Extraction of nanocrystalline cellulose from Kapok fiber as potential water-resistant composite.

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Abstract. This study focused on synthesizing of nanocrystalline cellulose (NCC) from kapok fiber with the combination of chemical and mechanical methods. The as-synthesized NCC is used to produce nanocrystalline cellulose (NCC) paper. It was determined that the optimum condition to synthesis nanocrystalline cellulose (NCC) from kapok fiber using acid hydrolysis process treatment was at 45˚C, 2 hours and 50% concentration of H₂SO₄. The particle size of the synthesized nanocrystalline cellulose (NCC) was 262.9 - 455.6 nm. The disappearance of peaks 1731.89, 1594.68, 1505.51 & 1240 cm⁻¹ in the (FTIR) spectroscopy was related to the removal of non-cellulose component such as lignin and hemicellulose. Based on the contact angle, the value gained for that contact angle of the filter paper was increased from 29.650˚ to 40.261˚ and 32.002˚ due to presence of nanocrystalline cellulose (NCC) in the filter paper. It was determined that the modified filter paper with the nanocrystalline cellulose (NCC) had the highest water-resistances property and was considered the most hydrophobic of all the other papers.

1. Introduction

Natural fibers, also known as cellulose-based fibers, is a term that indicates various fibers that are naturally present in plants, animals, and minerals [1]. On a worldwide scale, natural fibers are considered as one of the environmental friendly components for the formation of composite materials, which have excellent physical, mechanical and chemical properties [1]. Lignocellulosic biomass includes different types of natural organic matters which mostly refer to the plants or plant-based materials. As shown in Figure 1, microfibrils is mainly consists of lignin, hemicellulose, and cellulose in the cell wall structure [2].
Cellulose plays an important role in producing paper and paper packaging material. Cellulose which converted to nanocellulose is having bigger surface area and this eventually will improve the properties of the composite or packaging material. Active packaging is known for the use of nanoparticles. Nanoparticles such as nanocellulose are incorporated into polymer films for the prevention of microbial surface contamination of foods. Azeredo et. al., (2017) mentions that nanocellulose matrices may be applied as coatings to conventional polymers. One of the benefits of incorporation of nanomaterials in food packaging is that it will help in food waste reduction by retaining the freshness of packaged food. The packaging industry presently uses mainly unsustainable coatings of wax, plastics, or aluminum, and many other materials in order to manufacture competitive packages based on paper [4].The main function of the packaging is to protect from the three major classes of external influences that are chemical, biological, and physical influences. In the paper and paperboard making industry, nanocellulose acts as an additive and a strengthening agent. This will make the paper denser and stronger due to the greater surface area of nanocellulose. It also provides smooth surfaces to printing papers, improving printing quality. Paper and paperboard which are used as packaging purposes are sheet materials made from an interlaced network. The interlaced network consists of cellulose fibers derived from wood by using sulfate and sulfite. This project aims to extract nanocellulose from Kapok fiber using alkaline hydrolysis pretreatment for nanocomposite application.

2. Methodology
This research was conducted in UCSI University laboratory, Kuala Lumpur. Kapok fiber was purchased from a retail store in Wakaf Bharu, Kelantan. The consumable used in this experiment such as chloroform (Trichloromethane), acetic acid (ethanoic acid), sodium hypochlorite (NaClO), sodium hydroxide (NaOH) and sulphuric acid (H₂SO₄) were analytical grade and used per receive without any further purification.

2.1. Pre-treatment Process
For the extraction of cellulose from the kapok fiber, two different pre-treatment processes were conducted. The processes were labelled as Process 1 and Process 2. Both pre-treatment processes used were similar chemical methods which included of alkaline hydrolysis and bleaching treatment, however, process 2 was conducted with an extra treatment procedure, which was the chloroform treatment.

