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Abstract

The aim of this study was to estimate the influence of purification and treatment with textile finishing agent procedures on structural, mechanical, and water barrier properties of bacterial cellulose (BC) in order to predict the end-use properties. Kombucha fungus generated by Komagataeibacter xylinus species, formerly known as Gluconacetobacter xylinus, was used to produce the BC material. The BC was purified with 0.5% sodium hydroxide (NaOH) solution and treated with 5%, 10%, and 20% concentration of N, N-dimethylol 4,5-dihydroxy-ethylene urea (DMDHEU). By Fourier transform spectroscopy (FTIR) and X-ray diffractometer (XRD) was estimated, that the purification with a weak alkali solution was effective to remove amorphous matter of the BC material. Scanning electron microscope (SEM) images demonstrated the BC structure, similar to a non-woven textile fabric with clearly visible three-dimensional networks of fine cellulose fibers. After the purification process, the BC material tensile strength increased by 52%; however, the strain decreased by 93%. BC material after treatment with 20% DMDHEU regained deformability and tensile properties analogous to untreated samples. Water vapor permeability (WVP) values increased and water absorption capacity (WAC) decreased in BC material with increasing DMDHEU concentration. According to the FTIR results, the crosslinking reaction of DMDHEU and adjacent BC molecules was proved. The treatment with DMDHEU restores the amorphous properties of BC material, and therefore blocks water absorption, and the decrease in the water absorption parameter might be determined.

Keywords

Bacterial cellulose, DMDHEU, Komagataeibacter xylinus, mechanical properties, water barrier properties

In recent years, scientists have increasingly focused on biodegradable polymer properties that are particularly attractive in helping to solve the environmental problems of dwindling energy and natural

resources, and aim to improve environmental sustainability. Cellulose is the most abundant natural polymer on earth that can be synthesized by bacteria in natural or standardized media.[1,2] Bacterial cellulose (BC) is used as an environmentally friendly, renewable, and non-toxic material.[3] It has properties including high purity, high crystallinity, high wettability, and excellent mechanical strength, and is used in pharmacy, biomedical science, biobased packaging, paper, food, and the textile industry as a potential cruelty-free leather or to introduce a new paradigm of seamless clothing by 3D formation of BC membrane.[4-6] BC nanofibers are investigated as a structural reinforcement of polymer composites.[7]

No extra purification processing is required for BC, as the material has no lignin, pectin, or hemicellulose.[8] Nevertheless, according to the results of previous studies, the raw material of BC (a byproduct of kombucha drink) is unsuitable for use in the textile industry as it is sticky, is difficult to control, is subject to rapid aging, and wrinkles during washing and drying.[9 13] Purification and finishing technological processes are necessary for BC material to improve in terms of appearance and physical-mechanical properties. Various concentrations from 0.5% to 5% NaOH, potassium hydroxide (KOH), and sodium dodecyl sulfate (SDS) have been used for BC washing.[1,14,15] Alkalization is one of the most common purification processes used to remove the undesired impurities in BC material, such as lignin-like substances, byproducts (sugar, proteins, rod-shaped bacteria, homogeneous melanoidins which impart a yellowish color, nucleic acids), and other organic compounds.[16,17] It was proved that alkaline solution penetrates into the amorphous areas of cellulose and disrupts the crystalline regions by creating new crystalline lattices[18] Purification processes with a higher than 6% alkaline solution cause changes in the BC structure, which has a significant influence on the mechanical properties of this material.[19,20] BC material grows in a liquid medium and forms a gellike material, which needs to be converted into a film during the drying process. In previous studies it was estimated that drying conditions affect the mechanical, structural, and water barrier properties of the BC film.[21]

In this study, BC material was treated with N, N- dimethylol 4,5-dihydroxy-ethylene urea (DMDHEU), which is the most widely used chemical finish for textiles that can be applied for anti-wrinkle finishing and is stable and durable.[22] Some modified versions account for over 85% of all chemicals consumed in the textile industry might be characterized as being able to contain a very reduced formaldehyde content (0.3%).[23,24] The problem with formaldehyde-free linkers is that they require a higher temperature for the reaction and are more expensive.[25] Another disadvantage is that the catalyst used with the carboxylic acid linkers (sodium hypophosphite) can diminish some garment colors. In this study, a formaldehyde-free commercial product, Arkofix NZK, was used, which meets the requirements for children's wear (under 2 years) in: Japan Law 112-1973, Finnish Law SFS 4996/1986, and Eco-Tex Standard 100.[26,27] N-methylol crosslinker forms crosslinks between the cellulose chains in amorphous regions of the fiber, thereby increasing the fiber's elastic recovery from deformation.[28] DMDHEU improves the dimensional stability of cellulose material; moreover, it meets sustainability requirements, which should help to obtain better balance of physical BC properties and reduced environmental impact.[29,30] Jung et al. recognized that DMDHEU had four hydroxyl groups capable of making use of the crosslinking process in cellulose.[31] Due to containing four hydroxyl and the N-methyl groups, DMDHEU is one of the handiest crosslinking chemicals that can crosslink cellulose.[32]

