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RESEARCH ARTICLE

Effect of Nanocellulose Reinforced Recycled Paper Towards Tensile Strength

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ABSTRACT - The idea of paper recycling was done due to environmental issue. However, the properties of recycled paper like tensile will decrease each time it is recycled. Addition of reinforcing filler may increase the properties of recycled paper. Thus, in this research, nanocellulose was used as reinforcing filler in recycled paper. The objectives of this research are to fabricate papers from recycled paper and to investigate the effect of different nanocellulose and pressure of compression molding in paper fabrication towards tensile strength of recycled paper. Two types of nanocellulose used were commercialized cellulose nanofiber (CNF) and cellulose nanocrystals (CNC). CNC from filter paper and empty fruit bunches (EFB) were isolated via acid hydrolysis. The recycled paper was fabricated using traditional methods (net molding) and further processed with compression molding. All recycled paper with nanocellulose increases in tensile strength. The best nanocellulose for improving recycled paper is CNF. Pressure of compression molding also influences the enhancement of recycled paper strength. The tensile strength recorded 98% improvement at 15.28 MPa with 5wt% of CNF at 10 MPa of compression molding pressure.

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1.0 INTRODUCTION

Paper waste is one of the known problems nowadays and has affected our environment fatally. Paper is traditionally identified with reading and writing but packaging now accounts for over 41% of all global paper use. Estimates suggest that global paper consumption in 2025 will amount to 500 million tons, which is about 1.6% growth a year[1] . To pursue a better environment, recycling of paper waste is a strategy to reduce the effect to environment through remarkable greenhouse gas emissions reduction by saves in waste management practices[2]. Recycled paper is easy to crack up and tear apart, because of that the paper becomes low quality and hard to commercialize [3]. This is due to the recycling process itself which utilizes the aging and densification process, that resulted to the reduction on tensile strength and bonding of the recycled paper [4]. Recycled paper which has gone more refining process compared to fresh produced paper has a stiffening impact to the paper fiber that restraints it to obtain a strong interfibrous bonding. Hagel et al. reported that recycling process have a capability to reduce up to 30% of the original fibre strength due to various factors such as shortening of fibres, micro indentations during compression and flexibility loss [5]. Hence, this poses a problem to obtain high tensile strength recycled papers to increase its commercial utilization.

One of the methods to overcome this problem is by adding reinforcing filler like nanocellulose into the recycled paper. Reinforcing fillers serve a purpose beyond mere occupation of space within paper structure. Fillers affect paper's structure, appearance and many measurable properties that determine paper's end-use performance in different applications. It has played an especially prominent role in the manufacture of paper products intended for printing [6]. Nanocellulose like cellulose nanocrystals (CNC) and cellulose nanofiber (CNF) is one of the good reinforcing fillers to be added in recycled paper. Nanocellulose can be categorized by its size and structure. CNF has an average diameter around 20–50 nm. Meanwhile, the average range of CNC diameter and length are 5–70 nm and 100 nm, respectively [7]. Interesting properties of nanocellulose including low density, low thermal expansion coefficient, high strength, high stiffness, and easily modifiable surface make nanocellulose an ideal nanoscale building block for constructing macroscopic high-performance materials [8]. Previously, CNF from various sources like has proved the ability to improve the properties of recycled paper [9-12]. Meanwhile, the usage of CNC as a reinforcement element for recycled paper was not as much as CNF and the CNC used was modified and able to increase the mechanical properties [13]. However, comparison between these nanocelluloses have not been reported elsewhere. The objectives of this research are to fabricate recycled paper from office waste paper and to investigate the effect of CNCs and CNF as well as pressure of compression molding in paper fabrication towards tensile strength of recycled paper.

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2.0 MATERIALS AND METHODS

2.1 Materials

Recycled paper that has been used in this study is office waste paper (OWP). Recycled paper was collected around Universiti Malaysia Pahang. Hydrogen peroxide with concentration of 30% was used to bleach the paper and was bought by Sigma Aldrich. Filter paper was used for CNC extraction and acid sulfuric with concentration of 98% from Sigma Aldrich was used for acid hydrolysis. CNF derived from EFB was purchased from Universiti Putra Malaysia.

