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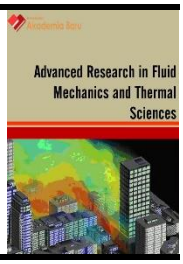

Extraction and Characterisation of Cellulose Nanocrystals Structures from Waste Office Paper

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ABSTRACT

The increase of consumption of paper has led to environmental issues due to the landfill and incineration activities. Subsequently, one of the major cellulosic waste materials includes the waste office paper (WOP). This wastage undergoes a value-added process, which is by having it converted into cellulose nanocrystals (CNC) that is prepared by an environmentally friendly and cost-effective method. Hence, this study is performed to synthesise CNC from WOP by acid hydrolysis method with the optimum acid concentration chosen based on the selected parameters. Until present time, limited research has been carried out on the preparation of CNC from WOP. Furthermore, the process of preparing CNC from WOP is easier and it is more energy and time efficient due to the minimum amount of lignin, compared to other cellulosic wastes. The alkali pretreatment and acid hydrolysis methods were conducted before further analysis by using X-ray Diffraction (XRD) in order to establish the crystallinity index (CrI) and Fourier-transform infrared (FTIR) spectroscopy, which show the confirmation of non-cellulosic materials that has been removed by alkali treatment with the 5 wt % concentration. In addition, via the Scanning Electron Microscopy (SEM), it was found that the surface morphology of the alkali-treated WOP was smoother than the untreated ones. From the result obtained, the optimum acid concentration is 30 wt %, with 90 minutes of hydrolysis time and 45°C temperature with the highest CrI and crystallite size of 36.35% and 32.92 nm, respectively. The extraction of CNC from WOP provides possible application as reinforcement in the nanocomposite industry. Thus, the use of WOP can be considered as green material, where it aligns with the sustainable design and development.

Keywords:
Cellulose nanocrystals; wastepaper; acid hydrolysis

1. Introduction

As other lignocellulosic raw materials, wastepaper has attracted attention due to its useful contribution in many celluloses and its derivatives application areas, including in papermaking [1-2], textile, composite [3-4], radiator [5], solar cells [6], biocomposting [7], road construction [8] and

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pharmaceutical. The large use of various types of papers, such as packaging paper, advertising paper, newspapers, and waste office papers, in everyday life reflects that paper is an important material in the world and it has significant effects to human. With the advent of new computer technology and the growing popularity of personal computers, there has been an increase in paper usage regarding all types of printers, including laser printers [9].

Paper use worldwide over the past four decades has increased by 400%, with the United States becoming the largest paper consumer. The same source revealed that over the last two decades, the country's consumption has grown about 126% from 92 million tons, representing only 5% of the world's population [10]. In Malaysia, about 1.9 million tons of papers and paperboards were consumed in 2017, which shows a significant increase in the figures, as compared to 1.6 million tons of similar items in the previous year [11]. In Malaysia, the three categories of recyclables are the paper, plastic and bottle; however, the recycling rate is very low. In Kuala Lumpur, for example, the current recycling rate is at 4.5% of the wastes generated. In vision, the rate is to increase to 16% by 2005 and 20% by 2020, in which the plan remains effective to date [12-16].

Wastepaper, being a cellulose biomass, gives a potential wellspring of crude material for the creation of cellulose nanocrystals (CNC). Previous study had found that wastepaper constitutes up to 70–100% of organic parts, which consist of cellulose, hemicellulose, lignin, and numerous compounds of lignin. The obtained cellulose is reported to have very special properties, such as crystal arrays, high level of whiteness and viscosity [17]. Cellulose, due to its structure, is a compound that has high potential to be used as a nanomaterial, in view of its abundance and its fibrillar nanostructure. This polysaccharide has a very special feature that makes it an extraordinary material, such as having high rigidity and mechanical properties, and is low in cost and biodegradable [18].

