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Cellulose Nanofibers from Waste Paper and their Utilization as Reinforcement Materials in Poly((*R*)-3-Hydroxybutyrate-*co*-(*R*)-3-Hydroxyhexanoate Bionanocomposite

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ABSTRACT

Waste paper is the second-highest municipal solid waste collected in Malaysia and as current practice, it is recycled for further use in the manufacturing of low-grade products. Instead of continuously utilizing waste paper for low-grade products manufacturing, it can be used as a feedstock to produce high bioproducts such as cellulose nanofiber (CNF). Hence, this study explored the potential of waste paper as a feedstock for CNF production. The waste paper was subjected to a different number of cycles of wet disk milling (WDM): 0, 5, 10, 15 and 20 cycles. The presence of nano-sized cellulose was confirmed by FE-SEM micrographs, where CNF with diameter size 20 – 40 nm was formed after 10 cycles of milling. It was also revealed that the obtained CNF possessed appropriate properties as a reinforcement material. The tensile strength and Young's modulus of poly((R)-3-hydroxybutyrate-*co*-(*R*)-3-hydroxyhexanoate (PHBHHx) increased by 19 and 12%, respectively after the reinforcement of 1% CNF. Overall, this study portrays that waste paper could be utilized as a raw material for CNF production, without the need for chemical pretreatment.

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INTRODUCTION

Over the years, the demands of cellulose increases because of its strength, biodegradability, renewability, and biocompatibility especially in paper, board

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and composite industries (Arévalo & Peijs, 2016; Gao et al., 2013; Sreekala et al., 1997; Wang et al., 2010). Despite of its excellent properties, cellulose production however causes environmental issues mainly because of the harmful chemical usage and inefficient effluents discharge to extract it from the raw material (Abraham et al., 2013; Prakash et al., 2017). In order to minimize the environmental pollution, wastes comprising cellulose such as waste paper can be reprocessed to produce high value-added bioproducts.

Waste paper is a combustible and post-consumer waste that is usually disposed in a trash bin before being piled up on land. Currently, waste paper is the second-highest municipal solid waste collected in Malaysia behind organic waste (47%), which is around 15% (Jereme et al., 2015). As a current practice, waste paper is recycled for further use in the manufacturing of new fibrous products such as low-grade paper and board (Hietala et al., 2018). Nevertheless, the recycling process may shorten the fibers and results in strength reduction. Hence, instead of utilizing waste paper for low-grade products, it can be used as a feedstock to produce value-added bioproducts such as cellulose nanofiber (CNF). Indirectly, the use of secondary raw material like waste paper will result in higher resource efficiency, thus may avoid resource depletion and unnecessary disposal of wastes (Hietala et al., 2018).

CNF is now mainly derived from various plant fibers that have been refined to nanoscale. Due to the fibrillation process, the hydrogen bonds (H-bonds) are broken down, hence forming nano-sized fibers in diameter called CNF. In general, CNF consists of alternating crystalline and amorphous domains (Chang et al., 2012). It also has long and flexible linked fibers around 20 to 100 nm in diameter, and several micrometers in length (Tibolla et al., 2014).

It is worth noting that CNF nowadays has received much attention from various fields due to its superior physical properties, which mainly derived from its nano-size structure as well as its chemical, biological and mechanical characteristics such as having high specific surface area, high crystallinity, thermally, chemically and rheologically stable, biodegradable, biocompatible and lack of toxicity (Lin & Dufresne, 2014). Many studies have been conducted to determine CNF potentials in various fields ranging from household materials to high-tech industrial applications. CNF is expected to be a promising renewable nanomaterial that can replace uses of non-renewable materials in various products manufacturing including as a reinforcement material to produce bionanocomposites.