Cellulose can be obtained through a bacterial fermentation process that composes new material in the textile field. Only a few studies have investigated BC processing by textile finishing agents. According to Modolo et al., the potential for chemical finishing to reduce the hydrophilicity of the BC surface with a negative effect from the activity of ureases has been proved.[33] Han et al. analyzed the BC purification process using 3% NaOH solution and a bleaching procedure using 5% hydrogen peroxide (H_2O_2) solution, while Song et al. investigated the effect of wetting agents on BC material without the purification pro- cess.[34,35] The BC material dyeing procedures using *in situ* and *ex situ* methods have

been described, with the positive impact of BC cultivated with direct and basic dyestuffs that have no influence on the crystallinity of the BC material.[36,37]

Summarizing the known research, it can be stated that the interest in BC material is high, but only a few works investigate BC for applications in the field of textiles. Researchers have concentrated on modification of BC material, which grows in synthetic media, but the properties of the BC film grown in the kombu- cha drink medium have not yet been sufficiently studied. Also, the influence of DMDHEU chemical finishing on the properties on kombucha film have not been reported in scientific research. The purpose of this study is to extend the knowledge about the processing of BC material produced from kombucha, and to characterize the effect of alkaline purification and DMDHEU treatment procedures on structural, mechanical, and water barrier properties of BC, in order to predict the end-use physical-mechanical and hygienic properties relevant to the textile industry.

Material and methods

Cultivation

Genomic DNA extraction was performed using the PowerWater®DNA Isolation Kit (MO BIO Laboratories, Inc., Carlsbad, CA, USA) following the manufacturer's instructions.

Identification and confirmation of the genus of the bacteria and yeast in *Kombucha fungus* were based on molecular characterization. The molecular method employed was partial sequencing, using 16S rRNA gene and 18S rRNA gene as the targets for the identification of bacteria and yeast, respectively. Identification was performed by bacterial 16S rRNA gene amplification using 341F/907R primers. Identification of yeast was performed by ITS3/ITS4 primers. The amplified DNA was outsourced to Baseclare Netherlands for sequencing. The sequence of nucleotide alignments obtained was referenced against the NCBI database by the nucleotide BLAST program.

The results illustrated that the bacterium was similar to *Komagataeibacter xylinus* (formerly known as *Gluconacetobacter xylinus)* strains, with 99% identity and 89% query cover. The yeast exhibited 99% identity and 100% query cover to *Zygosaccharomyces bailii.*

For the study, 4g of green tea leaves (pure Ceylon green tea "Impra Royal Elixir Green," Sri Lanka) were infused in 1000 ml boiled water along with 100 g of sucrose (Dan Suker, "Panevezio plius" sugar, Lithuania). After 15 min, tea leaves were removed and cooled to room temperature for about 1-2 h, and 100 ml of 6% yeast extract (apple cider vinegar "Extra Line," Lithuania) was added. The prepared culture medium was incubated with a piece of *Kombucha fungus* (MB "Arbatologija," Lithuania), and fermentation was carried out under standard room conditions (20 ± 2°C temperature and 65 ± 5% relative air humidity) in static cultivation conditions for 7 days (Figure 1(a)). The membrane, grown on the surface of the liquid medium (i.e., a floating, thick, gel-like material) was removed for the treatment and drying procedures.

Purification

In order to eliminate organic impurities, the pieces of BC material were washed under running tap water and purified in a weak alkali solution. The purified BC samples were immersed in 0.5% NaOH (CAS N 1310-73-2, Sigma-Aldrich, Portugal) with a solution ratio of 1:10 and were shaken in the waterbath (Julabo SW22) for 24 h at 30 ± 0.2°C at a speed of 120 rpm. The washing solution was changed after 1, 2, 4, and 8 h.

Treatment

The BC treated (BC_Treated) samples after purification were immersed for 8 h in the application liquor with 100 g/L of DMDHEU (ARKOFIX NZK LIQ M1000, Switzerland) and 15% magnesium chloride ($MgCl_2$) (Sigma-Aldrich) used as catalyst, related to the different concentrations (5%, 10%, and 20%) of DMDHEU.

Drying

After purification and treatment procedures, the BC samples were subjected to convective drying. Wet BC samples were covered with laboratory tissue paper (Joseph paper PRAT DUMAS JPPF-350-800, Spain) of high quality and excellent absorption to remove excess moisture and to avoid additional impurities. All samples were dried on the horizontal silicone surface in the laboratory oven (SNOL 60/300 LFN) at 25 ± 1°C until the specimen gained a constant weight. In previous studies it was noted that washed BC samples become wrinkled, which complicates the investigation of the material properties.[38] Wet samples were pinned along the entire contour during the drying process to ensure a less wrinkled surface. After the regular drying, the BC_Treated samples were dried for 4 min at 150°C to cause the crosslinking reaction.

Mechanical properties

The tensile properties of dried BC samples were investigated using a Tinius Olsen H10 KT tensile test machine. BC untreated (BC_Untreated), BC purified (BC_Purified), and BC_Treated specimens were used for the test. Their lengths were 100 mm, widths were 10 mm, and gauge lengths were 55 mm. A crosshead rate of 100mm/min was set. The load-extension curve was recorded to determine the average value of tensile strength, tensile modulus, and extension at break for at least 10 samples.

Structural properties

The structure of the BC film was analyzed using Fourier transform infrared spectroscopy (FTIR). The spectra were recorded using a Perkin-Elmer Spectrum X II FTIR System spectrophotometer in the range of 600 cm^{-1} to 4000 cm^{-1} with a 4 cm^{-1} resolution.

