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Effect of Hair Particle Filler on the Characteristics of Green Composite Based on Bacterial Cellulose

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Abstract. *The growing demand for sustainable materials and effective waste management has encouraged the development of environmentally friendly composites. Green composites are a promising alternative to conventional materials for environmental preservation. In this study, bacterial cellulose derived from nata de coco was used as a matrix, while haircut waste particles served as a filler. Composites were fabricated via compression molding at 170 °C with filler contents of 0, 15, 25, 35, and 45 wt.%. The synthesized materials were characterized through tensile testing, scanning electron microscopy (SEM), and Fourier transform infrared (FTIR) spectroscopy. Increasing hair-particle content increased the elastic modulus from 44.11 ± 3.02 MPa (A1) to 130.46 ± 4.94 MPa (A2), indicating enhanced stiffness of the composite; however, further increases in filler content slightly reduced the modulus due to possible void formation and weaker interfacial adhesion. In contrast, the tensile strength decreased progressively from 11.54 ± 0.56 MPa (A1) to 5.16 ± 0.52 MPa (A5) with increasing filler content. SEM observations revealed the formation of voids and weaker matrix–filler interactions at higher filler contents, which contributed to the reduction in tensile strength. FTIR spectra showed the presence of O–H, C–H, and C=O functional groups, suggesting possible interactions between bacterial cellulose and hair particles. Overall, a filler content of 15 wt.% provides the best balance between stiffness and structural integrity, demonstrating the potential of hair waste as a sustainable reinforcement in bacterial cellulose-based green composites.*

Keywords: *Green Composite; Tensile Strength; Elastic Modulus; Internal Morphology; Functional Groups.*

Type of the Paper: Regular Article.

1. Introduction

Environmental sustainability has become an increasingly critical issue in the modern era, driven by the growing use of synthetic materials that pose negative environmental impacts and the decreasing availability of natural raw materials. Therefore, the use of environmentally unfriendly materials needs to be limited and replaced with more sustainable alternatives. One promising alternative is composite materials, which are structures composed of two or more materials with different properties that are combined to produce superior characteristics, such as high specific strength, good fatigue resistance, and resistance to extreme conditions [1]. Composite materials made from natural and biodegradable constituents are known as green composites or environmentally friendly composites. Green composites are designed to minimize environmental

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impacts throughout their life cycle, aligning well with the principles of environmental conservation and sustainable development [2].

Composite materials have been widely used as raw materials in various industrial sectors, including automotive, furniture, household appliances, medical applications, and sports equipment [1]. In line with the growing emphasis on sustainability, the development of composite materials should focus on utilizing environmentally friendly resources that are abundant yet underutilized. One promising alternative is bacterial cellulose derived from *nata de coco*, which has traditionally been used primarily as a food product, as well as from human hair cutting waste, often discarded and underutilized. The incorporation of these materials is expected to support the development of sustainable and value-added green composites.

Nata de coco is a product obtained from the fermentation of coconut water by the bacterium *Komagataeibacter xylinus*, which metabolizes sugars into a three-dimensional gel-like structure of bacterial cellulose with a high degree of purity [3]. Bacterial cellulose derived from *nata de coco* exhibits good crystallinity and superior mechanical properties, making it highly promising for application as a composite material [4]. In general, bacterial cellulose demonstrates a crystallinity index of approximately 84–89%, a high degree of polymerization, a large specific surface area with fiber diameters ranging from 20 to 100 nm, as well as high flexibility and tensile strength, with a Young's modulus reaching 15–18 GPa [5]. Over the past few decades, research on bacterial cellulose has increased significantly, particularly in relation to its development into nanocomposites through incorporation with polymers, nanoparticles, and biomolecules [6].

Despite its excellent mechanical properties and strong potential as a composite constituent, bacterial cellulose derived from *nata de coco* possesses a highly hydrophilic structure due to the abundance of hydroxyl functional groups. This hydrophilic nature results in a high water absorption capacity, causing bacterial cellulose to absorb excessive moisture, which may limit its performance in structural composite applications [7]. Therefore, incorporating relatively hydrophobic components, such as human hair fibers, may help reduce moisture sensitivity and improve the dimensional stability of bacterial cellulose-based composites [8]. In addition, the presence of such reinforcement may also influence the mechanical properties of the resulting composite.

Human hair is a natural keratin-based fiber with a high protein content (approximately 65–95%) and a densely packed cortical structure, which contributes to its favorable mechanical properties. Human hair typically has a diameter of about 50–80 µm, a density of approximately 1.32 g/cm³, tensile strength in the range of 150–350 MPa, and elongation at break of up to 216.9% [9]. Previous studies have demonstrated that human hair can serve as a reinforcement material in polymer composites, improving properties such as tensile strength, flexural strength, impact resistance, and hardness at appropriate reinforcement fractions [10]. In addition, the relatively hydrophobic nature of human hair compared to cellulose-based materials suggests the potential to reduce moisture sensitivity. Therefore, the utilization of human hair as a filler not only improves composite performance but also offers a sustainable solution for managing hair waste, which is difficult to degrade and may contribute to environmental pollution [11]. However, most previous studies have focused on the use of human hair in fiber form within polymer-based composite matrices. The utilization of human hair in particulate form as a filler in bacterial cellulose-based composites remains limited. Moreover, the influence of hair-particle content on the mechanical behavior and microstructure of bacterial cellulose composites has not been systematically investigated. Therefore, exploring the incorporation of human hair particles into bacterial cellulose matrices is important to better understand their influence on the mechanical performance and microstructure of green composites.