Besides the use of degradable reinforcement materials, an attempt has also been widely conducted to replace synthetic polymers as one of the environmentally friendly solutions to minimize environmental pollution. Poly((*R*)-3-hydroxybutyrate-*co-(R)*-3-hydroxyhexanoate (PHBHHx) has been highlighted as one of the potential bio-polymer for various polymer-based products manufacturing. Despite of its biodegradability, the shortcoming of PHBHHx lies on its low thermal, crystallinity and mechanical properties

compared to synthetic plastics (Jing, 2012). Hence, this limits its usage for only certain applications. In fact, the low crystallization rate of the polymer serves as an obstacle during the industrial processing of the material (Buntinx et al., 2017). In order to tackle this obstacle, researchers have looked into techniques such as adding nucleating fillers, microfillers or nanofillers and the incorporation of CNF is expected to help overcome this matter (Xu et al., 2017). Improved property of PHBHHx will widen its usage in various applications, range from household to high-end applications, and indirectly will widen the utilization of PHBHHx as single-use plastics, and ultimately contributes to the less accumulation of single-use plastics.

Hence, this study explored the feasibility of WDM to produce CNF from waste paper. The potential of waste paper-CNF as a reinforcement material for PHBHHx bionanocomposite was also evaluated by analyzing the morphological, thermal and mechanical properties.

MATERIALS AND METHODS

Materials

Waste papers were collected from Administrator Office, Universiti Putra Malaysia. The papers were cut to approximately 3 cm using a milling cutter and stored in a plastic bag at room temperature prior to further processing. Poly((*R*)-3-hydroxybutyrate-*co*-*(R)*-3-hydroxyhexanoate (P(HB-*co*-11%-HHx)) (Mn 126,000, Mw 352,000 Da after purification) was supplied by Kaneka Corporation Japan and used as received.

Production of Cellulose Nanofiber by Wet Disk Milling

Waste papers were soaked in distilled water at a concentration of 2 wt% for 72 hours prior to wet disk milling (WDM). The suspension was then subjected to wet disk mill grinder (Multi mill, Grow Engineering Co. Ltd., Adachi-ku, Tokyo, Japan) for 5 - 20 cycles at 1800 rpm to produce CNF. The CNF slurry was then stored in a sealed bottle at 2°C prior to usage and an approximate 10 mL of slurry suspension was freeze-dried for characterization.

Production of Bionanocomposite

Bionanocomposites made up of 95 – 99 wt% PHBHHx and 1 – 5 wt% CNF were melt blended using a Brabender Plastograph EC internal mixer (Brabender GmbH & Co. KG, Germany) with a mixing speed 60 rpm for 20 min at 150°C. Water molecules from the CNF suspension were evaporated from the hopper, resulted in the formation of PHBHHx/CNF bionanocomposite lump. The PHBHHx/CNF bionanocomposites were then hot-pressed at 150°C for 5 min under 10 atm of pressure to obtain bionanocomposite sheets.

Morphological Analysis

The morphology of CNF was analyzed by field-emission scanning electron microscopy (FE-SEM) (Sirion 200, FEI, Eindhoven, Netherlands) (Yasim-Anuar et al., 2017). The CNF was silver-coated for 50s using vacuum sputter-coated prior to FE-SEM analysis.

X-ray Diffraction Analysis

The crystallinity index (*Crl)* of CNF from the waste paper was analyzed using X-ray diffraction (Shimadzu XRD-6000, Kyoto, Japan). The data was generated with a scan range from 5° to 50°. The *Crl* was determined using the following equation:

$$Crl = \frac{I_{002} - I_{am}}{I_{002}} \times 100\%$$

It is to note that I_{002} at an angle of 2θ=23 and I_{am} at an angle of 2θ=18 correspond to the cellulose and amorphous region respectively (Yasim-Anuar et al., 2018).

Thermogravimetric Analysis

Thermal stability and decomposition temperature of CNF obtained from waste papers and PHBHHx/CNF bionanocomposites were determined by the thermogravimetric analyzer (TGA) (TG 400, Perkin Elmer, Waltham, MA, USA). Samples weighing in a range of 9 – 10 mg were analyzed at a heating rate of 10°C/min under a nitrogen flow of 20 mL/min, from 50 to 550°C (Yasim-Anuar et al., 2019).

