Bacterial Cellulose as Reinforcement in Paper Made from Recycled Office Waste Pulp

Evren Ersoy Kalyoncu,[a,]* and Emrah Peşman [b]

Bacterial cellulose, produced during fermentation of Kombucha tea, was investigated relative to its ability to modify the characteristics of pulp from recycled office wastepaper. The produced bacterial cellulose wet films were dispersed and added rates of 5%, 10%, and 15% to the recycled office wastepaper. The Fourier-transform infrared spectroscopy analyses, scanning electron microscopy images and thermogravimetric analysis values were determined in order to characterize the pulp samples. The results of these analyses showed similar changes as the amount of added bacterial cellulose increased, which also meant an increased amount of filler attaching to the fiber matrix. The burst index and tensile index values were protected while the tear index value partially decreased as the amount of added bacterial cellulose increased. The brightness values of the bacterial cellulose reinforced papers did not change after thermal aging, while the changes in the yellowness values were quite limited. Higher water absorption rates, and lower air permeability values were obtained from bacterial cellulose reinforced recycled office wastepaper sheets, which corresponded to the addition of increased bacterial cellulose amounts. Considering the mechanical and physical properties of the reinforced paper, bacterial cellulose represents a promising alternative for the reinforcement of office wastepaper.

Keywords: Bacterial cellulose; Office wastepaper; Kombucha; Filler; Physical and mechanical properties

*Contact information: a: Department of Material and Material Processing Technologies, Karadeniz Technical University, Arsin 61900 Trabzon; b: Department of Forest Industrial Engineering, Artvin Coruh University, Artvin 0800 Turkey; *Corresponding author: eersoy@ktu.edu.tr*

INTRODUCTION

The fibers used in the pulp and paper production industry are generally obtained from wood. However, researchers have been looking for suitable alternative raw materials to wood due to the increase in global population, awareness of society, new laws being enacted, the advancement of technology, and the difficulty in obtaining and using forest resources. Recycling and reuse have been important issues around the world since the 1980s (Ruth and Harrington 1997). Recycling wastepaper resources and reusing them in paper production were among the first alternative raw materials to wood, due to their advantages, especially environmentally and economically (Bajpai 2014). Since it makes up a major portion of all wastepaper, recycling of office wastepaper (OWP) has become a meaningful and challenging process. However, the quality of the paper produced from wastepaper is often lower than those produced from virgin pulp, due to the shortened fiber length and reduction in tensile strength achieved during production (Lei *et al*. 2018).

Cellulose, the most basic component of the paper, is one of the most abundant polymers in nature and is generally obtained from plant sources but can also be obtained from different sources, *e.g.*, bacteria (Orue *et al*. 2017*)*. Bacterial cellulose is an interesting and promising polymer, and it has been gathering attention due to its excellent properties, *i.e.*, its exceptional physical-chemical properties, environmentally friendly processing properties, low production costs, hydrophilicity, high mechanical properties, excellent biocompatibility, and biodegradation properties (Ullah *et al.* 2016; Gao *et al*. 2019; Abol-Fotouh *et al.* 2020). Compared to natural plant cellulose, bacterial cellulose (BC) is remarkably similar in chemical composition and fiber structure (Gallegos *et al.* 2016), but BC exhibits superior structural and mechanical properties. (Klemn *et al.* 2011; Fillat *et al.* 2018). It is characterized by its high purity (free of lignins, pectin, and hemicelluloses) (Reiniati *et al.* 2017), strength, mobility, higher crystallinity, higher specific surface area, higher degree of polymerization, higher mechanical properties, higher and stronger biological adaptability, and higher liquid absorbing and holding capacity (Miyamoto *et al.* 1989; Yamanaka *et al*. 1989; Ross *et al.* 1991; Klemm *et al.* 2001; Klemm *et al.* 2005; Bäckdahl *et al.* 2006; Ashori *et al.* 2012; Chen *et al.* 2016; Torres *et al.* 2019).

Using symbiotic consortia of bacteria and yeast kombucha culture (SCOBY) is one way to produce bacterial cellulose (Dima *et al.* 2017; Kaminski *et al*. 2020). In this way, bacterial cellulose can be produced with a culture that is more economically available and easy to propagate (Kaminski *et al*. 2020). It was originally used to produce some functional drinks, *e.g.*, kombucha or Manchurian tea (Güzel and Akpınar 2018). Kombucha is a traditionally consumed fermented drink made *via* the fermentation of water, tea, and sucrose mixture by SCOBY (Dufresne and Farnworth 2000; Teoh *et al.* 2004; Malbaša *et al.* 2011; Goh *et al.* 2012; Villareal *et al.* 2018). With the fermentation process, bacteria synthesise a cellulose network on the nutrient medium surface and the synthesized cellulose is not consumed with kombucha (Domskiene *et al.* 2019). BC is considered as an attractive biopolymer due to its ability to be produced by simple technology. Many studies have stated that *Acetobacter xylinum* is the primary bacteria in produced bacterial cellulose mat (Hesseltine1965; Jankovic and Stojanovic 1994; Liu *et al.* 1996; Mayser *et al.* 1995; Sievers *et al.* 1995; Greenwalt *et al.* 2000). *Acetobacter xylinum* is the nature's most prolific cellulose-producing bacterium.

The use of biotechnology in paper production is seen as an important benefit for economic and environmental purposes (Basta and El-Saied, 2009). Because of its structural properties, BC is considered an attractive biopolymer for pulp and paper. The purpose of this study was to investigate the usefulness of bacterial cellulose as an additive in the reinforcement of recycled OWP and to improve the quality of recycled OWP. Considering the increasing global demand for recycled papers in recent years, it is quite remarkable with this study to improve the worse quality of recycled papers by BC reinforcement. This study is expected to provide new insights for the reusability of OWP and create additional usage possibilities of these papers in different areas for different purposes.