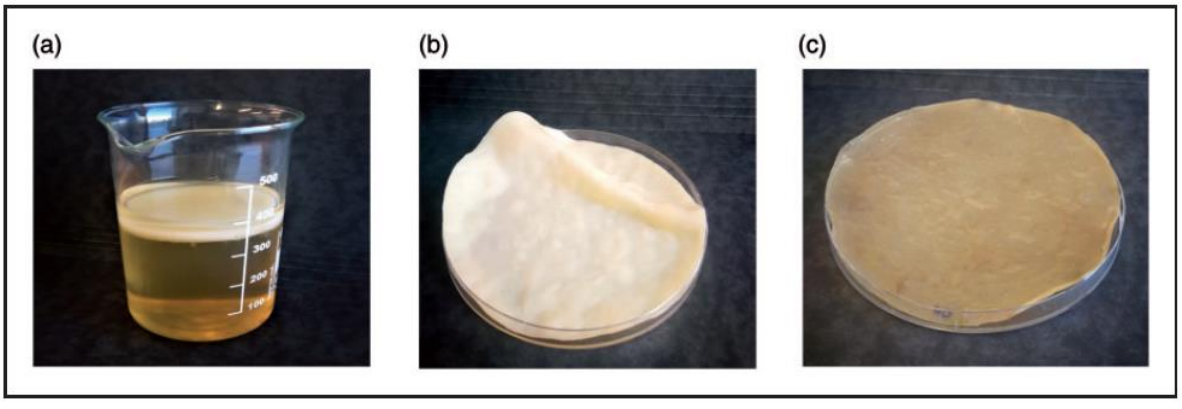

Figure 1. Bacterial cellulose membrane: (a) during the growing process; (b) in the wet state; (c) in the dry state.

A scanning electron microscope (SEM) Quanta 200 FEG at 30 kV was used to investigate the surface morphology of the BC material. The images of dry samples were taken with x 10,000 magnification. All experiments were performed for untreated, purified, and treated BC samples, which were analyzed using the typical values of infrared spectra.[39] The characterization of the crystalline area, amorphous area, and the degree of crystallinity of the BC samples was analyzed with a Bruker SMART X2S X-ray diffractometer (XRD). The crystallinity index (Cr. I., %) of BC samples was calculated from the height of the amorphous (Ia) and the total intensity (I0) according to equation (1):

$$\text{Cr. I.} = (I0\text{-}Ia)/I0 \quad (1)$$

Water barrier properties

The thickness of the BC samples was measured using a DPT 60 digital indicator, with an accuracy of 0.01 mm. The samples were weighed by laboratory precision balance Kern EG-N, with an accuracy of 0.001 g. The average values of the measurements were calculated for investigations and analysis.

Water vapor permeability (WVP) was investigated for the desiccant method according to standard E96/ E96M-10.[40] Before testing, all samples were at standard atmosphere (temperature T = 23 ± 2°C) and relative humidity (RH = 50 ± 5%) conditions. For the desiccant method, silica gel was freshly dried in a ventilated oven at 125 ± 5°C for at least 16 h and was cooled in a sealed container for at least 6 h. Freshly dried silica gel was placed in the pot until half full, then a circular test specimen of BC material (45 ± 5 mm) was placed centrally over the open end of the pot. A clamping ring was placed centrally on the top of the specimen and the ring was tightened. The weight of the pot with the samples was measured. Then all samples were placed in a desiccator with a humidity of 80% for 24 h.
The WVP of the material was calculated by equation (2):

$$WVP = (WVT \cdot T)/\Delta P \quad (2)$$

where WVT is water vapor transmission (g/h-m^2), T is the thickness (mm), and ΔP is vapor pressure difference (mmHg).

The water absorption capacity (WAC) was investigated according to standard 20158.[41] WAC is the weight of absorbed water by a material when it is saturated with water under specified conditions. WAC (%) was calculated for each specimen using equation (3):

$$WAC\ (\%) = ((m_2\text{-}m_1)/m_1) \cdot 100 \quad (3)$$

where m_1 is the weight of the test specimen in the dry state (g) and m_2 is the weight of the test specimen in the wet state (g).

Specimens were weighed to the nearest 0.01 g. The container was filled with distilled water. The samples were immersed in water and left in the conditioned- atmosphere room for 24 h. The samples were removed from the water after 24 h and were drained for 60 s. They were weighed and the WAC parameter was calculated.

Contact angle (CA) was measured using the water drop contact angle method with a Pocketgoniometer PG-3 according to standard 1598 9.[42] Ten measurements were made for each sample with distilled water, and the average values were calculated.

Results

Structural properties

The structure of raw BC material is usually bulky due to the presence of binding agents (mostly sucrose, obtained from the growing medium) and other organic impurities. Purification of BC material with alkaline solution is used to wash off byproducts and organic compounds of the cellulose fiber network. Cellulose inner structure fragments, during the treatment with DMDHEU, are distributed in the matrix and the surface of the cellulose material becomes smooth.