Mechanical Analysis

The tensile properties of PHBHHx/CNF bionanocomposites were measured based on ASTM D638 and ASTM D790, using Intron Universal Testing Machine (P5567, 30kN, INTROP Universiti Putra Malaysia, Malaysia). The reported values were the average value of five measurements of each sample.

RESULTS AND DISCUSSION

Characterization of Cellulose Nanofiber from Waste Paper

Morphological Observation. The morphology of waste paper and CNF was observed by FE-SEM as shown in Figure 1. From the FE-SEM micrograph, an irregular structure and presence of bundles of fibers with a size larger than 1000 nm can be observed for waste paper prior to WDM (Figure 1a). Reduction in diameter size could be observed after WDM, and this could be seen as early as 10 cycles. The diameter size of waste paper was reduced from approximately 100 – 900 nm for 5 cycles, to 20 – 40 nm after 10 cycles, 10 – 25 nm

after 15 cycles and 5 – 25 nm after 20 cycles of WDM. This might be attributed to shear and frictional force at the surface layer of cellulose fibers caused by milling stones during WDM processing. During WDM, cellulose fibers were grounded by two counteracting grinding disks at extremely high speed, impact forces and friction. This physical damage then causes fibrillation by breaking down the hydrogen bonds of cellulose fibers, which leads to the transverse cleavage of them along the longitudinal axis (Dubey et al. 2018). Hence, the large bundles of fibers can be disintegrated into individual nanosize fibers with the network structure.

From the morphologies of CNF after WDM as shown in Figure 1 (a-e), the single fibril obviously reduced to nanoscale size by increasing the milling cycles, suggesting that CNF was successfully extracted from cellulose fibers by using the present method. In fact, it was also revealed that no obvious difference could be observed by increasing the WDM cycles up to 20 cycles. This was in contrast with other nanofibrillation methods like ultrasonication. Our previous research on ultrasonication revealed that by prolonging the ultrasonication to more than 9 hours might lead to re-agglomeration of CNF, and eventually increased the diameter size of CNF (Yasim-Anuar et al., 2018). This might be attributed to the reformation of H-bonds between CNF as well as due to the attraction of van der Walls forces, which mainly occurred due to stress on the CNF surface. This phenomenon occurred mainly due to the longer processing time and adhesion of CNF to one another (Yasim-Anuar et al.,