EXPERIMENTAL

Materials

Re-pulping non-printed office wastepaper

Non-printed office wastepapers, which were obtained from various commercial suppliers, were used in this work. The ash amounts of the office paper used in the study at the beginning and after the re-pulping process were determined as 14.2% and 10.1%, respectively. The OWP was cut into small pieces (approximately 2.5 cm x 2.5 cm) and transferred in a 2 L capacity laboratory pulper. The loading of the pulper was performed manually, while the system heating and temperature control were done digitally. The temperature in the pulper was set to 50 °C, and the consistency was adjusted to 15% by adding a calculated amount of hot tap water. The samples were allowed to wait 5 min to become wet, and then the repulping was performed at a speed of 1000 rpm for 10 min, and 1 wt.% of NaOH was added for easy swelling and fibrillation during the process. For each trial, 100 g of oven-dried white office paper pieces were used.

Preparing the bacterial cellulose

The bacterial cellulose used as an additive in this study was obtained from a yeast-bacteria kombucha culture (SCOBY).

The nutrient broth medium for growing BC was produced as follows: 1000 mL of boiled water was mixed with 2 black tea bags and 200 g of sucrose (as a carbon source), which was then cooled to 25 °C. The fermentation process was performed by adding cider vinegar (4 to 8% acetic acid concentrations) and symbiotic culture of bacteria and yeast (SCOBY), obtained from commercial suppliers. Culture, generally includes acetic acid bacteria *(Acetobacter*, *Gluconobacter*, and *Komagataeibacter*), lactic acid bacteria (*Lactobacillus*), and various yeasts (*Saccharomyces* and *Zygosaccharomyces*) (Bokulich and Bamforth 2013; Villarreal-Soto *et al.* 2018). At the end of the fermentation period (15 d), a BC gel biomass with a thickness of approximately 1 cm on the surface of the fermentation media was recovered, which was first put into alcohol and then boiled in water for 40 min, in order to clean off the bacteria. Afterwards, the BC gel was immersed in a 0.1 M NaOH solution and boiled for 20 min, which occurred twice to ensure cellulose purification. Finally, the BC gel was washed with distilled water until the pH reached approximately 7.0. This process yielded the wet cellulose membrane used later in the study.

Paper Sheets Making

The produced BC wet films were dispersed into a slurry with a homogenizer (Silent Crusher Homogenizer, Heidolph, Schwabach, Germany) at a stirring speed of 25000 rpm for 5 min to separate bundles and to easily mix in the wastepaper fibers. Using this method, the hydroxyl groups on the cellulose molecular chain were exposed and the formation of a hydrogen bond between the plant fiber and the bacterial fiber was facilitated (Yuan *et al.* 2016). The concentration of the obtained slurry was determined and added to the OWP pulp at rates of 5%, 10%, and 15%. All paper sheets, including the control, were prepared from a 2% consistency pulp mixture. For each BC additive, 10 paper sheets were prepared on a Rapid -Köthen sheet former with a diameter of 20 cm, according to TAPPI standard T205 sp-02 (2006). The handsheets were equilibrated at 23 °C ± 1 °C and 50% ± 2 % relative humidity before testing their physical properties.

Paper Characterization

Physical, mechanical, and optical properties of recycled office wastepaper

The basis weight and thickness of sheets were determined according to TAPPI standard T410 om-08 (2013) and TAPPI standard T411 om-05 (2005), respectively. The density of the test papers was calculated by the relationship between basis weight and thickness. The tensile strength, burst strength, and tear strength of the sheets were determined according to TAPPI standards T494 om-01(2006), T403 om-02 (2010), and T414 om-12 (2012), respectively. The ISO brightness and yellowness of the paper sheets were measured accordance to ISO/DIS standard 2470 (2016) and ASTM standard E313 (2005), respectively. The color values of the paper sheets were calculated using the L^*, *a**, *b** values, which were measured with a UV-spectrophotometer (Konica-Minolta, cm-2600d, Osaka, Japan) using a UV filter according to TAPPI standard T527 om-13 (2013). In addition, the effect of thermal aging on the optical properties of BC reinforced pulp in comparison to the control was also investigated. Ten different test paper sheets were used for each measurement. Thermogravimetric (TGA and DTGA) analyses and thermal aging analysis (15 h at 105 °C) were performed to determine the thermal properties. The water absorption (g/m^2) capacity was determined in accordance with TAPPI standard T441om-13 (2013) with a Cobb sizing tester. The results were reported as an average of 5 measurements per each sample. To measure the air resistance (s/100 mL) of the paper samples (felt side), a Gurley porosimeter was used in accordance with TAPPI standard T460 om-02 (2006).

Scanning electron microscope (SEM) imaging

A Carls Zeiss Evo LS-10 scanning electron microscope was used to study the bacterial cellulose, wastepaper fiber, and filler interaction of the samples (before imaging all samples were coated in gold).

Fourier-transform infrared spectroscopy-attenuated total reflectance characterization

The FTIR-ATR (Fourier-Transform Infrared Spectroscopy-Attenuated Total Reflectance) spectra of the handsheets were measured using a Shimadzu IR Prestige-21 FTIR device with a Pike Miracle ATR attachment. The measurement range was 600 cm^{-1} to 4000 cm^{-1}, the resolution was 16 cm^{-1}, and the number of repetitions was 16.

Thermal analysis

The thermogravimetric analyses of the samples were determined using a Perkin Elmer STA 6000 device. During the test, the temperature was increased from room temperature to 600 °C by 10 °C/min steps, with a nitrogen gas flow of 20 mL/min.