The SEM images of BC_Untreated, BC_Purified, and BC_Treated samples at a magnification of x 10,000 are shown in Figure 2. Figure 2(b) indicates that weak alkaline purification successfully removed the unwanted impurities from the BC material. The structure—similar to a non-woven textile, with clearly visible three-dimensional networks of fine cellulose fibers—was obtained on the SEM image of the BC_Purified sample. Cellulose fibers were determined to have a diameter of 100 nm in previous research.[38] The bulking effect of treatment with DMDHEU is evident in Figure 2(c), showing the BC_Treated sample. Some small particles are found to be attached to the BC surface; it might be that unidentified impurities are not well removed from the surface of the BC material.

FTIR

The FTIR experiment results were used to analyze the change in functional groups within the BC material. The FTIR spectrum of BC_Untreated shows the main peaks at 3339 cm^{-1}, 2896 cm^{-1}, 1730 cm^{-1}, 1426 cm^{-1}, 1236 cm^{-1}, and 1050 cm^{-1} (Figure 3), which are associated with O-H stretching (hydroxyl groups), C-H stretching (methyl groups), O-H bending (carbonyl bonds), and C-O stretching respectively.

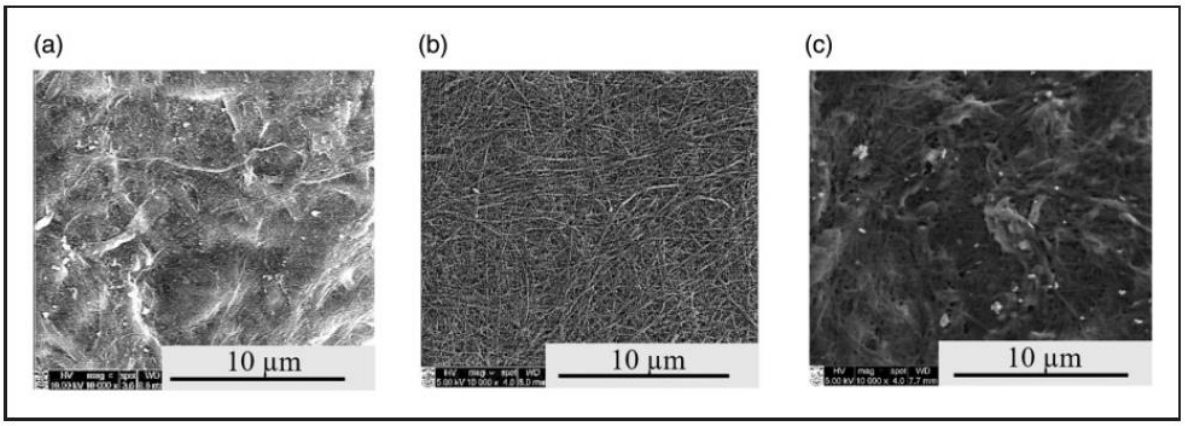

Figure 2. SEM pictures at x 10,000 magnification: (a) BC_Untreated; (b) BC_Purified; (c) BC_Treated.

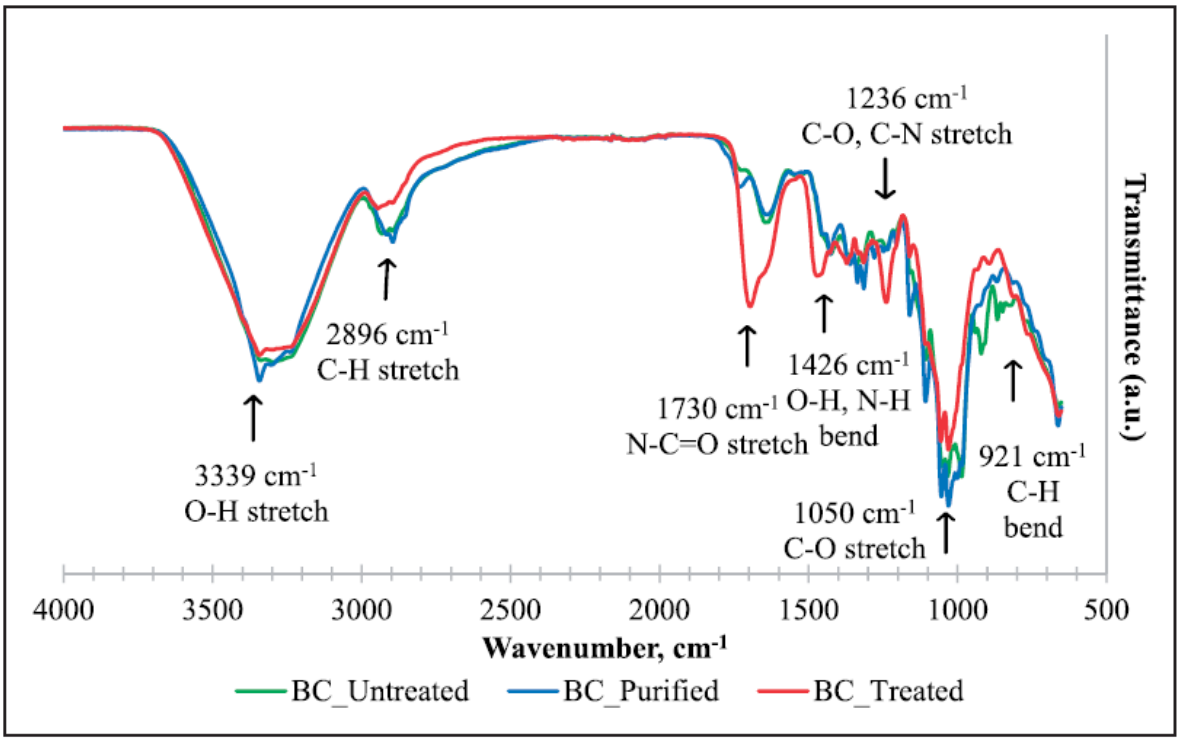

Figure 3. FTIR spectra of BC_Untreated, BC_Purified, and BC_Treated.