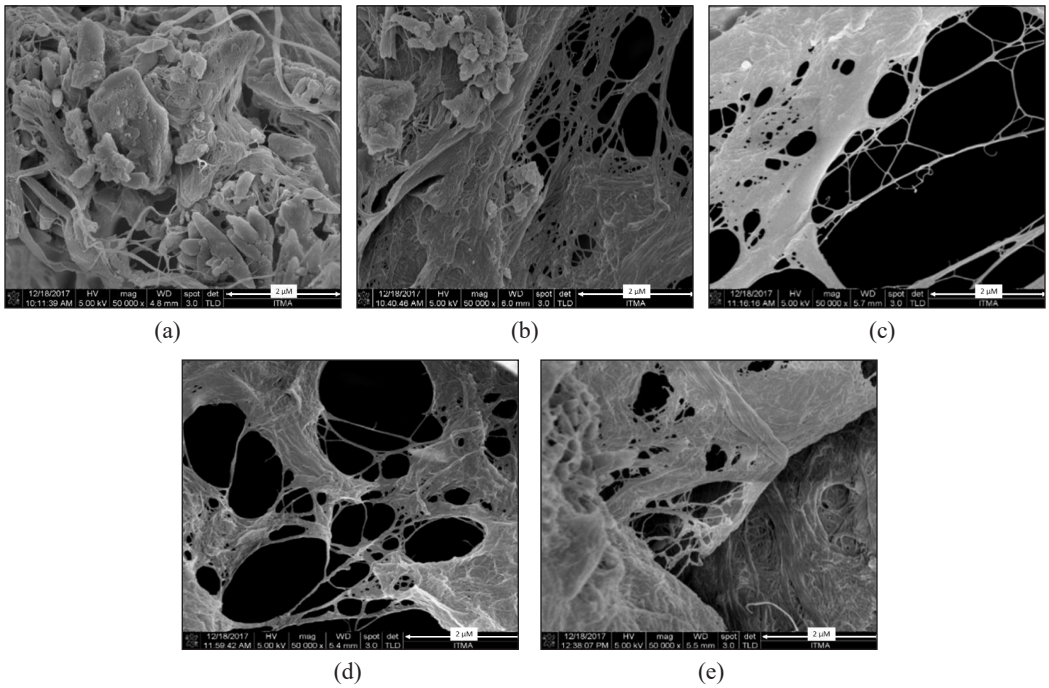

Figure 1. FE-SEM micrographs of (a) cellulose from waste paper and CNF after (b) 5, (c) 10, (d) 15 and (e) 20 cycles. The magnifications for (a) and (b-e) were 10,000x and 50,000x, respectively.

2018). This, however, did not occur to CNF produced by WDM and this proved that this method was more efficient in nanofibrillating CNF into smaller diameter sizes.

Thermal stability and Crystallinity Index of Cellulose Nanofiber

Thermogravimetric analysis was carried out to investigate the thermal stability of CNF. The obtained TG and DTG thermograms of cellulose from waste paper and CNF were plotted as shown in Figure 2. A small weight loss was observed around 50 – 170 ºC mainly due to the evaporation of water from CNF (Yasim-Anuar et al., 2019). The dramatic weight losses were found in a temperature range between 200 – 350 ºC for all samples, which corresponds to the decomposition of cellulose. This finding indicates that cellulose decomposes rapidly at a higher temperature. It was identified that the temperature degradation ($Td_{50\%}$) reduced by prolonged the WDM process. The $Td_{50\%}$ reduced from 340 ºC to approximately 328 – 336 ºC after milling. The reduction was highly attributed to the exposure of CNF to high shear and frictional forces, caused by grinding stones during WDM. In addition, thermal degradation might occur early due to the reduction in diameter size and increment of surface area (Abraham et al. 2013).

This result also corresponded to the *Crl* which was analyzed by XRD. Figure 3 shows that all samples exhibited similar diffraction peaks at around 2θ=21.5 – 23.5. The *Crl* of cellulose fiber was approximately 85% prior to milling, and the value reduced to 80 – 82% after milling. Mechanical forces from high shear and frictional force during WDM could damage the crystalline structure of CNF, hence reduce the *Crl* values. This was supported by reports from Jang et al. (2015), Norrrahim (2018) and Zakaria et al. (2015). Hence, it can be noted that by prolonging the milling process, the crystalline structure of CNF might be disrupted, thus reducing the *Crl* value.

Based on the morphology and average diameter, it was revealed that by WDM the waste paper up to 10 cycles was able to produce CNF with size less than 100 nm, which