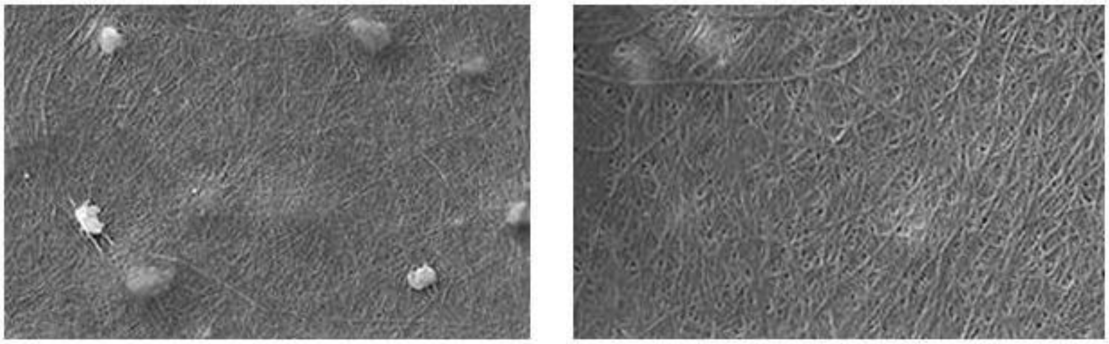
RESULTS AND DISCUSSION

Characterization of Dispersed Bacterial Cellulose and Bacterial Cellulose Reinforced Recycled Office Wastepaper Fibers

Scanning electron microscopy (SEM)

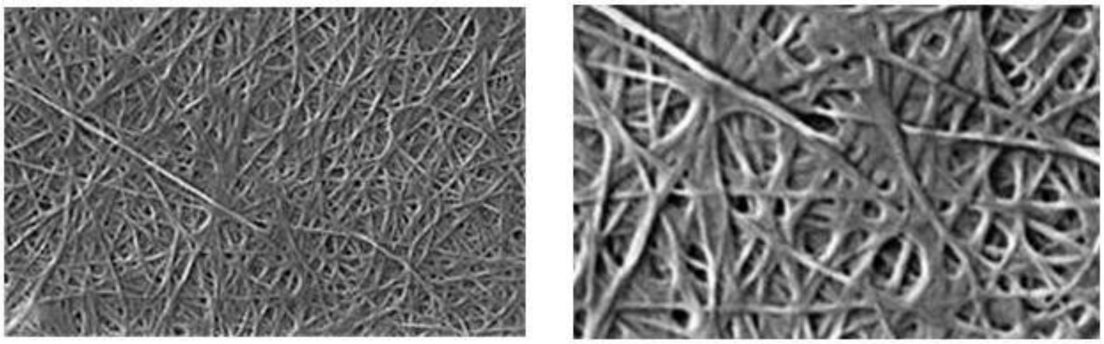
In order to investigate any improvements and/or modifications from the addition of BC to recycled OWP pulp characteristics in detail, SEM images of the samples were examined.

Figure 1 shows the SEM images of BC at 10000 X, 20000 X, 40000 X, and 100000 X magnifications. As shown in the SEM images, the dispersed BC typically presented itself as an interwoven mesh of fibrils. As can be seen from the images, BC had a tight mesh structure. In the 10000 X magnification images, it can be seen that the bacterial cellulose became an almost gapless structure due to the nano-sized fiber thickness. It is also seen from the image at 100000 X magnification that the dispersed BC had fibrils that were approximately 60 nm to 70 nm in width.



(a) Bacterial cellulose fibers (BCF)
10.00 KX

(b) Bacterial cellulose fibers (BCF)
20.00 KX



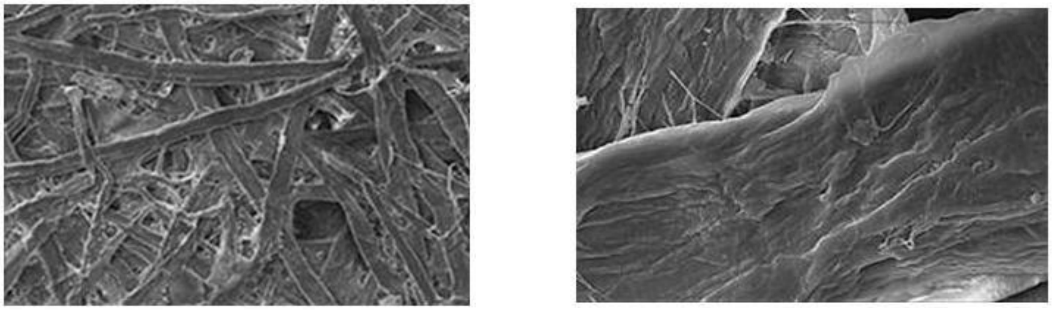
(c) Bacterial cellulose fibers (BCF)
40.00 KX

(d) Bacterial cellulose fibers (BCF)
100.00 KX

Fig. 1. Scanning electron microscope (SEM) images of bacterial cellulose

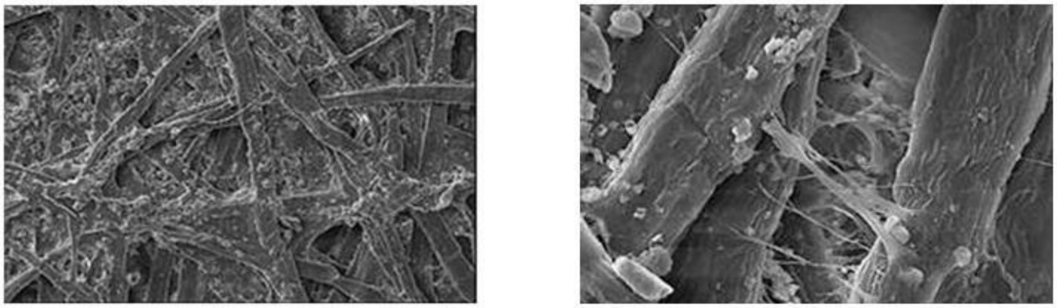
Figure 2 shows the comparative SEM images of the control and 5%, 10%, and 15% BC reinforced office wastepaper samples, magnified at 1000 X and 10000 X. As can be seen in Fig. 2, it was observed that the fiber thicknesses of the wood-based cellulose fibers were quite thick compared to the BC fibers, which were approximately 8 microns to 10 microns in diameter. When the images at 1000 X magnification were examined, it was observed that the gap structure between the fibers decreased and the $CaCO_3$ content increased with the addition of BC. Yuan *et al.* (2016) also stated in their study that with the addition of BC, the fiber-fiber binding capacity increased, the gaps between the fibers became filled, and therefore the paper structure tightened. Bacterial cellulose has a unique three-dimensional network structure (Wu *et al.* 2015). As shown in Fig. 2, the BC attached to the plant fibers to form a compact network during the paper sheet production. The fact that BC fibers combined with the OWP cellulose fibers and filled in the thin gaps between each fiber (as shown in Fig. 2B1, 2C1, and 2D1) due to their thin diameter, suggested that the physical strength properties and barrier properties of resulting paper had been improved.