2.1.1. Process 1 (without chloroform treatment). For Process I, 5g kapok fiber was dried in the oven at 100 °C for 24 hours. Then, alkali process treatment was carried out by adding the dried kapok to a 1 L beaker containing 10 % (w/v) of sodium hydroxide solution and the process was conducted for 3 hours at 80°C with constant stirring. 500 ml of 10 % (w/v) of sodium hydroxide solution was used for the alkali pre-treatment process, which followed according to the mass to volume ratio in which for every 1 gram of kapok fiber used, 100 ml of 10 % (w/v) of sodium hydroxide solution was added. Followed by, the residue fiber is then added to another 1 L beaker containing 2% (v/v) of sodium hypochlorite solution for 2 hours at 80°C with constant stirring. The amount of 2% (v/v) of sodium hypochlorite solution was used for the bleaching pre-treatment process was according to the mass to volume ratio in which for every 1 gram of alkali treated fiber used; 100 ml of 2% (v/v) of sodium hypochlorite solution was added.
2.1.2. Process 2 (with chloroform treatment). For Process 2, 5g kapok fiber was dried in the oven at 100 °C for 24 hours. Then the dried kapok was added to a 1 L beaker containing chloroform and was conducted for 1 hour at 45°C. 500 ml of chloroform was used for the chloroform pre-treatment process which followed according to the mass to volume ratio in which for every 1 gram of kapok fiber used, 100 ml of chloroform was added. The treated fiber was placed into 1 L beaker containing 10 % (w/v) of sodium hydroxide solution was conducted for 3 hours at 80°C with constant stirring. The amount of 10 % (w/v) of sodium hydroxide solution was used for the alkali pre-treatment process was according to the mass to volume ratio in which for every 1 gram of chloroform treated kapok fiber used; 100 ml of 10 % (w/v) of sodium hydroxide solution was added. The residue fiber is then added to a beaker containing 2% (v/v) of sodium hypochlorite solution was conducted for 2 hours at 80˚C with constant stirring. The amount of 2% (v/v) of sodium hypochlorite solution was used for the bleaching pre-treatment process was according to the mass to volume ratio in which for every 1 gram of alkali treated fiber used; 100 ml of 2% (v/v) of sodium hypochlorite solution was added.

2.2. Synthesizing the Nanocrystalline Cellulose (NCC)
After the pre-treatment process, the remaining cellulose residue was added to a 1 L beaker containing 5% (v/v) of sulphuric acid solution, was conducted for 1-2 hours at 45 & 90 °C with constant stirring. The amount of 5% (v/v) of sulphuric acid solution was used for the acid hydrolysis treatment process was according to the mass to volume ratio in which for every 1 gram of cellulose fiber used, 100 ml of 5% (v/v) of sulphuric acid solution was added. Then cold distilled water at 10 °C was poured into the beaker. The amount of cold distillate water was poured was according to the volume to volume ratio in which for every 1 ml of 5% (v/v) of sulphuric acid solution, 10 ml of cold distilled water was required. Then the diluted solution was centrifuged using a centrifugation equipment, Sigma 2-6 Including Imbalance Sensor-10223 model, at 3000 rpm for 12 minutes and was repeated for 2 times. The residue was ultrasonicated using an ultrasonicator, Hielscher Ultrasonic Sound Protection Box model, at 100% amplitude for 10 minutes with constant stirring. The experiment was repeated using 20, 30, 40, 50 and 60% (v/v) of sulphuric acid solution.

2.3. Crosslinking and Curing Process
After the pre-treatment and acid hydrolysis process, the nanocrystalline cellulose (NCC) was mixed in 25 (v/v) % acetic acid solutions and stirred for 30 minutes at 45°C The amount of 25 (v/v) % acetic acid solutions solution was used for the crosslinking process was according to the mass to volume ratio in which for every 1 gram nanocrystalline cellulose (NCC) used, 100 ml of 25 (v/v) % acetic acid solutions was added. The solution was placed in the oven at 100 °C for 15 minutes for the curing process. Thus, the nanocrystalline cellulose (NCC) which underwent crosslinking and curing process was known as treated nanocrystalline cellulose (NCC) in this experiment.