There are four main classes of cellulose nanomaterials that differ in terms of their morphology, particle size, crystallinity, and some other properties due to the different sources and extraction method. The main classes of cellulose are cellulose nanocrystals (CNC), cellulose nanofibrils (CNF), bacterial cellulose (BC), and electrospun cellulose nanofibres (ECNFs) [19]. However, the CNC of natural fibre has been widely used due to the advantages of its high crystallinity that can be acquired via a chemical approach, for instance, acid hydrolysis.

Meanwhile, the major element in lignocellulosic biomass that is mostly localised in the plant cell walls is about 35–50% cellulose. Cellulose can not only be found in plant cells, such as hemp, cotton, flax, jute, and wood, but also in agricultural residues, such as sugarcane bagasse, tunicate, algae, and sea animals that consist of protein and carbohydrate. The common extraction method in obtaining the nanocellulose is by chemical or mechanical treatment, or by both treatments combined [20]. However, the utilisation of waste office paper (WOP) as the starting material for CNC has not been much exploited. Therefore, this opens the opportunity to maximise the renewable and recyclable sources of CNC, instead of by using other common lignocellulosic fibres, such as sisal, kenaf, hemp, flax, grass, and many more.

However, acid hydrolysis is a famous method used in producing a stable colloidal suspension of the CNC [21]. In this method, the acid helps in degrading the amorphous region of the hemicellulose and lignin; thus, the crystal cellulose becomes the domain structure. Essential parameters, such as temperature correlation, hydrolysis time and acid concentration, must be considered in the acid hydrolysis, as it could affect the result of CNC obtained. In order to make the cellulose more accessible and the hydrolysis more effective, chemical pretreatment is always done prior to the hydrolysis process. The most common pretreatment used is the alkali pretreatment, in which the cellulose fibre is subjected into base solutions [22].

This study aims to prepare and characterise the CNC derived from WOP by acid hydrolysis method with the optimum conditions that consider the temperature correlation, hydrolysis time and acid

concentration. This is due to the lack of previous study on WOP that includes any wastepaper, either printed or non-printed paper, as the raw material for the extraction of CNC. The CNC obtained will be further analysed and characterised by using selected tools, such as X-ray Diffraction (XRD) analysis, Fourier-transform infrared (FTIR) spectroscopy and Scanning Electron Microscopy (SEM). Therefore, a definite CNC structure and crystallinity can be evaluated due to the optimum condition of hydrolysis focusing on the effect of hydrolysis time and concentration.

2. Methodology

2.1 Sample Preparation

The printed and non-printed waste office papers (WOPs) obtained from the UTeM campus area were used as a raw material in this experiment. Approximately 2 kg of printed and non-printed WOP was prepared. Firstly, the WOP was cut into smaller pieces of about 2–4 cm and was then washed four to five times with hot water. After that, the sample of the WOP was kept at room temperature for 24 hours. Next, the smaller pieces sample was ground down by applying the high-speed blender at soft to medium-hard to ground the brittle and fibrous sample rapidly and efficiently.

2.2 Pre-treatment by Alkali Treatment Method

The sample of the WOP was treated with the 5 wt % concentration of sodium hydroxide (NaOH) at room temperature for 24 hours in order to eliminate the inks and fillers and break the hydrogen bonding between the various cellulose chains. After that, the mixture was filtered and rinsed numerous times with distilled water until the pH becomes neutral. Then, the resulting mixture was dried via the drying oven for 3 hours at a temperature of 100°C.