However, as shown in Fig. 2B2, 2C2, and 2D2, the thin and interwoven network structure of the BC had also trapped the filler pigment materials in the structure of the waste paper and prevented it from being washed away from the fiber matrix during the re-paper production. From the images at 10000 X magnification, it was seen in more detail that the BC, especially in the ratio of 10% and 15%, kept the fillers on the fiber with its three-dimensional network structure.



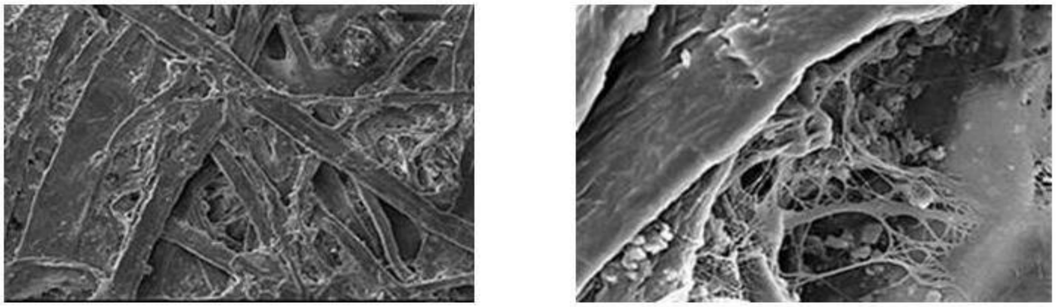
(A1) Office wastepaper fibers
1.00 KX

(A2) Office wastepaper fibers
10.00 KX



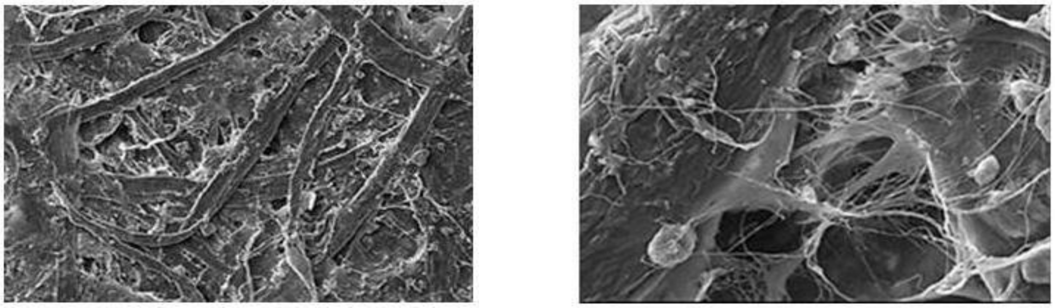
(B1) 95% OWPF + 5% BCF comp.
1.00 KX

(B2) 95% OWPF + 5% BCF comp.
10.00 KX



(C1) 90% OWPF + 10% BCF comp.
1.00 KX

(C2) 90% OWPF + 10% BCF comp.
10.00 KX



(D1) 85% OWPF + 15% BCF comp.
1.00 KX

(D2) 85% OWPF + 15% BCF comp.
10.00 KX

Fig. 2. Scanning electron microscopy images of office wastepaper sheets at 1.00 KX and 10.00 KX magnification without BC (A1 and A2), with 5% BC (B1 and B2), with 10% BC (C1 and C2), and with 15% BC (D1 and D2).

Fourier-transform infrared spectroscopy-attenuated total reflectance analysis

Figure 3 shows the FTIR-ATR spectra of the bacterial cellulose and recycled office wastepaper.

In both spectra, the characteristic peaks of cellulose, apparent at wave numbers of 1023 cm^{-1} and 1151 cm^{-1}, were quite evident. However, the BC spectra showed absorptions signals at 750 cm^{-1}, and 3240 cm^{-1} and stretching bands at 710 cm^{-1} and 3270 cm^{-1}. However, OWP only showed absorption peaks at 710 cm^{-1} and 3270 cm^{-1} bands. Sugiyama *et al.* (1991) reported that signals in the bands at approximately 750 cm^{-1} and 3240 cm^{-1} indicated Iα crystalline cellulose, while signals at approximately 710 cm^{-1}and 3270 cm^{-1} indicated Iβ crystalline cellulose. In this case, the crystal structure of wood-based, cellulose-rich office wastepaper is predominantly Iβ type, while bacterial cellulose contains both Iβ and Iα type polymorphisms. Bacterial cellulose created major peak heights (compared to office paper) in the 1540 cm^{-1} and 1640 cm^{-1} band area. These peaks corresponded to the amide bonds and were related to the proteins and bio residues that originated from the culture medium or bacteria during the production of bacterial cellulose (Dubey *et al*. 2002).