2.4. Synthesizing the Nanocomposite Paper
A filter paper was teared into small pieces and added to the blender. The blender was used to further shred the treaded pieces of filter paper. Then, 50 ml of distilled water was poured into the blender. The slurry containing the mixture of shredded filter paper and distilled water was filtered out using a strainer. The remaining pulp in the strainer was added into a 100 ml beaker. 20 ml of distilled water was poured into the 100 ml beaker containing the pulp. The nanocrystalline cellulose (NCC) synthesized from the optimized acid hydrolysis process was also added into the 100 ml beaker containing the filtered pulp and was mixed for 10 minutes using a magnetic stirrer. Finally, the mixed pulp was filtered using the strainer and was placed in the oven to dry. The drying process of the pulp was carried out weighted until a constant weight of the pulp. This step was repeated using the treated nanocrystalline cellulose (NCC) synthesized from the crosslinking and curing process.
3. Results and Discussion

In this chapter, the results obtained through observation of the samples and using the characterization equipment was discussed. The description of the results was explained in this chapter with prove of figures and tables. The characterization equipment used was Nicolet iS5 FTIR Spectrometer with a wavelength range of 200 – 4000 cm\(^{-1}\), the Malvern Zetasizer Nanosizer based on dynamic light scattering (DLS) method, which has a measurement range: 3.8 nm – 100 microns (diameter), and Optical Tensiometer (SI-CAM2000Y).

3.1. Functional Group Analysis of Kapok Fiber, Process 1 and Process 2

Figure 2(a) shows the FTIR spectra for the raw kapok fiber, which was used as a base reference to determine the functional groups that were presented in the raw fiber. Figure 2(b) shows the FTIR spectra of the nanocrystalline cellulose (NCC) obtained through Process 1, and Figure 2(c) shows the FTIR spectra of the nanocrystalline cellulose (NCC) obtained through Process 2.

![Figure 2(a)](image)

![Figure 2(b)](image)
Figure 2. FTIR spectra of (a) Raw Kapok Sample (b) Kapok Sample from Process 1 (c) Kapok Sample from Process 2

The absorption band which corresponds to the hemicellulose was observed at 1728–1736 cm\(^{-1}\) [5]. In this experiment, the absorption band which corresponded to hemicellulose was observed in the raw kapok fiber at 1731.89, which satisfies in the range given by Mohamed et al., (2017). Lignin present at the peaks in the range of 1,500–1,600 cm\(^{-1}\) corresponding to the aromatic skeletal vibration [6]. From Figure 2(a), the absorption band at 1594.68 cm\(^{-1}\) was attributed to the C=C unsaturated linkages, the aromatic rings present in lignin; while 1505.51 cm\(^{-1}\) corresponded to the C=C stretching vibration in the aromatic structure of lignin. Both peak points satisfy the range given by Morán et al., (2008). The absorption band at 1240 cm\(^{-1}\), which could be attributed to hemicellulose and lignin, was further observed in the FTIR spectra. The peak statement of 1240 cm\(^{-1}\) was agreeable with the results obtained by Wulandri et al., (2016). Based on Figure 2(b) and 2(c), the absorption band at 1731.89, 1594.68, 1505.51 & 1240 cm\(^{-1}\) were completely disappeared after undergoing both process treatments, indicating that the hemicellulose and lignin were successfully removed from the fibers. Both figures showed the presence of the vibration of C-O-C in the pyranose ring indicated by the absorption peak at 1047.49 and 1050.99 cm\(^{-1}\). The peak statement of 1047.49 and 1050.99 cm\(^{-1}\) were agreeable with the results obtained by Morán et al., (2008) and Wulandari et al., (2016). The bending modes of the absorbed water were shown at 1633.80 and 1634.01 cm\(^{-1}\). The bands at 1639–1641 cm\(^{-1}\) were due to the carboxylate group, as well as, the bending mode of the absorbed water in the samples [5]. Finally, the O-H stretching vibrations of the O-H group were shown as peak 3347.18 and 3332.83 cm\(^{-1}\). The peak of the O-H group satisfies the range stated by Morán et al., (2008), which was 4,000–2,995 cm\(^{-1}\).