In the BC_Purified samples it is seen that there is a significant reduction of specific peaks of sugar (D-glucose, sucrose) that were obtained in the 1000 cm^{-1} to 500 cm^{-1} regions (Figure 3); this shows the removal of sugars in the samples after the purification process.

The infrared spectra of the DMDHEU-treated samples showed slight decreases at 3344 cm^{-1}, 2899 cm^{-1}, and 934 cm^{-1}. The main changes after the treatment process can be seen at three peaks: a significant increase at 1698 cm^{-1}, which is attributed to the appearance of N- C=O groups, a significant increase at 1470 cm^{-1}, which is attributed to the appearance of N-H groups, and a significant increase at 1239 cm^{-1}, which can be attributed to C-O or C-N stretching groups (Figure 3). A strong absorption band at 1698 cm^{-1} indicates an increase in the carbonyl content caused by carbonyl groups in the DMDHEU, and it overlaid native carbonyl group absorptions in cellulose. Two N-methylol groups existing in DMDHEU are capable of reacting with the hydroxyl groups of the cell wall polymer. This peak has shifted to a lower wavenumber (from 1730 cm^{-1} to 1698 cm^{-1}) after the treatment. It might be explained by hydrolysis of the ether groups in the condensed DMDHEU. These three main peaks lead to important changes to the chemical structure of the samples, because DMDHEU was detected in the FTIR curves, and could show that the BC membrane successfully underwent chemical crosslinking with the DMDHEU agents.

XRD

In the XRD analysis, the crystallinity of the BC_Untreated sample was 29% (Table 1), which was considered low because of the high amount of amorphous medium remaining in the sample. The crystallinity of the BC_Purified sample was 1.5 times higher (47%), which proved the effectiveness of the washing procedure when the amorphous matter of the medium was washed off. A reduction in crystallinity was noted after the treatment procedure, falling to 10%. This was expected due to the degradation of the crystalline structure of BC during the treatment, and showed the domination of amorphous areas in the BC after the addition of the DMDHEU.

Table 1. XRD parameters of BC samples

Parameter	BC_Untreated	BC_Purified	BC_Treated
Crystalline area (m^2/g)	9203.55	17,495.85	223.71
Amorphous area (m^2/g)	22,420.28	19,794.98	2098.88
Degree of crystallinity (%)	29.10	46.91	9.63

In other research, the crystallinity of BC was found to range 25% to 92%, and to mostly depend on the growing media (usually it is a synthetic one), static or shaken culture, and the percentage of the alkali solution used (usually 1-3%).[43-45] Moreover, it was noticed that different kinds of chemical applications are used for BC treatment, which reduced the crystallinity of the material as also found in our experiment.[46,47]

The XRD patterns of the BC_Untreated sample showed three diffraction peaks at 14.24°, 16.54°, and 22.92° (Figure 4). The peaks are usually attributed to crystallographic planes of 101 (amorphous region), 10 (amorphous region), and 200 (crystalline region), and characterize cellulose of type I (triclinic). Two peaks in the BC_Purified samples were recorded at 14.68° and 22.47°; these are assigned to the cellulose of 1a and 1b phases. The BC_Treated samples had three peaks at 14.55°, 16.88°, and 22.76°. X-ray diffractograms indicated the amorphous area after the purification was washed off; after the treatment with DMDHEU, the amorphous area increased more than in the BC_Untreated sample.