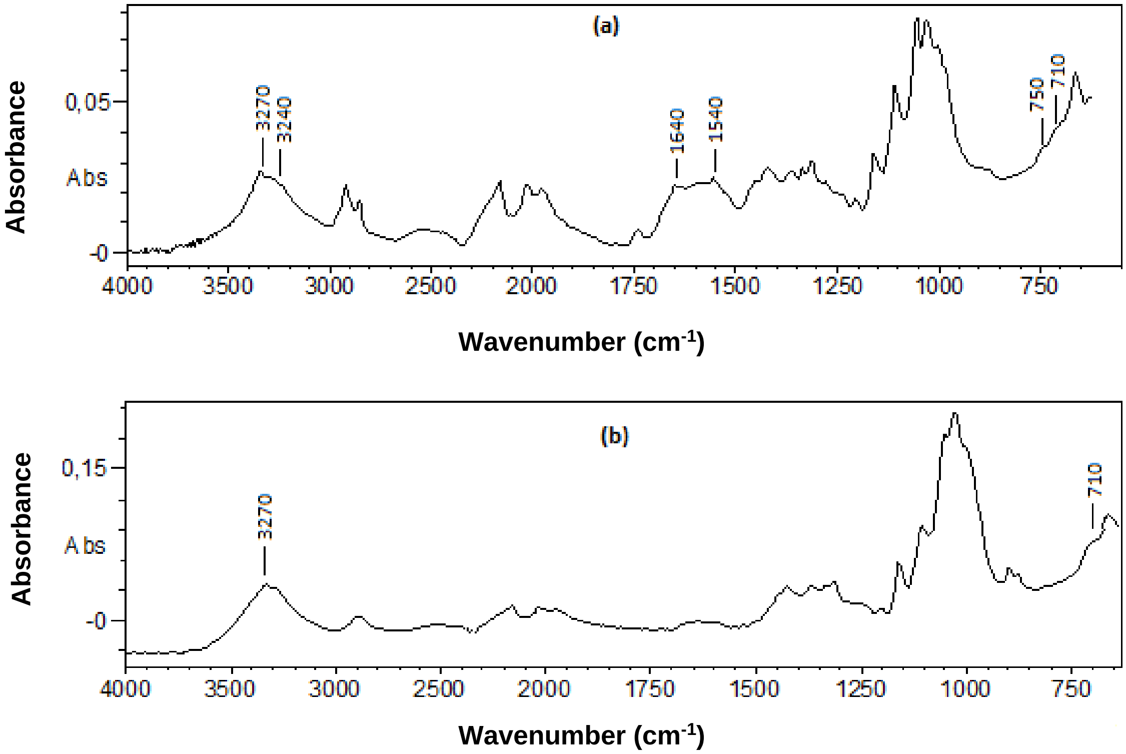


Fig. 3. The FTIR-ATR spectra of the bacterial cellulose (a) and recycled office wastepaper (b)

Figure 4 shows the FTIR-ATR spectra of the recycled office wastepaper and recycled office wastepaper samples reinforced with 5%, 10%, and 15% bacterial cellulose additives. The peak intensity significantly increased, especially at 874 and 1426 cm^{-1}, which was attributed to the amount of bacterial cellulose increasing. This increase at the 874 cm^{-1} and 1426 cm^{-1} band was thought to be caused by the filling material ($CaCO_3$) found in the office paper. Pesman and Tufan (2017) reported that the 874 cm^{-1} and 1426 cm^{-1} bands were associated with calcium carbonate ($CaCO_3$) in their study. It was thought that the BC created an interlaced fibrillar structure between the $CaCO_3$ and cellulose fiber that made up the paper; this structure held and prevented the filler contained in the structure of the waste office paper from being washed away during the paper production process. The band at 1643 cm^{-1} was the absorption peak due to the adsorption of water by the sample (Gao 2011) and the peak at 1652 cm^{-1} was due to water OH bending (Tabarsa *et al.* 2017). These peaks decreased relatively in the 15% BC reinforcement OWP sample. The band at 897 cm^{-1} is assigned to the amorphous region found in cellulose (Poletto *et al.* 2014). As a result of the weakening of the signal from the amorphous regions with the addition of bacterial cellulose with high crystalline properties, a decrease was seen in the height of the peak at the 897 cm^{-1} band.

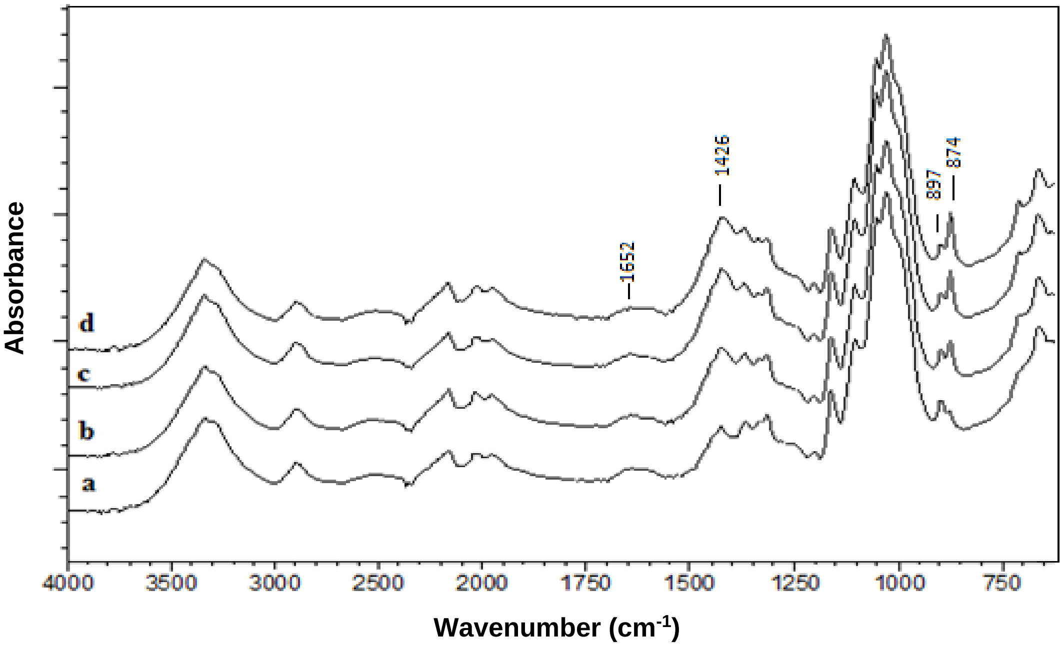


Fig. 4. The FTIR-ATR spectra of recycled office wastepaper (a) and bacterial cellulose (b: 5%, c: 10%, and d: 15%) reinforced recycled office wastepaper