2.3 Preparation of Cellulose Nanocrystals (CNC) by Acid Hydrolysis

The acid hydrolysis was conducted using different concentrations, which are 20 wt %, 30 wt %, 40 wt %, and 64 wt % of sulphuric acid (H_2SO_4). Meanwhile, the hydrolysis times were 30 min, 60 min and 90 min. The temperature correlation was fixed at 45°C and the ratio of the cellulose-to-acid solution was fixed to 1:14 (w/v). Therefore, the parameter was designed to study if the concentration will affect the hydrolysis time. The alkali-treated WOP was mixed with sulphuric acid and the experiment was performed by using different hydrolysis times and concentrations. The mixture was stirred continuously until the hydrolysis time was completed. The cellulose that has been hydrolysed was dialysed several times with distilled water until the pH is 7. After that, the sample was dried in the oven for 3 days at a temperature 40°C. The CNC acquired will be kept at room temperature for it to be analysed.

2.4 Scanning Electron Microscopy (SEM)

Scanning Electron Microscopy (SEM) is a common technique used to study the microscopic arrangement by scanning the ruptured surface of materials. In this study, Scanning Electron Microscopy (SEM) model Zeiss EVO 50 was used to study the effectiveness of the alkali treatment of sodium hydroxide (NaOH) that was evaluated through surface morphology and structure of the CNC-WOP. Thus, the surface morphologies of the raw and alkali-treated WOP were observed. A drop of diluted fibrils (raw WOP and treated NaOH) was placed onto the specimen holder. The samples were then dried, and a thin layer of the samples was put on the aluminium studs and layered with gold

using a mini sputter coater before the further analysis. The accelerating voltage was 2 kV with a resolution of up to 10 nm, while the magnification out of 5 to 1 000 000X was used to capture the samples' appearance.

2.5 FTIR Analysis

The elemental chemical compositions for the different chemical stages in WOP were determined by FTIR analysis with the use of FT-IR6100 Spectrum GX-FTIR spectrophotometer (Perkin Elmer, Germany) machine. This analysis was carried out at room temperature, and the three samples that were examined from different stages of the chemical treatment of WOP are: (i) raw WOP; (ii) alkali-treated WOP; and (iii) acid hydrolysed WOP, which has the highest crystallinity index (CrI). The samples were well combined with the potassium bromide (KBr) in order to prepare the homogenous suspensions and the samples were later pushed into the transparent pallets. The transmittance mode was examined at a resolution of 4 cm^{-1}, within the range of 500–4000 cm^{-1}.

2.6 XRD Analysis

In order to evaluate the crystallinity index (CrI) and crystallite size of cellulose extracted from the WOP, the crystal structure of CNC-WOP was analysed using the PAN analytical X'PERT PRO MPD X-ray Diffraction (XRD) analysis. The monochromatic CuKα radiation source of λ = 0.154060 at the scanning rate of 0.5°/min was used to perform the experiment at room temperature. The CrI of the three WOP samples are: (i) raw WOP; (ii) alkali-treated WOP; and (iii) acid hydrolysed WOP, which can be measured according to the crystal peaks shown by the XRD data analysis. The crystal structure and crystallite size were investigated in the 2Θ range from 10–40°.

3. Results

3.1 Alkali Treatment of Waste Office Paper (WOP)

Alkali treatment is one of the stages done to synthesise the CNC from the WOP. The main purpose of alkali treatment in synthesising the CNC is to remove the hemicellulose and other impurities in the WOP to attain high purification of the cellulose fibre. According to Arzu *et al.,* [23], the common method to significantly eliminate the hemicellulose and further impurities from the outer surface of the fibre cell wall, either before or after the bleaching treatment, is by alkali treatment. The alkali concentration, extraction time and temperature during the cellulose extraction were found to have affected the α-cellulose content, whiteness index and cellulose yield. Furthermore, the CNC structure behaviour for the swelling and shrinkage of the fibre, the rate of agglomeration and the disruption of the tensile strength were also affected by the alkali treatment [24].

3.2 Scanning Electron Microscopy (SEM) Analysis

The results of the surface morphology for the untreated and alkali-treated WOP were studied after completing the process of purification by alkali treatment. Based on the observation in Figure 1, the surface morphology of the untreated WOP was covered by many impurities, such as hemicellulose, lignin and other non-cellulosic materials that are undesirable for any reinforcement. Meanwhile, the surface morphology of the alkali-treated WOP was smoother than that of the untreated WOP. This indicated that the impurities had been successfully removed by the alkali treatment.