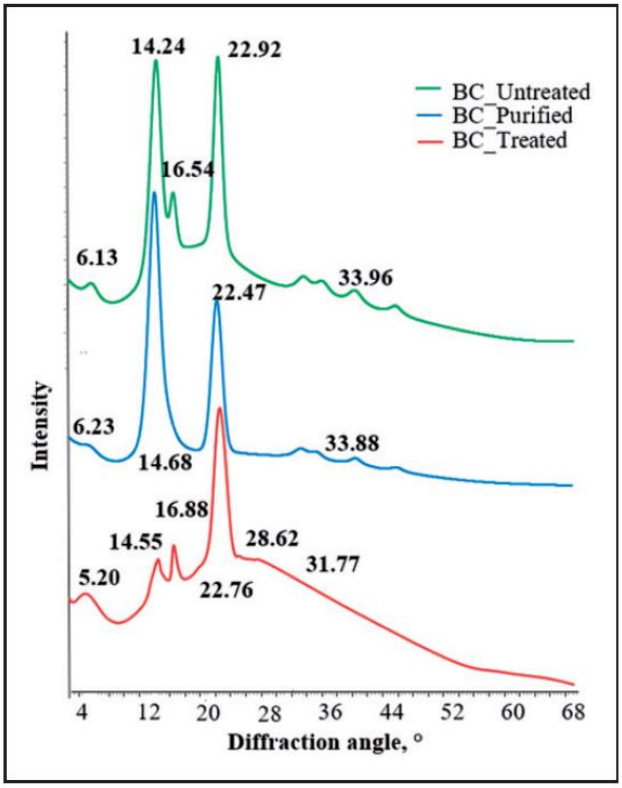

Figure 4. X-Ray diffractograms of BC samples

Mechanical properties

Tensile properties

The purification process of the BC material is one of the most important factors for improving the its tensile strength (Figure 5). The tensile strength of the samples increased by 52% after the purification with NaOH solution (Table 2). Unfortunately, the washed BC material lost its ability to deform and strain decreased by 93%. This could also be related to the removal of the impurities and sugars, or amorphous parts of the cellulose network; BC fibrils obtained better interaction with one another, as we noticed after FTIR analysis.

After treatment, the samples regained their ability to deform significantly. For the samples treated with 5% DMDHEU, the deformation recovered by 85%; for the samples treated with 10% DMDHEU, the deformation recovered by 87%; and for the samples treated with 20% DMDHEU the deformation recovered by 89%. In Kamruzzaman et al.'s study, DMDHEU was blended with MgCl2 as the acid catalyst to improve the formation of crosslinking bonds among the molecules of cellulose chains.32 The development of those bonds in the amorphous areas of the fiber improved the resistance to distortion and enhances the elasticity properties, as we also see in our experiment.

The loss of tensile strength due to the presence of DMDHEU was 4% when treated with 5% DMDHEU, 8% when treated with 10% DMDHEU, and 49% when treated with 20% DMDHEU compare to BC_Purified. According to Chen et al., this phenomenon could be related not only to the DMDHEU appearance, but also to the MgCl2 catalyst used for the crosslinking process.48 The thickness of the BC samples was reduced after removing binders during washing (Table 3); therefore, the material became stiffer and a higher tensile strength was estimated. SEM and FTIR analysis have confirmed that the applied treatment changed the structure of the BC material by causing the crosslinking of cellulose, and the deformation properties of the material were improved.

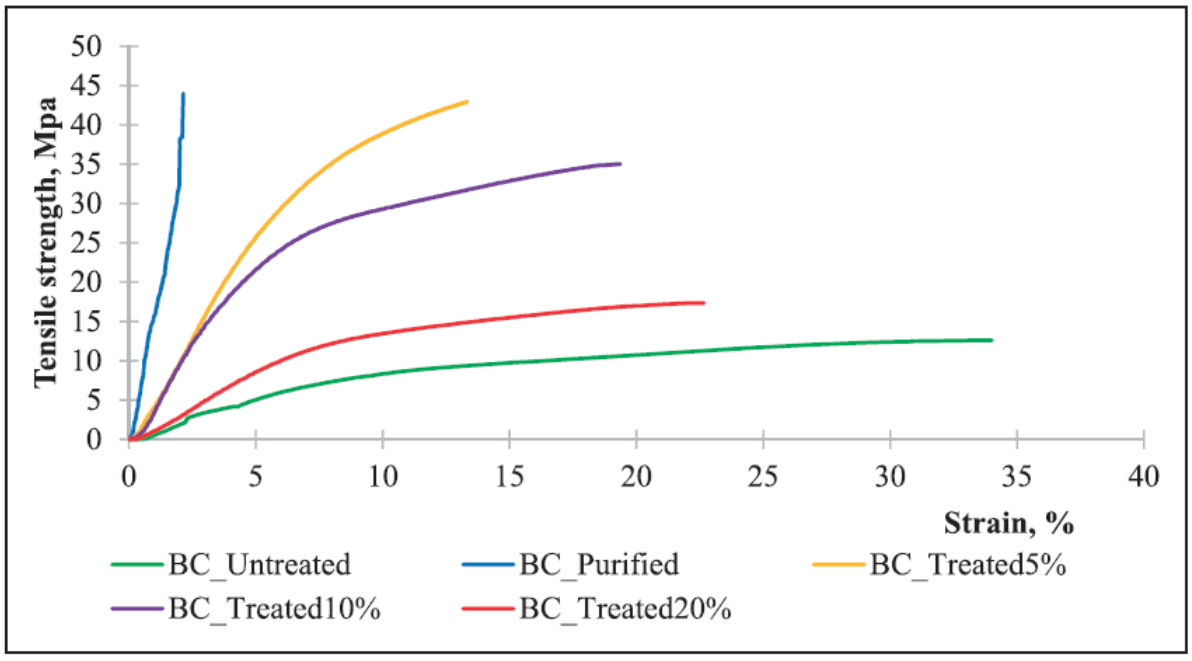

Figure 5. Tensile strength-strain curves for BC samples.