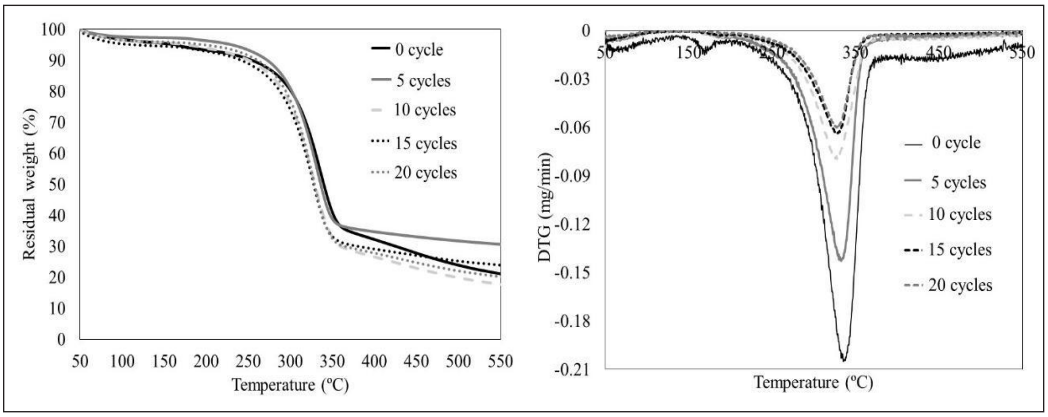

Figure 2. The TG and DTG thermograms of CNF produced by different milling cycles

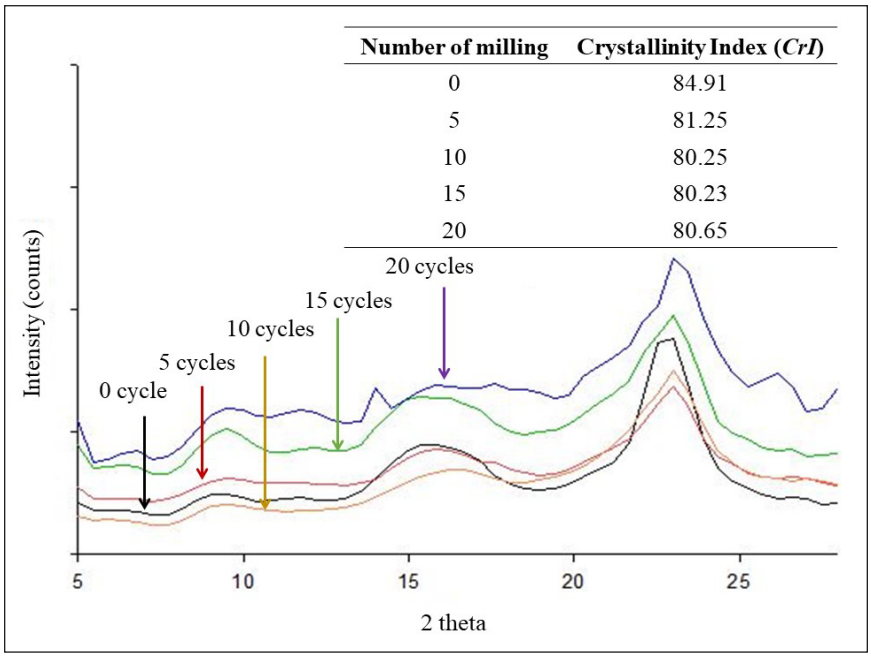

Figure 3. The XRD diffraction patterns of CNF produced by different milling cycles

fulfilled the standard of Malaysian NanoVerify. In fact, by taking into consideration the crystallinity and thermal stability, the results only showed a slight reduction of about 3.2 and 5.5%, respectively after 10 cycles of milling, in comparison to the cellulose before being nanofibrillated. It was also revealed that the CNF produced using this nanofibrillation method was having comparable properties with CNF produced from other nanofibrillation methods as shown in Table 1. This indirectly proves that WDM exhibited excellent CNF formability as compared to other fibrillation methods.

Evaluation of CNF Production from Waste Paper by Wet Disk Milling

Conventionally, CNF is mainly produced from lignocellulosic biomass, agricultural waste, softwood and hardwood. In order to produce CNF from these sources, pretreatment is needed to extract only cellulose and remove other impurities (Yasim-Anuar et al., 2019). In contrast to other sources as shown in Tables 1 and 2, pretreatment is unnecessary and can be skipped for waste paper as they are mainly composed of cellulose. Concerns regarding the ink contained in the waste paper may rise, however, the deinking process can be skipped if the targeted application does not involve direct contact with foods and humans, such as for the manufacturing of automotive compartment, building and construction materials. Regardless of having impurities, which mostly comes from ink, the CNF produced from waste paper is having similar characteristics with CNF produced from other sources and this was highly due to the effectiveness of WDM.