Even though both samples have the sample functional group, sample obtained form Process 1 showed narrower peaks compared to the sample obtained from Process 2. Process 1 also consists of fewer steps compare to Process 2, thus reduces the time consumption that was required to conduct the experiment. Not only that, but the kapok residue after the alkali process obtained from Process 2 was dark orange in color compare to the normal light brownish color obtained by sample using Process 1 as shown in Figure 3. Johar et al., (2012), stated that these color changes are due to the removal of non-cellulosic materials and other impurities such as lignin, hemicelluloses, pectin, and wax upon chemical treatment. Due to the dark orange color change in Process 2 which may have indicated the presence of impurities mixed with the kapok residue during the chloroform treatment process. The cellulose residue from Process 2 formed nanocrystalline cellulose (NCC) sample which was orange in color while the cellulose residue from Process 1 formed nanocrystalline cellulose (NCC) sample which was white in color. Thus, Process 1 was considered the most optimal process treatment and procedure that was used to synthesized the nanocrystalline cellulose (NCC) sample.
3.2. Effect of acid hydrolysis process on the synthetization of nanocrystalline cellulose (NCC)

The effect of each concentration was presented in Table 1. Based on physical observation of the nanocrystalline cellulose (NCC) color obtained, the nanocrystalline cellulose (NCC) produce at acid hydrolysis concentration of 30, 40 and 50 % of sulphuric acid was considered the most suitable concentration used in this research experiment. Samples obtained from 30, 40 and 50 % were white in color, which indicates that the nanocrystalline cellulose (NCC) was able to maintain the cellulose structure. White fibers had the highest cellulose content [9]. Based on physical observation, it was seen that the particle size was smaller as the concentration was increased from 30 to 50 %. The average particle size was strongly reduced as a consequence of greater cellulose depolymerization produced by the acid [10]. The temperature used for samples with a concentration of 5 and 20 % of sulphuric acid was at 90 °C, while the other samples were at 45 °C. Based on physical observation of the particle size and color, from the sample with a concentration of 5 and 20 % sulphuric acid, it was seen that there was a large decrease in particle size compare to the other samples, however, the color of the sample was not white in color, the particle was dark brown or blackish in color which indicated that the nanocrystalline cellulose (NCC) was degraded. All the samples had undergone the same conditions of 1 to 2 hours of reaction time. It was observed that the physical results obtained were the same for all the samples. At 1 hour of reaction time, only small parts of the celluloses were broken down, the majority of the celluloses were intact. However, at 2 hours it was seen the celluloses were broken down and reduce in size. This statement was agreeable with Sun et al., (2016), who mentioned that the particle size of the cellulose exhibited a continuous decrease as the acid hydrolysis process continued. Based on the optimization that was conducted for this research experiment, the optimal condition to obtain the nanocrystalline cellulose (NCC) was 50 % of sulphuric acid, 45 °C and 2 hours.

Table 1. Observation results of the samples

| No | Parameters | Results |
|----|------------|---------|
| 1  | Concentration (%) | 5 |
|    | Temperature (°C) | 45 & 90 |
|    | Time (hr) | 1-2 |
| 2  | Concentration (%) | 20 |
|    | Temperature (°C) | 45 & 90 |

- The cellulose fiber particle size was reduced after 2 hours. The cellulose fiber reminded white in color at 45 °C but turn dark brown in color (degradation of nanocrystalline cellulose (NCC)) at 90 °C.
3.3. Particle Size Analysis