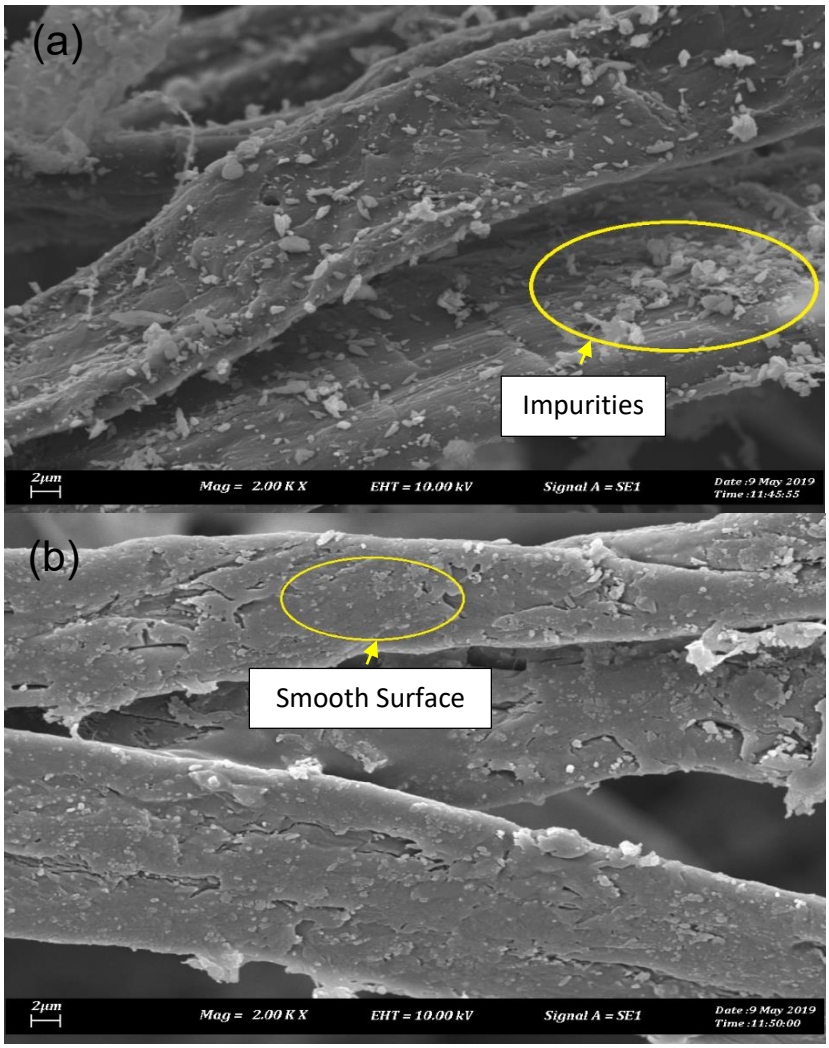

Fig. 1. SEM micrograph for: (a) untreated WOP and (b) alkali-treated WOP under the magnification of 2000X

The surface morphology of the WOP was analysed in order to characterise the structural change that had occurred during alkali treatment. Compared to the alkali-treated WOP, the surface of the untreated WOP was not smooth, as there was a presence of white spots on the surface that indicated impurities. From observation, it was found that there were impurities, such as hemicelluloses, lignin, waxes, and ink contaminants, in the untreated WOP. Indication of impurities or localised damage of the fibres by the white spots on the surface of the WOP fibre may be due to the handling process. Hence, based on the observation, the impurities have been successfully removed by the alkali treatment. Consequently, this observation is in accordance with the current research work by Hashim *et al.,* [25].