Table 1
Properties of CNF produced from different sources and nanofibrillation methods

Source of cellulose	Fibrillation methods and conditions	Findings	Ref.
OPMF-cellulose	*Method:* Electrospinning *Condition:* Voltage: 15kV - Concentration: 6% (w/v) - Solvent: ([EMIM]Cl), ([C10MIM][Cl]) and DMF	Diameter: 200 – 500 nm *Crl*: 56% $T_{d50\%}$: 254°C	Yasim-Anuar et al. (2017)
Sisal fibers *(Agave sisalana)*	*Method:* Ultrasonication Condition: Time: 2 h Power: 750 W Solution: Ethanol	Size: 50 nm *Crl*: 78.9% $T_{d50\%}$: -	Sosiati et al. (2017)
Tunicate, unbleached hardwood *(Eucalyptus grandis×E. ur ophylla)* and softwood *(Picea abies)*	*Pre-fibrillation:* Cellulose suspension was subjected to enzyme hydrolysis and TEMPO-oxidation *Method:* High-pressure homogenization: *Condition:* Pressure: 925 bar (1st pass) and 1600 (subsequent passes) Passes: 5	Diameter size: 10 – 70 nm *Crl*: - $T_{d50\%}$: 313.8°C	Zhao et al. (2017)
Oil palm empty fruit bunch (OPEFB)	*Method*: Wet grounded using Fritsch Pulverisette 7 nano-grinder *Condition*: Time: 30 min Speed: 722 rpm Concentration: 5% (w/v)	Diameter size: 10 – 100 nm *Crl*: 85.09% T_{max}: 350°C	Supian et al. (2020)
Waste paper	*Method:* Wet disk milling *Condition:* Cycles: 10 cycles Speed: 1800 rpm Concentration: 2% (w/v)	Diameter size: 20 – 40 nm *Crl*: 80 – 82% $T_{d50\%}$: 328 – 336ºC	This study

The utilization of waste paper and chemical-free nanofibrillation method (WDM) for CNF production was to promote the idea of using renewable and sustainable reinforcement material for polymer composites. The implication of the processing method on CNF production was evaluated according to the Principles of Green Chemistry (Anastas & Warner, 1998; Cinelli et al., 2017). Based on the overall evaluation, it was observed that by nanofibrillating waste paper using wet disk milling method was able to fulfill six criteria of the green chemistry principle. Among the criteria are, no waste generation, hazardless

Table 2
Pretreatment for extracting cellulose prior to nanofibrillation

Sources	Pretreatment	Nanofibrillation	Findings	Ref.
Henequen *(Agave fourcryodes)*	Dewaxed with a (3:1, v/v, toluene and ethanol) for 72h at 45°C, followed by KOH solution at room temperature for 8 h. Then, the fibers were treated with 3M HCl, followed by acid hydrolysis and bleaching with a mixture of $NaClO_2$ and glacial acetic acid (5:1) for 4 h at 50°C.	Homogenizing at 15,000 rpm for 4 h.	Diameter size: 51 – 103 nm *Crl* (%): - T_{max}: -	Fazeli et al. (2018)
Balsa tree	Bleached with a mixture of $NaClO_3$ and CH_3COOH solution at 70°C for 1 h. The process was repeated 3 times.	Grinding using ultra-fine friction grinder for 30 passes at 1500 rpm.	Diameter size: 1-100 nm *Crl* (%): - T_{max}: -	Kumode et al. (2017)
Oil palm mesocarp fiber	Treated with 5% (w/v) $NaClO_3$ solution at 70 °C for 90 min, followed by 6% (w/v) KOH solution at room temperature for 24 h.	Extruded at 100 rpm for 1h.	Diameter size: Less than 100 nm *Crl* (%): - T_{max}: -	Yasim-Anuar et al. (2019)
Waste paper	No pretreatment was carried out.	Wet disk milling at 1800 rpm for 10 cycles.	Diameter size: 20 – 40 nm *Crl* (%): 80 – 82% $T_{d50\%}$: 328 – 336ºC	This study

processing, chemicals and solvents-free process, use of renewable feedstocks as well as process effectiveness and safety level (Cinelli et al., 2017).