Particle size of the sample produced had been analysed using Dynamic Light Scattering (DLS) technique. The samples stated in Table 2, were the nanocrystalline cellulose (NCC) synthesized from both pre-treatment processes and the optimal condition for acid hydrolysis process treatment. The results obtained from the DLS analysis showed that the particle diameter of nanocrystalline cellulose (NCC) obtained using Process 1 has a lower average particle diameter compare to the sample obtained from Process 2 as shown in Table 2. Khan et al., (2019), mentions that nanoparticles have size ranges from few nanometer to 500 nm. Thus, Process 1 was considered the most effective method in producing nanocrystalline cellulose (NCC) with a diameter below 500 nm.

Table 2. Diameter of the nanocrystalline cellulose (NCC) particles obtained from Process 1 and 2

| Samples                   | Average Diameter of nanocrystalline cellulose (NCC) particles (nm) |
|---------------------------|---------------------------------------------------------------------|
| Sample obtained from Process 1 | 262.9 - 455.6                                                        |
| Sample obtained from Process 2 | 1669 – 1998                                                          |

3.4. Functional Group Analysis of Filter Paper and modified Filter Papers

Figure 4(a-c) shows the Fourier Transform Infrared (FTIR) spectroscopy characterization for the filter paper and the synthesized nanocomposite papers. Both Figure 4(a) and 4(b) showed peaks 3332.59 cm\(^{-1}\) and 3334.45 cm\(^{-1}\) which correspond to the O-H band for the filter paper, which satisfies the range mention by Wang et al., (2016). However, there was a slight change observed in Figure 4(b) at the peak of 895.03 cm\(^{-1}\), which showed a narrower peak compared to the FTIR spectra of Figure 4(a). This indicates that there was an increase in the cellulose concentration in the filter paper after mixing it with nanocrystalline cellulose (NCC). There were major changes to the composition of the filter paper that was observed based on Figure 4(a) and 4(c). FTIR spectra of the filter paper with treated nanocrystalline cellulose (NCC) showed a broader absorption peak at 3334.05 cm\(^{-1}\) which corresponds to the O-H stretching vibrations arising from abundant free hydroxyl and hydrogen-bonded hydroxyl groups, and the peak at 1639.73 cm\(^{-1}\) which relate to the O–H bending vibration of the adsorbed water showed a narrower band. When comparing the Figure 4(b) and 4(c), it was seen that the absorption at 1159.69 cm\(^{-1}\), 1028.91 cm\(^{-1}\), 1159.79 cm\(^{-1}\), and 1030.65 cm\(^{-1}\), related to anti-symmetrical bridge C–O
stretching and C–O–C stretching vibrations respectively, also become stronger, indicating glucose molecules are successfully cross-linked to the surface of filter papers.

Figure 4. FTIR spectra of (a) Filter Paper (b) Filter Paper with Nanocrystalline Cellulose (NCC) (c) Filter Paper with Treated Nanocrystalline Cellulose (NCC)
3.5. Hydrophobicity Analysis

Based on Table 3, it was determined that all the papers used in this experiment were considered having hydrophilic surface properties. However, it was observed that the modified filter paper with the nanocrystalline cellulose (NCC) was most hydrophobic compare to the other two papers used while the unmodified filter paper with the treated nanocrystalline cellulose (NCC) seemed to be the most hydrophilic. Nanocrystalline cellulose (NCC) was considered hydrophilic insoluble nanoparticles in water but poorly dispersed in nonpolar solvent, as well as a nonpolar polymer matrix for nanocomposite preparation [14]. Based on the statement given by Tan et al., (2019), it was considered that the nanocrystalline cellulose (NCC) and the treated nanocrystalline cellulose (NCC) that were used, were insoluble in water and may had increased the hydrophobicity of the filter paper surface. The result of the contact angle for both modified filter paper may be caused by the increased in the surface roughness of the filter paper [15]. The results of the contact angle obtained for the filter paper with the treated nanocrystalline cellulose (NCC) may be due to the crosslinking process by acetic acid thus altered the cellulose structure of the nanocrystalline cellulose (NCC) particle. Acetylation is the reaction that introduces an acetyl functional group CH3-C(=O)- onto the surface of cellulose and the reaction only occurs on the cellulose chains located on the surface of the nanoparticles [16].