3.3 Acid Hydrolysis of Waste Office Paper

One of the main processes to isolate the CNC from cellulosic materials is by acid hydrolysis method. According to a study by Phantong *et al.,* [21], acid hydrolysis can easily hydrolyse the disordered region in cellulose chains, thus leaving the remaining part known as ordered region. This method also leads to the removal of amorphous part of raw materials for an extraction of CNC with

a high degree of crystallinity. In order to obtain a highly purified CNC, the hemicellulose and lignin must be completely removed.

3.4 X-ray Diffraction (XRD) Analysis

The patterns of the XRD analysis were analysed to study the influence of the constraints used after undergoing the acid hydrolysis treatment by sulphuric acid (H_2SO_4). The XRD analysis for the untreated WOP, alkali-treated WOP and CNC of the WOP obtained at optimum condition was showed in Figure 2. Based on the diffraction patterns, there were three well-defined peaks of cellulose I. This peak is present at around 2Θ = 15°, 22° and 34°. From this analysis, the crystallinity index (CrI) and crystallite size were calculated using Segal Equation and Scherer Equation, respectively, and the results are shown in Table 1 and 2.

Based on Figure 2, it can be concluded that the crystallinity peak increases upon the purification by alkali treatment and after undergoing the acid hydrolysis treatment. This was due to the alkali treatment that had successfully removed partially of the non-cellulosic materials in order to obtain the highly purified nanocellulose. This assumption was supported by Phantong *et al.,* [21], which stated that the application of alkali could be used in alkali treatment as an agent in eliminating the amorphous polymer of hemicellulose and the residual lignin.

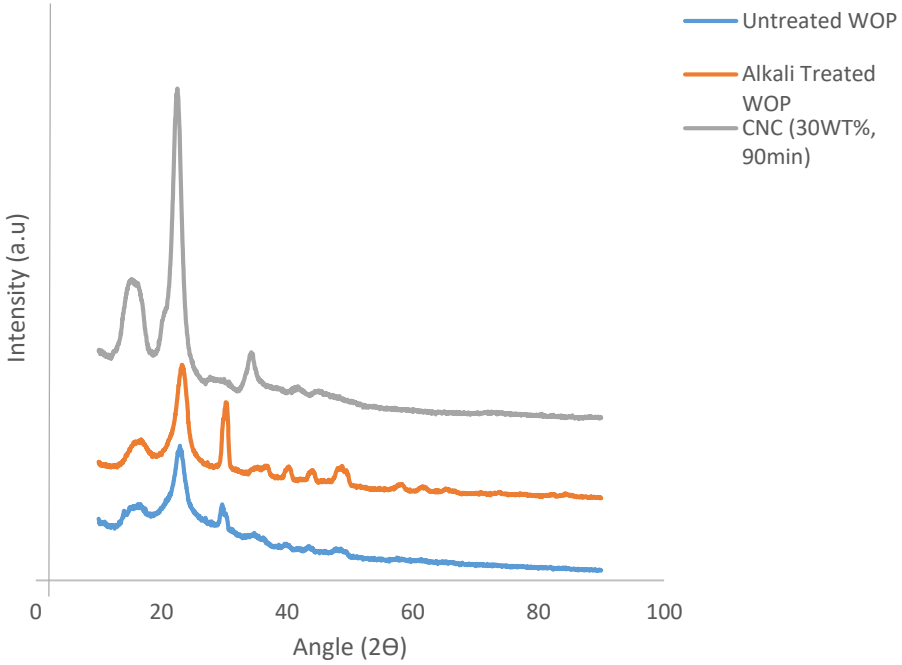

Fig. 2. X-ray Diffraction patterns of CNC WOP using different treatments

Meanwhile, the CrI and crystallite size were calculated, as shown in Table 1 and 2. The percentage of CrI increases upon the purification by alkali treatment and after undergoing the acid hydrolysis treatment. The increasing CrI might be caused by the elimination of amorphous cellulose throughout the acid hydrolysis process. It was found that the concentration of acid used, and the hydrolysis time had affected the process of acid hydrolysis and the higher concentration and reaction time used had resulted in increasing CrI. This explanation is validated by Phantong *et al.,* [21], which claimed that the reaction time, temperature and acid concentration were the key controlling aspects that had affected the properties of the nanocellulose acquired.