Thermogravimetric analysis (TGA)

The weight loss diagrams of BC, OWP, 5%, 10%, and 15% BC reinforced OWP are shown in Fig. 5, with the results being dependent on the temperature. Weight loss due to the increasing temperature was first observed in the BC sample. This was due to the fact that the internal structure of BC is more compact than the structures of OWP and 5%, 10%, and 15% BC reinforced OWP. Bacterial cellulose differs from plant cellulose due to its high crystalline structure. The fibers that were desorbed physically adsorbed free water *via* evaporation as the heat increased, which is associated with amorphous regions rather than crystalline regions. (Gao *et al.* 2011).

The inflection point temperature was determined to be 333.3 °C for the BC sample and 368.9 ° C for the recycled OWP sample (with a difference of approximately 35 °C between them). The differences in the internal structures of BC and OWP fibers and the evaporation of water were the reasons for this temperature difference. The results indicated that BC was more resistant to high temperatures. The thermal stability was associated with its dense and higher crystalline network structure at the nanoscale (Gao *et al.* 2011).

In addition, the inflection point temperature of the papers obtained by reinforcing recycled OWP with 10% and 15% bacterial cellulose increased from 368.9 °C to an average of approximately 371 °C compared to non-reinforced paper. It is thought that the reason for this increase is that the papers containing bacterial cellulose contained more filling material. The residual amount in office paper, 5%, 10%, and 15% bacterial cellulose reinforced office paper samples at 550 °C were determined to be 23.4%, 24.0%, 26.5%, and 27.2% respectively. This increase in the amount of residue was due to the increase in the amount of inorganic matter contained in bacterial cellulose and the rate of filler trapped in the medium.

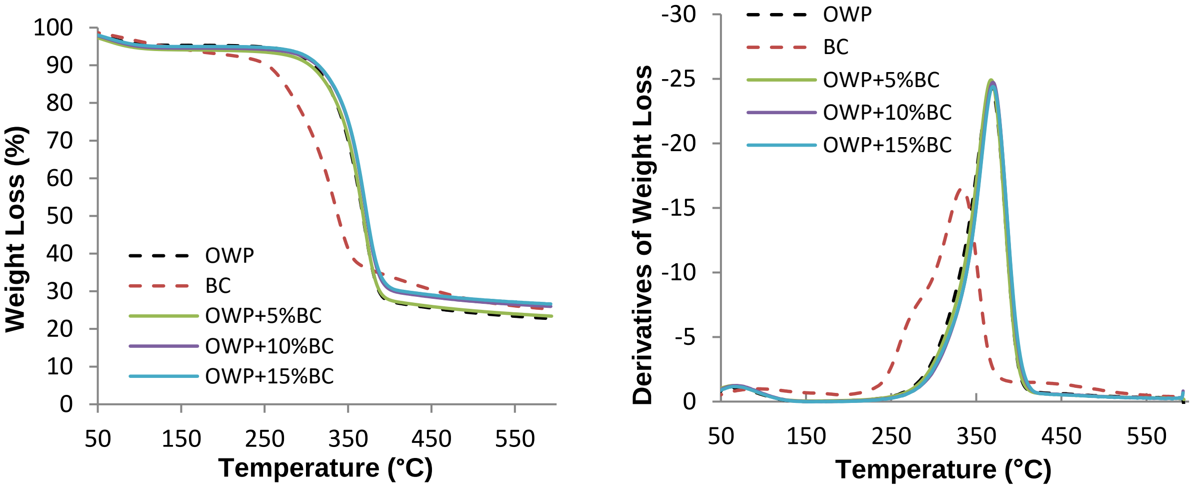


Fig. 5. The TGA and DTGA curves of the bacterial cellulose reinforced recycled office paper samples

Mechanical, Physical, and Optical Properties of Bacterial Cellulose Reinforced Recycled Office Wastepaper

Mechanical and physical properties

Tensile strength has been used to estimate the changes in the inter-fiber bond strength (Koubaa and Koran 1995; Niskanen *et al.* 1999; Forsström *et al.* 2005; Johansonn 2011). The tensile index of the fibers is especially dependent on the binding ability of the fibers in network, regardless of the chemical properties (Ashori *et al.* 2008).

Table 1. Mechanical and Physical Properties of BC Reinforced OWP

	Non-Reinforced OWP		95% OWP + 5% BC		90% OWP + 10% BC		85% OWP + 15% BC	
	Mean value	Std. dev.	Mean value	Std. dev.	Mean value	Std. dev.	Mean value	Std. dev.
Density (kg/m^3)	619.86	4.58	621.40	6.19	626.51	4.74	635.10	6.49
Tensile Index (Nm/g)	30.89	1.41	31.86	2.81	29.90	1.90	31.85	2.13
TEA Index (Nm/g)	0.33	0.04	0.33	0.07	0.30	0.06	0.31	0.04
Tear Index (mN · m^2/g)	34.75	6.24	33.72	5.87	31.13	3.68	29.93	4.45
Burst Index (KPa · m^2/g)	2.28	0.08	2.28	0.08	2.27	0.12	2.22	0.04
Cobb (g/m^2)	100.53	6.50	117.85	6.58	114.64	2.80	112.32	3.31
Air Resist. Gurley (s/100 mL)	5.87	0.15	23.33	0.40	83.03	2.10	113.80	3.52