2.2 Isolation of CNC

CNC preparation was done by acid sulfuric hydrolysis and the raw materials were from filter paper and EFB. Filter paper was blended with deionized water with solid to liquid ratio 1:75. For the raw material of filter paper, sulfuric acid was added slowly to the solution until it reached a concentration of 32%. While for EFB, sulfuric acid was added until it reached a concentration of 64%. Ice bath was used to keep the temperature below 20 °C. Then, the mixture was heated to 50 °C for 3.5 hours. The cellulose suspension was centrifuged 5 times at 4800 rpm until it became turbid. The cellulose suspension was dialyzed (dialyzed tubing with MW cut-off of 14000) against deionized water until the suspension reached the neutral state (pH~7). After that, the cellulose suspension was ultrasonicated using high intensity ultrasonication (QSonica ultrasonicator) for 30 minutes at an output of 500W, frequency of 20 kHz and 20 amplitudes. Finally, the cellulose suspension was freeze dried. Nanocellulose obtained from filter paper and EFB were donated as CNC-fp and CNC-efb respectively.

2.3 Paper Fabrication

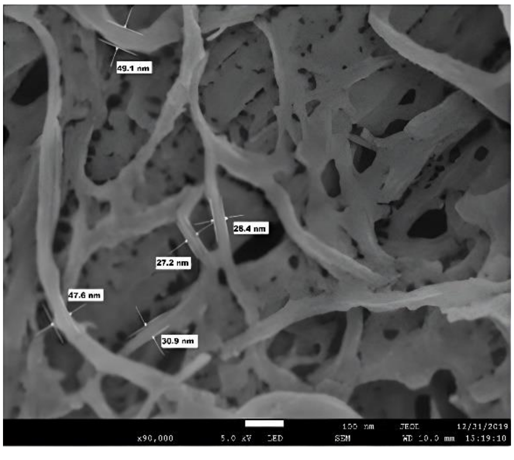
3 pieces of recycled paper (A4 size) were cut into small pieces (3cm×3cm) and then the paper was mixed in 1 L of water. The mixture was blended using a mechanical blender at 3000 rpm for 26 minutes. Then, hydrogen peroxide was added to mixture for bleaching purposes. The ratio for hydrogen peroxide to paper in this method was 1:3. Then, 1 wt.% of nanocellulose were mixed in paper pulp mixture. The mixture was stirred gently for 1 hour using a magnetic stirrer at level 4 and was left for 24 hours in the chiller. The mixture of paper pulp was poured into a netting mold in A4 size into the basin with 2 L of water inside to get the shape of a paper. Then, the paper sheet in the netting mold was dried for 2 days and was placed at room temperature. After that, the paper sheet was placed into hot compression molding at 70 °C and pressure of 5 MPa for 10 minutes. The procedures were repeated for nanocellulose concentration at 3 wt.% and 5 wt.% and pressure of 10 MPa.

2.4 Characterization

Nanocellulose size and dimension was observed via Field Emission Scanning Electron Microscope (FESEM) and Transmission Electron Microscopy (TEM). Prior FESEM analysis, samples were coated with platinum sputter for conductivity to prevent charging effect on the sample. Then, the samples were scanned onto an audiovisual display unit at 5 kV and 50 kX magnification. For TEM analysis, CNC samples were diluted in deionized water at a concentration of 0.08 mg/ml and the suspension was sonicated for 10 minutes for sample preparation.

A paper has standard international size which is (8.3x11.7 in) or (210x297 mm) in A4 size. Standard test method that was used for tensile strength is using a tensile tester by Shimadzu (AGX-V Series) tensile testing machine. While thickness of the paper, it was measured according to TAPPI test method T494. For the tensile, the paper was cut in measurement of 200mm in length and 25mm for the width using TAPPI T494.

3.0 RESULTS AND DISCUSSION



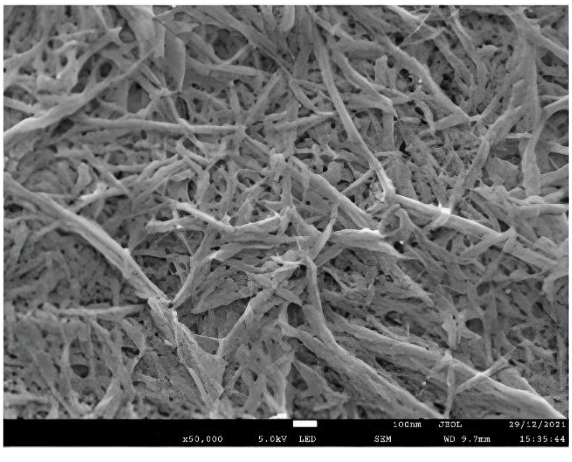


Figure 1. (Left): FESEM image of CNF under 90 kX magnification (Reproduction of image from John Wiley & Sons Ltd with permission from Yahaya et al., Synthesis of Active Hybrid Films Reinforced with CNF as Active Packaging Material 2022; 45: 8: 1448-53[14] and Figure 2. (Right): CNC-fp image under 50 kX magnification.