The performance of composite materials depends not only on the properties of the individual components but also on the quality of the interfacial interaction between the matrix and the reinforcement. Strong interfacial bonding enables efficient stress transfer, whereas weak interfacial adhesion can lead to a significant deterioration of composite mechanical properties [12]. Therefore, to achieve optimal interfacial bonding in this study, an additive was introduced during the composite synthesis process. One environmentally friendly additive that has been widely used is citric acid. Citric acid is a weak organic acid containing three carboxyl groups; it is biodegradable and non-toxic, making it suitable for use as a binding or crosslinking agent in natural polymer–based materials [13]. Citric acid can form ester bonds between its carboxyl groups and the hydroxyl groups present in both the matrix and the reinforcement, thereby enhancing interfacial adhesion, improving mechanical properties, and reducing water absorption and swelling behavior in composite materials [14].

Based on the potential of bacterial cellulose as a matrix, human hair cutting waste in particulate form as a filler, and citric acid as an environmentally friendly binding agent, this study investigates the development of bacterial cellulose-based green composites reinforced with human hair particles. This research aims to evaluate the effect of hair-particle content on the mechanical properties and microstructural characteristics of the resulting composites. Mechanical performance was evaluated through tensile testing to determine the ultimate tensile strength (UTS) and elastic modulus. The internal morphology and the distribution of fillers in the matrix were also investigated using scanning electron microscopy (SEM), while Fourier transform infrared (FTIR) spectroscopy was employed to identify the functional groups present in the synthesized composite materials.

2. Materials and methods

2.1. Materials and Instruments

The equipment used in this study included a Hung Ta tensile testing machine (HT-2402, 10 kN capacity), a blender, a hot press machine, a cold press machine, and a 200-mesh sieve. The materials used in this research included bacterial cellulose derived from nata de coco, human hair-cutting waste particles, citric acid, and sodium hydroxide (NaOH). Nata de coco used in this study was obtained from a local producer in the Jember area, while the human hair cutting waste was collected from local hair salons in the Jember area. The bacterial cellulose was oven-dried at 100 °C for approximately 2 h and subsequently ground using a blender to obtain cellulose powder before use. The human hair particles used in this study had a particle size below approximately 74 µm, corresponding to the 200-mesh sieve used for particle size selection.

2.2. Method and Procedure

The synthesis of the green composite began with the preparation of the composite constituents, namely nata de coco and human hair cutting waste. The nata de coco was processed to obtain bacterial cellulose powder by crushing and oven-drying it at 100 °C for approximately 2 h to reduce its moisture content. Meanwhile, the human hair cutting waste was cleaned and treated with a 5% NaOH solution for 1 h to remove surface impurities, followed by drying under sunlight. The dried alkalized hair was then ground using a blender and sieved through a 200-mesh sieve to obtain hair particles with sizes below approximately 74 µm.

The amounts of bacterial cellulose powder and hair particles were determined based on their mass fractions, as listed in Table 1. Citric acid was added at 5 wt% of the total mass of the composite constituents, with the total composite mass fixed at 8 g. In this study, citric acid acted as a crosslinking agent that promotes esterification reactions between the hydroxyl groups of cellulose and the functional groups of keratin under elevated temperature conditions.

All composite constituents were mixed using a blender to obtain a homogeneous mixture. The mixture was then placed into a mold with dimensions of 10 × 1 × 0.6 cm and compacted using a cold press. Subsequently, the specimen was subjected to hot pressing at a temperature of 170 °C under a pressure of 5 MPa using a hydraulically assisted hot press. The temperature was maintained at 170 °C for 5 min as the dwell time during the pressing process. After hot pressing, the composite specimen was allowed to cool under pressure before being removed from the mold. The synthesized composite specimens were then subjected to further characterization.

Table 1. Mass fraction variations of hair particle filler and bacterial cellulose in green composites

Type of Sample	Mass Fraction of Filler: Bacterial Cellulose (% total mass of composite)
A1	0 : 100
A2	15 : 85
A3	25 : 75
A4	35 : 65
A5	45 : 55

2.3. Tensile testing

Tensile testing of the green composite materials was conducted using a Computerized Control Universal Testing Machine (UTM) Hung Ta HT-2402 with a maximum load capacity of 10 kN. The composite specimens were prepared in rectangular shapes with dimensions of approximately 10 × 1 × 0.6 cm. Before testing, the cross-sectional area and gauge length were measured and used as input parameters for the computer-controlled tensile testing system.