The effect of BC on the tensile index and TEA (tensile energy absorption) properties are shown in Table 1. The fillers added to office paper, in order to increase their printing properties, had a detrimental effect on paper strength (Brown 1992). Most filler tends to reduce the strength properties of the paper by partially blocking the bonding between the fibers (Hubbe and Gill 2016). Since the fillers prevent fiber-to-fiber bonding, the tensile strength of paper containing fillers are adversely affected (Al-Mehbad 2004). In Table 1, it is seen that the tensile index value is maintained, due to the increasing ratio of BC, despite the filler adhesion. Moreover, with a 5% BC addition, there is a slight increase in tensile strength over the non-reinforced OWP paper. It is believed that this is achieved with the structural properties of BC. Bacterial cellulose has suitable structural properties, *e.g.*, high crystalline, high tensile strength, and wide specific surface area of elasticity (El-Saied *et al.* 2008).

The tear index decreased as the percentage of BC in the paper increased. This is due to the increasingly brittle nature of the paper as bonding is increased. The fracture zone becomes smaller, so that less energy is consumed during the tearing.

Unlike the tear index values, the burst index values stay approximately the same as the BC ratio increases. While this situation is clearly seen in the samples with an addition of 5% and 10% BC, a decrease of only 0.06 units was observed with the addition of 15% BC. All three values remained close to each other as well as to the original burst strength value.

The gaps between the fibers in the paper sheets increased as the BC ratio was increased. This caused the paper density to improve and the tensile, tear and burst indexes to increase (Gao *et al.* 2011; Tabarsa *et al.* 2017; Campano *et al.* 2018). As a result, although the addition of BC to the waste office paper pulp was expected to favorably affect the mechanical strength properties of the paper sheets, it was observed that the tensile index and burst index values were nearly identical but the tear index values had a moderate decrease. This was due to the fact that while the additional BC enhanced the fiber to fiber bonding properties, the $CaCO_3$ filler in the waste office papers also remained within the fiber network structure and had little negative effect on the formation of hydrogen bonds between the fibers.

The determined water absorption capacity for recycled OWP and reinforced recycled OWP with 5%, 10% and 15% BC additives are shown in Table 1. Due to its interlaced microstructure, BC has a high-water binding capacity (Shah and Brown 2005, Skočaj 2019) and excellent water retention abilities (Xiang *et al.* 2017). As shown in Table 1, the water absorption capacity of the recycled OWP generally increased after the BC treatment. However, this increase gradually decreased as the BC percentage used for reinforcement increased. The treatment with BC increased the Cobb values considerably, from 100.2 to 117.8 g/m^2, for the paper reinforced with 5% BC. The water absorption capacity of the recycled OWP paper increased by 17.6% when reinforced with 5% BC. This increase was 14.0% for 10% BC and 11.7% for 15% BC. The increased inter fiber-to-fiber bonding ratio led to decreased porosity in the paper sheets and a reduced water absorption capacity, which could be further increased by an increased BC addition ratio (Gao *et al.* 2011; Tabarsa *et al.* 2017). It was observed that high barrier properties provided for OWP pulp against water, as a result of compatible bonding of bacterial cellulose fibers with wood-based fibers.

Fillat *et al.* (2018) stated that the water absorption capacity of composites produced from bacterial cellulose and paper sheets were decreased, while the barrier properties created against water increased. The properties are among the most important, especially for the food packaging sector, include barrier properties against water and biodegradability, which can be brought to paper by adding BC (Osong *et al.* 2016; Gao *et al.* 2019; Skočaj 2019).

The effect of BC on the porosity of reinforced OWP handsheet samples was determined according to the Gurley air permeability test. Table 1 presented the air resistance of the non-reinforced and reinforced handsheet samples with different ratios of BC to OWP. The obtained results for each reinforced samples showed that the resistance to air permeability drastically increased in all reinforced papers in relation to their original value, depending on the ratio of BC to OWP.

The handsheet samples reinforced with 15% BC exhibited the greatest air resistance values, 19 times greater than the original value. The results were in accord with previous studies. In the study by Yousefi *et al.* (2013), it was found that paper made with BC did not have air permeability. These findings confirmed that increased inter fiber bonding reduces the pore size and consequently leads to an increase in air penetration resistance with a greater addition of BC fibers (Fendler *et al.* 2007; Gao *et al.* 2011; Yousefi *et al.* 2013; Santos *et al.* 2017; Tabarsa *et al.* 2017). In addition, the results indicated that the closed structure of BC partially provided increased barrier properties to the produced paper, so that airflow from the paper surface was partially prevented (Santos *et al.* 2016a; Santos *et al.* 2017). The retention of the filler ($CaCO_3$) was also thought to be effective in terms of reducing air resistance. In a study by Serafica *et al.* (2002), a stronger and more flexible material was produced by immolating the added calcium carbonate to the BC membrane (Serafica *et al.* 2002; Gallegos *et al.* 2016).

Reinforcement with BC is thought to have a protective effect against atmospheric pollutants, which is the primary factor in paper degradation (Area and Cheradame 2011; Yousefi *et al.* 2013; Santos *et al.* 2016a). In addition, considering the importance of the barrier properties of paper used in the food packaging industry and the availability of biomaterials instead of petrochemicals-based products, BC is considered a promising material for this industry (Osong *et al.* 2016).