Table 2. Tensile properties of BC samples

Sample	Tensile strength (MPa)	Strain (%)	E modulus (MPa)
BC_Untreated	16.98 ± 0.83	34.62 ± 1.86	0.27
BC_Purified	35.42 ± 7.62	2.50 ± 2.0	17.74
BC_Treated 5%	34.13 ± 3.67	16.52 ± 2.08	3.32
BC_Treated 10%	32.58 ± 2.11	19.21 ± 2.43	3.27
BC_Treated 20%	17.96 ± 3.02	22.92 ± 2.23	1.92

Elwood et al.'s work discussed the possibility of cellulose degrading after the treatment, when DMDHEU forms a complex with the magnesium cation (Mg2+).[49] Since the 15% concentration of the catalyst was the same in all solutions with the DMDHEU, the tensile strength reduced with the increased DMDHEU treatment. It might be explained that DMDHEU molecules can penetrate the cellulose structure, crosslink with the hydroxyl groups in the amorphous region, and thus reduce the slipping of cellulose chains.[50] The same tendency could be identified in Reising et al.'s study, in which it was observed that increasing the alignment and the size of cellulose nanocrystals in the material improves the mechanical properties because of better load transfer between the crystals.[51] The influence of crosslinking was discussed by Lam et al. and Tania et al.[29,52]

Values of E modulus correlate with the results of tensile strength; E modulus drastically increased from 0.27 MPa to 17.74 MPa after the purification process, whereas the E modulus for the BC material treated with DMDHEU significantly decreased—as far as to 1.92 MPa for the BC_Treated 20% sample (Table 2). This indicates a tension behavior close to the BC_Untreated sample, which is characterized by the tensile nature of the elastic material.

Water barrier properties

Thickness

The thickness of the BC sample decreased by 74% after purification (Table 3), when all the impurities of the growing medium were washed out; this was proved by SEM, FTIR, and tensile analysis. The thickness of the sample after applying DMDHEU increases with higher concentrations of the treatment. Adding 5% DMDHEU, the BC material becomes 34% thicker; it increases by 162% after adding 10%; and in the case of 20% DMDHEU it increases by 293%. It is evident that treatment has a significant effect on the mechanical and structural properties of the BC material.

Table. 3. Thickness and water barrier properties of BC samples

Samples	Thickness (mm)	WVP (g·mm/m^2·kPa·h)	WAC (%)	Contact angle (°)
BC_Untreated	0.674	0.049	92.92	63.41
BC_Purified	0.178	0.009	171.23	87.60
BC_Treated 5%	0.238	0.034	89.98	77.40
BC_Treated 10%	0.466	0.070	18.98	68.56
BC_Treated 20%	0.700	0.108	18.08	63.52

WAC, water absorption capacity; WVP, water vapor permeability.

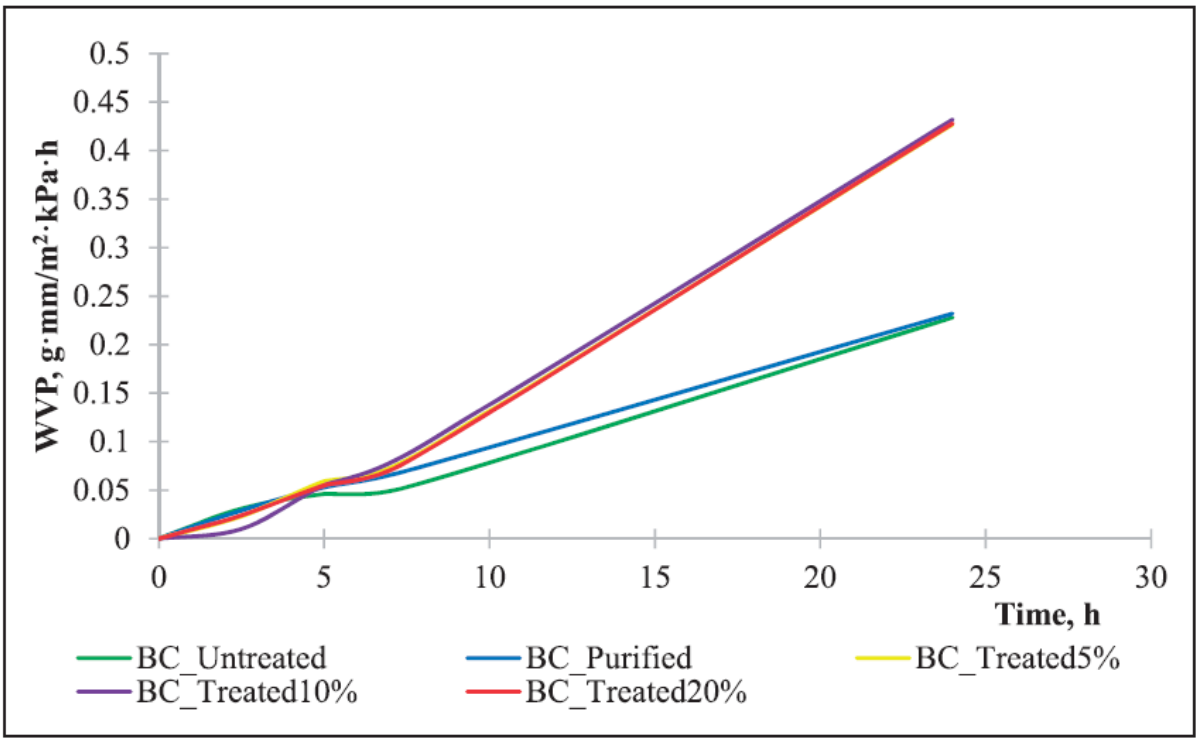

Figure 6. Curves for water vapor permeability (WVP).