In Fig. 1 and 2, CNF and CNC-fp are observed at magnification of 90kX and 50kX respectively by FESEM. Fig. 3 shows the image of CNC-efb by TEM. As reported by Yahaya et al., the stackable CNFs that is shown in FESEM image suggest that CNF formed an interconnected network of fibers with the average diameter less than 50 nm [14]. The image also confirms the long smooth fibril structure of CNF. The measured average diameter and length of CNC-fp is 26.86 nm and 358.50 nm respectively, resulted in the aspect ratio of 13. As shown in Fig. 2, the CNC rods were grouped with each other to form bundles that is due to the adopted freeze-drying process [15]. CNC in its original state would portray a rod like structure, with the average diameter of 20 nm and length between 100 – 500 nm [16].

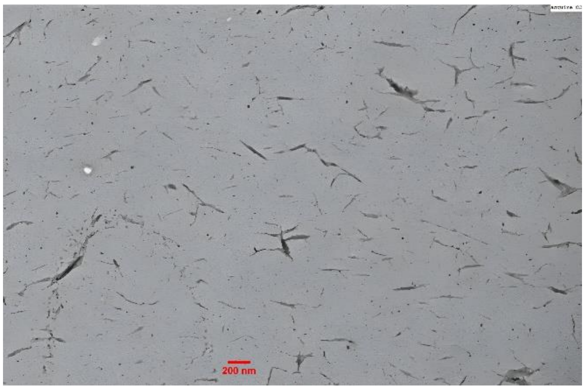


Figure 3. TEM image of CNC-efb.

The size and dimension for CNC-efb were obtained by the TEM analysis as shown in Figure 3. The average diameter and length measured is 30.73 nm and 168.57 nm respectively, resulting in the aspect ratio of 6.8. CNC-efb that is shown in Figure 3 has shown obvious rod-like structure that is commonly reported over the years.

The data for tensile strength for each type of nanocellulose that is incorporated into recycled paper was summarized in Table 1. The fabricated recycled papers were varied by the nanocellulose loading and the difference in pressure during melt compounding process. Overall, recycled paper reinforced with CNF have shown the highest tensile strength compared to the other nanocellulose.

Table 1. Tensile strength for CNF, CNC-fp and CNC-efb at different pressure and concentration.

Pressure (MPa)	Weight percent of filler (wt.%)	Tensile strength of recycled paper (N/mm^2)		
		CNC-fp	CNC-efb	CNF
5	0		8.33 ±2.35	
	1	8.31 ±1.93	6.83 ±0.64	13.67 ±1.51
	3	8.98 ±0.91	10.20 ±0.35	11.23 ±1.67
	5	6.83 ±1.05	9.24 ±0.49	7.31 ±0.62
10	0		7.72 ±0.75	
	1	7.54 ±0.64	8.37 ±1.34	14.59 ±0.49
	3	7.69 ±0.84	8.43 ±1.40	15.02 ±0.74
	5	9.18 ±1.25	6.23 ±0.03	15.28 ±1.10

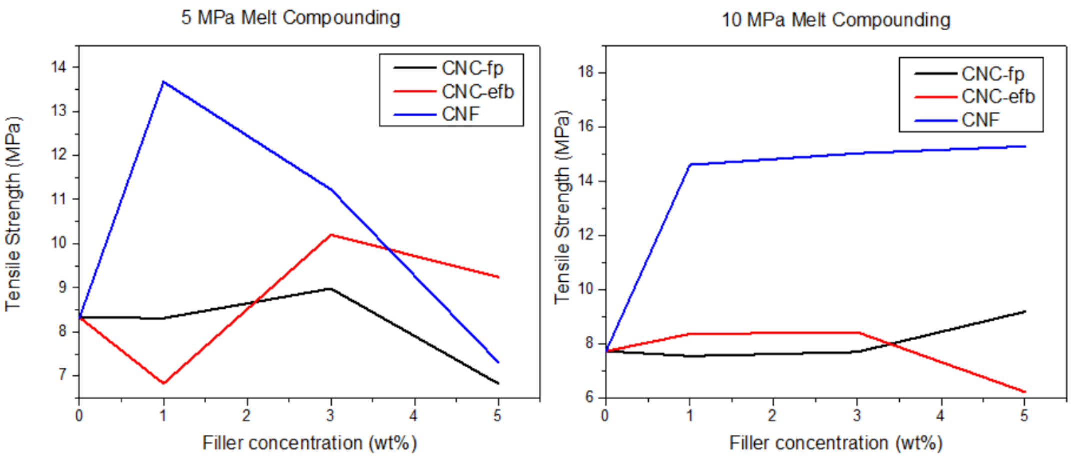


Figure 4. Tensile strength of the recycle paper at different nanocellulose loading.