Table 3. Contact Angle obtained for the filter paper and modified filter papers

| Type of paper used                                      | Contact Angle |
|--------------------------------------------------------|---------------|
| Filter paper                                           | 29.650˚       |
| Filter paper with nanocrystalline cellulose (NCC)       | 40.261˚       |
| Filter paper with treated nanocrystalline cellulose (NCC)| 32.002˚       |

3.6. Water Absorption Capacity

A surface is hydrophilic if it tends to absorb water or be wetted by water while a surface is hydrophobic if it tends not to absorb water or be wetted by water [16]. From the physical observation of the wettability of the papers, three papers were considered as hydrophilic. Based on Table 4, it was tabulated that the water absorbed by the filter paper with treated nanocrystalline cellulose (NCC) was slightly more than filter paper with nanocrystalline cellulose (NCC). This statement indicated that the modified filter paper with nanocrystalline cellulose (NCC) is more hydrophobic compared to the modified filter paper with treated nanocrystalline cellulose (NCC), which relates to the contact angle value obtained in Table 3. The value of water absorption of the unmodified filter paper was neglected due to the unmodified filter paper properties. The surface morphology of the unmodified filter paper seems to be fibrous and smooth, which indicates that filter paper was both hydrophilic and oleophilic [13]. However, the unmodified filter paper contained tiny pores which water passes through. This indicates that only allows a slight amount of water was absorbed, while the remaining was passed through the untreated filter paper. Not only that, the treated nanocrystalline cellulose (NCC) might have undergone a degree of acetyl substitution which had a strong influence on material properties like stiffness and deformability [16]. The modified filter paper with nanocrystalline cellulose (NCC) was considered to have the highest water-resistance compared to the unmodified filter paper and the modified filter paper with treated nanocrystalline cellulose (NCC).
Table 4. Water Absorption Capacity for the filter paper and modified filter papers

| Type of paper used                                      | Initial weight of the paper used | Final weight of the paper used with absorbed water | Weight of the water absorbed |
|---------------------------------------------------------|----------------------------------|---------------------------------------------------|-----------------------------|
| Filter paper                                            | 0.0945 g                         | 0.3841 g                                          | 0.2896 g                    |
| Filter paper with nanocrystalline cellulose (NCC)       | 0.0910 g                         | 0.5258 g                                          | 0.4348 g                    |
| Filter paper with treated nanocrystalline cellulose (NCC) | 0.0915 g                         | 0.5458 g                                          | 0.4543 g                    |

4. Conclusion

It was determined that Process 1 has the best conditions to produce the desired cellulose. The optimized conditions to synthesize nanocrystalline cellulose (NCC) using acid hydrolysis treatment were 50%, 45°C and 2 hours. Based on the FTIR spectra obtained for nanocrystalline cellulose (NCC), it was determined that the samples obtained throughout the time period of experimenting were considered nanocrystalline cellulose (NCC) that had non-cellulose components removed through the pre-treatment. The main objective that was stated was to determine the contact angle and water resistances property of the paper that consists of nanocrystalline cellulose (NCC) composition. Even though, the modified filter paper with the nanocrystalline cellulose (NCC) has a higher amount of water absorb compare to untreated filter paper, the value of water absorption of the unmodified filter paper was neglected due to the unmodified filter paper properties. It was determined that the modified filter paper with the nanocrystalline cellulose (NCC) had the highest contact angle value and was considered the most hydrophobic of all the other papers. In conclusion, the modified filter paper with the nanocrystalline cellulose (NCC) had the highest water-resistances property.

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