The breathability of the material was estimated using the WVP test. The WVP values of the BC_Purified sample decreased by 82% compared to the BC_Untreated sample (Table 3). The value changed from 0.049 gmm/m2kPah for the BC_Untreated sample to 0.009 g mm/m2 kPa h for the BC_Purified sample. Water vapor primarily occurs through the amorphous cellulose regions, so the absence of DMDHEU kept water on the surface for longer. The purification of the samples improved WAC values up to 171%, or about 1.8 times higher than in the untreated samples. The addition of DMDHEU had an influence

on WAC. With a higher concentration of DMDHEU, WAC values reduced from 90% (with 5% DMDHEU) to 18% (with 10% and 20% DMDHEU), which indicates the hydrophobic character of the treated BC material, which may depend on the appearance of the chemical treatment reaction with OH groups in the BC material. The susceptibility of these groups in the amorphous regions within the BC chains are related with the appearance of the functional groups (e.g., C-O, N-H, C-N) containing nitrogen after the treatment with DMDHEU.54 It could be explained that DMDHEU treatment blocks water absorption into the material and the total water absorption is reduced by 90%. As per the obtained results, it might be that DMDHEU treatment introduced hydrophobic groups containing nitrogen to the BC surface; therefore, after the purification process with NaOH, it created hydrophilic hydroxyl and carboxyl groups within the inner structure of the BC.55 Because we used the amorphous regions can reduce WVP. The significant decrease of WVP values after the purification process was shown by the XRD results, where it was determined that the alkaline solution used to purify the BC samples removed the amorphous parts and left the crystalline segments intact. WVP values rise with increased DMDHEU treatment concentrations, becoming twice as high with 20% DMDHEU than it was before the purification process (Figure 6). Water molecules can simply diffuse to H-bonds with OH groups in the polymer chains and can create new bonds with the DMDHEU matrix.53 According to the FTIR and XRD results, it was proved that the addition of DMDHEU restores the amorphous properties of the BC sample, and therefore it has an influence on the increase in WVP values.

The wettability of the BC material surface was evaluated through the measurement of the CA of water (Table 3). We can see that there was not a big difference in CA values, ranging from 63° to 87°. These results were measured in the first seconds, but after a while it changed and samples washed with NaOH absorbed all the water while samples treated with DMDHEU kept water on the surface for longer. The purification of the samples improved WAC values up to 171%, or about 1.8 times higher than in the untreated samples. The addition of DMDHEU had an influence on WAC. With a higher concentration of DMDHEU, WAC values reduced from 90% (with 5% DMDHEU) to 18% (with 10% and 20% DMDHEU), which indicates the hydrophobic character of the treated BC material, which may depend on the appearance of the chemical treatment reaction with OH groups in the BC material. The susceptibility of these groups in the amorphous regions within the BC chains are related with the appearance of the functional groups (e.g., C-O, N-H, C-N) containing nitrogen after the treatment with DMDHEU.[54] It could be explained that DMDHEU treatment blocks water absorption into the material and the total water absorption is reduced by 90%. As per the obtained results, it might be that DMDHEU treatment introduced hydrophobic groups containing nitrogen to the BC surface; therefore, after the purification process with NaOH, it created hydrophilic hydroxyl and carboxyl groups within the inner structure of the BC.[55] Because we used the anti-wrinkle product DMDHEU, which according to Vahid et al. reacts with the OH groups of cellulose,[56] the number of free OH groups (which are hydrophilic) should decrease following treatment with DMDHEU. The hydrophobicity should decrease due to a decrease in unregulated hydrophilic OH groups, and this tendency we can see in our experiment with reference to the WAC results.

Conclusion

Structural, mechanical, and water barrier properties of BC untreated, purified with NaOH, and treated with DMDHEU material samples were examined. SEM analysis clearly indicated the effectiveness of the weak alkali solution purification process and obtained clear images of the cellulose network structure that can be compared to a non-woven textile. The purification process was also proved regarding morphology by obtaining more crystalline areas in comparison to the untreated and treated samples. Subsequently, the purification and the tensile strength of the samples increased by half, and the material lost its ability to deform by 93%.

The main structural changes of the BC material after the DMDHEU treatment process were determined by FTIR and XRD analysis. Three increased peaks at 1698 cm^{-1}, 1470 cm^{-1}, and 1239 cm^{-1} showed that cellulose was successfully crosslinked with the agents of DMDHEU and the slipping of cellulose chains was reduced. Along with the mechanical properties, the samples regained their ability to deform after the treatment. The concentration of DMDHEU treatment also affected the water barrier properties. The significant decrease of WVP values after the purification process might be related to the removed amorphous parts of the BC material. After the addition of DMDHEU, the WVP values rose with increased treatment concentrations. According to the WAC results, DMDHEU treatment blocks water absorption into the material by 90% by introducing hydrophobic groups within the inner structure of the BC.

The results showed that BC material obtained through a natural fermentation process might be treated by conventional textile finishing methods. However, the properties of BC need to be further investigated to find the best combination of properties relevant to end-use product production and exploitation that leads to sustainable implications for the process of developing BC.

Declaration of conflicting interests
The authors declared no potential conflicts of interest with respect to the research, authorship, and/or publication of this article.

Funding
The authors received no financial support for the research, authorship, and/or publication of this article.

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