Table 1
Cellulose intensity peak and their respective CrI

Sample	Cellulose Intensity Peak (2Θ)	Crystallinity Index (%)
Untreated WOP	22.1448	24.8027
Alkali-treated WOP	22.3983	25.0336
CNC (20 wt %, 30 min)	23.0934	27.2593
CNC (20 wt %, 60 min)	22.8872	29.1696
CNC (20 wt %, 90 min)	22.5968	33.2459
CNC (30 wt %, 30 min)	23.0619	34.7495
CNC (30 wt %, 60 min)	22.5548	34.7522
CNC (30 wt %, 90 min)	22.6093	36.3501
CNC (40 wt %, 30 min)	-	-
CNC (40 wt %, 60 min)	-	-
CNC (64 wt %, 60 min)	-	-

Table 2
Crystallite size of CNC at different parameters

Sample	Position (°2Th.)	FHWM (°2Th.)	Crystallite Size (nm)
CNC (20wt %, 30 min)	23.0934	0.8400	9.6528
CNC (20wt %, 60 min)	22.8872	2.2380	3.6217
CNC (20wt %, 90 min)	22.5968	0.7380	10.9774
CNC (30wt %, 30 min)	23.0619	0.6396	12.6765
CNC (30wt %, 60 min)	22.5548	0.4428	18.2959
CNC (30wt %, 90 min)	22.6093	0.2460	32.9296

However, when the concentration reached 40 wt %, the sample of WOP was dissolved in the solution and the acid concentration became more than 60 wt %, causing the sample to turn black and dissolve with the acid. Consequently, characterisation on the sample of WOP with concentrations of 40 wt % and 64 wt % could not be conducted for further analysis. This might be due to the side reactions, such as dehydration and oxidation, which had occurred when the hydrolysis became black, as supported by Mikaela and Gunnar [26] who have mentioned that the solution turns black and dissolves in acid due to the solution and organic substances that reacted with one another. Besides that, one of the quite extreme oxidising agents is the sulphuric acid, which can dehydrate many organic compounds to produce carbon in the form of carbon and graphite.

Based on the crystallite size calculation shown in Table 2, it can be observed that the CNC extracted using 30 wt % of acid concentration for 90 min had the best crystallite size and a higher crystallinity index. The CNC was produced, as proven by the calculation done on the crystallite size, having a value of less than 100 nm. It can be concluded that the crystallite size was related to the concentration and hydrolysis time used. The higher crystallinity index had also affected the crystallite size, as the value of the crystallite size had increased upon the concentration used. According to French *et al.*, [27], the relationship between the crystallinity index calculated based on the Segal Equation was linear to the relative crystallite size. Therefore, in overall, the optimum conditions for the isolation of CNC are at 30 wt % concentration and 90 minutes hydrolysis time with the temperature of 45°C.

3.5 Fourier-transform Infrared (FTIR) Spectroscopy Analysis

The FTIR spectra of the untreated WOP, alkali-treated WOP and CNC of WOP with optimum conditions are shown in Figure 3. Based on the observation, CNC was produced in the form of type I structure and it was showed by the transmittance signal at 1428 cm^{-1}.