It can be seen that CNF reinforced recycled paper has recorded an increase in tensile strength for different loading amount and pressure, except for 5 wt% CNF with 5 MPa melt compounding pressure. The reduction in tensile strength is believed to be due to the pressure utilized is insufficient for the CNF to fuse efficiently with the recycled paper [17]. For 5 MPa pressure compression molding, CNF has recorded a 64% and 34% increase for CNF loading at 1 and 3 wt% respectively while at 5 wt% CNF has recorded a 13% reduction in tensile strength. At 10 MPa pressure compression molding, CNF has recorded a significant increasing amount, which is 89%, 94% and 97% for 1, 3 and 5 wt% respectively.

This observable increase for CNF compared to other CNCs as clearly indicated in the table 1 is due to CNF offer a higher aspect ratio compared to CNC. CNF is commonly known has a higher aspect ratio and surface area compared to CNC and provide a promising enhancement when incorporated as a filler in polymer matrices [18]. High aspect ratio resulted into a larger amorphous region, which contained bonded hydrogen between the CNF and recycled paper. The increase in bonded area promotes fiber to fiber contact hence increasing the bonded hydrogens and resulted in an entangled nanoscale network embedded in the microfiber network. This has led into the increase of the strength of the recycled paper. As reported by Latifah et al., the nanosized dimension of CNF resulted in increased surface area and in the number of available hydroxyl groups for interaction, thus promoting the formation of fiber-to-fiber contact and bonding, which in turn solidifies the structure of the paper [19]. This is believed to be the justification on CNF to have the highest tensile strength after incorporated into recycled paper at 5 wt% CNF. The increase of pressure shows significant improvement as it assist in interfiber joint between the cellulose and paper fibre [20].

CNC-fp has shown a 7% increase in tensile strength at 3 wt% CNC-fp for 5 MPa melt compounding pressure and a 18% increase at 5 wt% for 10 MPa melt compounding pressure. At 5 MPa melt compounding pressure, CNC-fp has shown a reduction of 0.3% and 18% for 1 wt% and 5 wt% CNC-fp respectively. Slight reduction (<1%) can be observed at 1 wt% and 3 wt% of CNC-fp at 10 MPa melt compounding pressure. For CNC-efb, an increase of 22% and 11% is observed for 3 wt% and 5 wt% respectively for 5 MPa pressure melt compounding. At 1 wt% CNC-efb for the same melt compounding pressure, it has shown 18% decrease in tensile strength. Different trend has been recorded for CNC-efb at 10 MPa melt compounding pressure, which have shown 8% and 9% increase for 1 wt% and 3 wt% CNC-efb respectively. At 5 wt% CNC-efb have shown a 19% reduction in tensile strength. While the CNC enhancement to the tensile strength of the recycled paper is not as significant as compared to CNF, it also possesses hydroxyl groups (OH^-) that could form hydrogen bond together with the recycled paper matrices that contributed to the increase in tensile strength. Balea et al. emphasized that this is due to the rod-like shaped structure and a large surface area of CNC that corresponds to the properties enhancement of the paper matrices [21]. Meanwhile in pressure aspect, it shows insignificant or less improvement as CNC has short fibre dimensions which may hindered the interfiber joint [20].

4.0 CONCLUSION

As a conclusion, nanocellulose specifically CNF has shown the highest potential to strengthen recycled paper which recorded a 97% increase in tensile strength. 5 wt% CNF recycled paper prepared with 10 MPa pressure compression molding has increased in tensile strength from 7.72 MPa to 15.28 MPa. This is believed to be the impact of high aspect ratio of the nanocellulose that possess the ability to enhance the tensile strength of the recycled paper. The pressure in fabrication of the paper sheet also influences the strength of the paper [20]. It is suggested that further research be done on other types of CNC to observe its compatibility with recycled paper as CNCs has known to differ in enhancing properties.

5.0 CONFLICT OF INTEREST

The authors declare no conflicts of interest to any financial or non-financial interests regarding political, personal and professional relationships in this manuscript.

6.0 AUTHORS CONTRIBUTION

A.R.H Hipeni (Writing – review and editing; Data curation; Formal analysis; Visualisation; Methodology)

T. J. Wei (Data curation; Investigation; Writing – original draft)

N. S. J. Shah (Data curation; Investigation; Writing – original draft)

K. N. M. Amin (Funding acquisition; Project administration; Supervision)

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