Optical properties

The optical properties values of the OWP sheets (the control and BC reinforced), before and after the aging process, are shown in Table 2. When cellulose is exposed to a thermal aging process, a common result of the thermal treatment, there is an increase in the yellowness value and a decrease in the brightness value (Ardelean *et al.* 2011). Under normal conditions, the loss of brightness, caused by aging, is greater in samples that are reinforced with BC (Santos *et al.* 2016b). But in the author's study, as shown in Table 2, the brightness values of the test paper sheets with BC reinforcement were stabilized against thermal aging, while the brightness value of the control test paper sheet sample was negatively affected by the thermal aging process. The brightness stability was achieved by trapping the filler contained in the structure of the waste office paper into the interwoven network structure of the bacterial cellulose and thereby preventing it from being washed away from the fiber matrix during paper production. However, as the BC ratio increased, the brightness values decreased. These reductions were approximately 11%, 17%, and 21%, for 5%, 10% and 15% BC ratios, respectively.

The color coordinates, L^*, a^*, and b^*, were also determined and represented in Table 2, where L^* describes the lightness of the color and coordinate a^* and b^*expresses the redness or greenness and yellowness or blueness, respectively. The a-axis runs from red (+a) to green (-a) and the b-axis from yellow (+b) to blue (-) (Hubbe *et al.* 2008). The

*L**, *a**, and *b** values were changed due to the effects of thermal aging, as shown in Table 2. It was seen that the *L** and *b** values of the test papers showed similar changes in the brightness and yellowness, respectively, against the changing conditions. As expected, when the paper sheets were aged, the *L** value decreased and the *b** value increased. Łojewska *et al.* (2007) stated that lignin-free papers can turn yellow during protracted storage due to the conjugated carbonyl derivatives that absorb blue light, which are formed as a result of oxidation of cellulose at high temperatures with the presence of humidity.

Table 2. Optical Properties of the Office Wastepaper Control and BC Reinforced Samples Before and After the Thermal Aging Process

			Non-reinforced OWP		Reinforced OWPs					
					(95% OWP + 5% BCF)		(90% OWP + 10% BCF)		(85% OWP + 15% BCF)	
			Non-aged	Thermal Aged	Non-aged	Thermal Aged	Non-aged	Thermal Aged	Non-aged	Thermal Aged
ISO Brightness (%)		Mean value	88.46	83.82	78.95	78.40	73.28	73.21	69.97	70.56
		Std. dev.	0.30	0.12	0.26	0.23	0.85	0.56	1.12	1,52
Yellowness (%)		Mean value	-9.87	-4.78	0.00	1.02	5.63	6.17	8.25	8.57
		Std. dev	0.46	0.24	0.39	0.27	0.96	0.66	1.11	1.53
CIE Values	L^*	Mean value	92.32	91.84	90.74	90.87	89.61	89.71	88.62	88.94
		Std. dev.	0.13	0.14	0.06	0.06	0.14	0.13	0.24	0.31
	a^*	Mean value	0.49	0.86	0.93	1.45	1.40	1.59	1.65	1.66
		Std. dev.	0.04	0.12	0.02	0.12	0.05	0.06	0.08	0.11
	b^*	Mean value	-4.94	-1.42	-0.16	0.78	2.20	2.66	3.36	3.64
		Std. dev.	0.17	0.13	0.4	0.19	0.45	0.29	0.6	0.77

It was observed from Table 2 that, the yellowness values of the test samples increased due to the increase in the BC reinforcement ratio added to the waste papers. This is thought to be caused by the distinctive yellowish color of bacterial cellulose. While the yellowness values of the test samples increased with thermal aging process, this increase amount decreased due to the increasing bacterial cellulose ratio. With the effect of thermal aging, the yellowness values of control and 5% BC, 10% BC and 15% BC reinforced papers increased by 5.09, 1.02, 0.54 and 0.32 units, respectively. This is attributed to the adhesion of the filling material on the fibers, due to the addition of bacterial cellulose.

As can be seen from Table 2, the *a** values increased depending on the added BC ratio added to the OWP. With the addition of 5% BC, the *a** value increased by 0.44 units compared to the control sample, while this increase doubled with the addition of 10% BC. Only 0.25 units of increase was observed at the *a** values with the addition of 15% BC. The thermal aging process increased the *a** value of the control sample by 0.4 units. The increase seen in *a** value was 0.52 units in waste papers with 5% BC reinforcement, while it was 0.19 units in waste papers with 10% BC reinforcement. The *a** value of 15% BC reinforced waste papers did not change with heat treatment.

The b^* values of all test samples showed similar changes with the yellowness values.

CONCLUSIONS

1. Bacterial cellulose (BC) used for paper reinforcing has the ability to fill the thin gaps between the recycled office wastepaper (OWP) fibers.
2. Bacterial cellulose also has good compatibility in bonding with wood-based fibers to form a compact network during the paper sheet production process.
3. Although BC has the potential to increase the mechanical properties of the papers produced *via* reinforcement, this effect was obscured by the increased retention of fillers, which were trapped within the interwoven network structure of the BC and were prevented from washing away from the fiber matrix of the waste paper during the re-paper production process. While the tensile and burst index values of the papers did not change, the tear index value was reduced, consistent with the effects of the attached filler.
4. As a result of the thermal aging process applied to the BC-reinforced sample test papers, it was observed that the brightness values of the papers did not change, and stability was achieved with the effects of attached filler.
5. Bacterial cellulose will have a promising role in the pulp and paper industry due to its positive results. It was observed that high barrier properties for air and water were achieved with the BC reinforcement of OWP pulp. Due to its high water holding capacity and high barrier properties for water and air, BC is thought to be a potential alternative reinforcement material for the paper industry and can also offer important opportunities for different industries as a resource for producing value-added products with the barrier properties it provides to the paper.

ACKNOWLEDGMENTS

The authors are grateful to Prof. Dr. Hüseyin Kırcı for his contributions to the study and Artvin Coruh University, Science-Technology Application and Research Center for providing the opportunity to work at their laboratories.

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Article submitted: July 21, 2020; Peer review completed: September 5, 2020; Revised version received and accepted: September 14, 2020; Published: September 23, 2020.
DOI: 10.15376/biores.15.4.8496-8514