The 1st principle highlights on waste management. Under this principle, waste generation should be prevented as it may lead to costly waste treatment and environmental issues. Considering a continuous nanofibrillation process of cellulose suspension by WDM to produce CNF, waste generation can be prevented using this method. Principle number 3, 4 and 5 highlight on the hazardless, chemicals as well as the solvent-free process. The WDM is hazardless as it does not involve the use of chemicals along with the treatment. Hence, it is relatively safer for the operator and environment to adapt this technology to produce CNF. Other than that, this study also fulfills the requirement of the 7th principle. As one of the post-consumer wastes, waste paper has not been fully utilized for valuable bioproducts manufacturing despite made up of cellulose. The use of waste paper as a feedstock for this study proves that the high-value-added CNF can be produced from discarded materials. This study also meets the requirement of principle number 12, which stress on accident prevention. WDM is conducted at atmospheric pressure and involves no corrosive chemicals, hence it is safe to be operated. Based on the overall evaluation, WDM

can be considered as an alternative, safe and environmentally-friendly nanofibrillation treatment that can produce CNF effectively.

Characterization of Bionanocomposites Reinforced CNF

To evaluate the reinforcement effect of CNF produced in this study, the CNF was incorporated in PHBHHx by liquid assisted melt-blending method. The mechanical analysis revealed that CNF was able to improve the tensile strength and Young's modulus of PHBHHx/CNF bionanocomposites as shown in Table 3.

Table 3
Mechanical properties of PHBHHx reinforced CNF

	Tensile strength (MPa)	Young's modulus (MPa)
Neat PHBHHx	28.05 ± 1.27	2320.1 ± 97.81
PHBHHx/1% CNF	34.67 ± 1.68	2636.28 ± 93.18
PHBHHx/3% CNF	33.66 ± 1.68	2537.72 ± 87.06
PHBHHx/5% CNF	33.43 ± 1.30	2613.87 ± 93.03

It was observed that both tensile strength and Young's modulus increased up to 19 and 12%, respectively after 1 wt% CNF was incorporated in the PHBHHx. Thus, this proved that the incorporation of CNF significantly improved the mechanical properties as compared to the neat PHBHHx. In fact, it could be reached at a low reinforcement level. This proved that CNF and PHBHHx had good interfacial adhesion mainly due to their hydrophilicity properties. The adhesion of CNF and PHBHHx was influenced by hydrogen bond formed between CNF-matrix chain and eventually created a strong fiber-matrix adhesion, thus able to enhance the mechanical properties of biocomposites (Kargarzadeh et al., 2017). This was in accordance with other studies that also incorporated a low amount of CNF in polymers such as thermoplastic starch (Fazeli et al., 2018), polyethylene (Yano et al., 2018) and polyether block amide (Ziaei-tabari et al., 2017).

However, it was also noticeable that the values were slightly reduced when 3 and 5 wt% CNF were incorporated in the PHBHHx, but these were still higher than those of neat PHBHHx as a reference. This might be because of the agglomeration of CNF in the polymer matrix. According to Herrera et al. (2016), CNF tends to agglomerate during melt-processing, and as the filler content increased, the size of the agglomerates increases, thus broaden their distribution. This indirectly leads to poor mechanical properties of the composites. In fact, according to Mittal et al. (2018), reduction in mechanical properties starts to occur after reaching a threshold limit and this is mainly due to poor mechanical interlocking, which may degrade load transfer between fibers and matrix. Indirectly, this explains the reduction in mechanical properties for PHBHHx bionanocomposites composed of 3 and 5 wt% CNF.

CONCLUSION

The environmentally friendly WDM process was found to be able to nanofibrillate waste paper from micrometer range to 20 – 40 nm after 10 cycles of milling. It was also revealed that the obtained CNF was able to improve the tensile strength and Young's modulus of PHBHHx by 19 and 12%, respectively higher than the neat polymer. Overall, the utilization of waste paper-CNF as a reinforcement material for PHBHHx bionanocomposite is possible as evidenced in this study, without compromising the mechanical performance of the bionanocomposites produced. The CNF produced by WDM was also found to have low environmental impacts, thus able to promote sustainable and safe nanofibrillation processing.

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