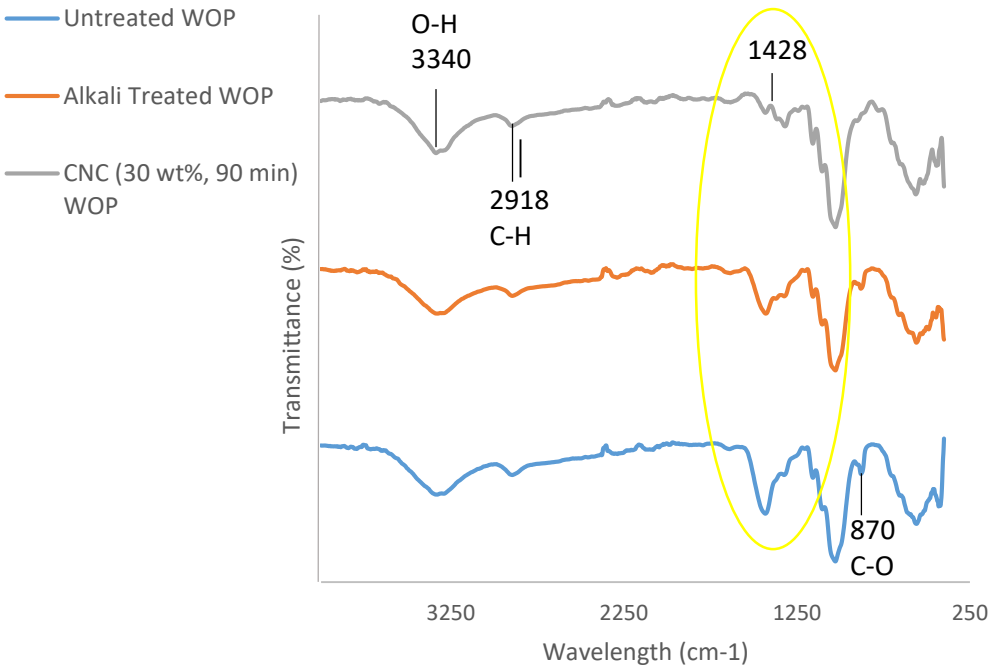

Fig. 3. FTIR spectra of untreated WOP, alkali-treated WOP and CNC of WOP

The peak at 870 cm^{-1} refers to the calcite mode, which is associated to the C-O stretching mode. It was observed that two bands at 870 cm^{-1} and 712 cm^{-1} had corresponded to the symmetric C-O stretching mode and O-C-O bending mode of the calcite, respectively, and calcium carbonate was produced [22]. However, the intensity of the band at 870 cm^{-1} lowered upon the purification by alkaline treatment and the intensity was absent after undergoing the acid hydrolysis treatment. This resulted in the extraction of highly purified nanocellulose, as the calcite was successfully removed by the treatment.

Both the untreated and alkali-treated WOP had peaks at wavelengths of 3340 and 2918 cm^{-1} due to the presence of hydroxyl group and aliphatic saturated C-H stretching vibration of the cellulose, respectively. Besides that, the presence of a broader peak that complemented the O-H stretching vibration at peak 3340 cm^{-1} showed that there was an increase in water absorption upon the treatment that resulted in the removal of amorphous cellulose [20]. Hence, it can be concluded that the difference in the FTIR spectra between the untreated WOP, alkali-treated WOP and CNC of WOP extracted had indicated success in the isolation of CNC from the WOP by acid hydrolysis method.

4. Conclusion

As a conclusion, the resultant cellulose nanocrystals (CNC) from waste office paper (WOP) has been successfully prepared based on the selected parameters with optimum acid concentration. It was found that the optimum acid concentration to isolate the CNC from WOP was at 30 wt % concentration with the hydrolysis time of 90 min and a temperature of 45°C. The CNC obtained was prepared in two stages of treatment, that is by alkali treatment using sodium hydroxide (NaOH) with the concentration of 5 wt % before it is subjected to the acid hydrolysis method using sulphuric acid (H_2SO_4) at selected parameters. The parameters selected were the acid concentration, hydrolysis time and fixed temperature correlation to the 45°C. The CNC obtained from the WOP provides other alternatives of the cellulosic source.

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