The tensile test was performed at a crosshead speed of 4.0 mm/min with a gauge length of 50 mm. During the test, the applied force and elongation were recorded and converted into stress (σ) and strain (ε) values to obtain stress–strain curves. The tensile strength was determined from the maximum stress before specimen failure, while the elastic modulus was calculated from the slope of the initial linear region of the stress–strain curve. Each composite formulation was tested three times ($n = 3$), and the results were reported as the average value of the measurements. The stress–strain data and elastic modulus calculations were processed using Microsoft Excel.

2.4. Morphology analysis (SEM)

The internal morphology of the green composite materials was examined using a scanning electron microscope (SEM, Hitachi Type 30). Fractured cross-sectional specimens obtained after tensile testing were used to observe the internal structure and fracture morphology of the composites. The fractured surfaces were directly mounted on the SEM sample holder and observed without additional surface treatment. SEM observations were carried out at an accelerating voltage of 10 kV, and micrographs were recorded at a magnification of 200×. The SEM images were used to evaluate the internal morphology of the composites, particularly the presence and distribution of voids within the composite structure. Void size estimation was performed based on the SEM scale bar using image measurement to provide a semi-quantitative description of the observed pores.

2.5. FTIR spectroscopy

Fourier Transform Infrared (FTIR) analysis was performed to identify the functional groups present in the synthesized green composite materials using an Alpha FTIR spectrometer. The infrared spectra were recorded in the wavenumber range of 4000–400 cm^{-1} using the attenuated total reflectance (ATR) mode. To obtain a better signal-to-noise ratio, each spectrum was measured at a resolution of 4 cm^{-1} with a total of 32 scans. The collected spectra were processed using baseline correction before analysis. The functional groups in the composite materials were identified based on the position and intensity of the absorption peaks expressed as percentage transmittance. Particular attention was given to absorption bands associated with cellulose, keratin from hair particles, and possible ester bonds formed due to the presence of citric acid.

3. Results and Discussion

The characteristics of the synthesized green composite were determined through tensile testing, SEM analysis, and FTIR analysis. Tensile testing produced stress and strain data, which were subsequently plotted as stress–strain curves to analyze the tensile strength and elastic modulus of the composite materials. The resulting tensile strength and elastic modulus values are presented in Table 2 and Fig. 1.

Table 2. Tensile properties of green composites with varying filler–matrix ratios

Sample	Tensile Strength (MPa)	Elastic Modulus (MPa)
A1	11.54 ± 0.56	44.11 ± 3.02
A2	9.64 ± 1.08	130.46 ± 4.94
A3	7.69 ± 0.74	129.85 ± 4.12
A4	5.76 ± 1.05	93.05 ± 8.70
A5	5.16 ± 0.52	98.76 ± 2.02

Table 2 presents the tensile strength and elastic modulus values of the synthesized composite materials. Based on these data, the tensile strength of the composites tends to decrease with increasing hair particle filler fraction. This behavior may be related to the limited interaction between the bacterial cellulose matrix and the hair particle filler, which limits the ability of the matrix to transfer stress to the filler phase. Weak interfacial adhesion limits effective stress transfer from the matrix to the filler, thereby reducing the ability of the filler to carry and distribute the applied load within the composite material [12].

In addition to interfacial interactions, the tensile strength of composite materials is also influenced by the distribution of the filler within the matrix. Non-uniform filler dispersion can lead to particle agglomeration accompanied by the formation of voids that act as stress concentration regions, leading to a faster failure of the material. In contrast, homogeneous filler dispersion enhances the structural integrity of the composite and reduces internal defects [15]. Therefore, the decrease in tensile strength observed in this study may also be associated with non-uniform filler dispersion and the potential formation of voids at higher filler contents.

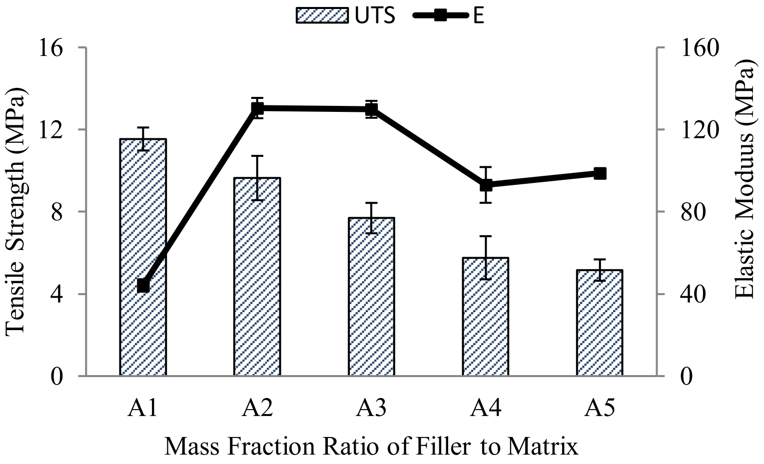

Fig. 1. Tensile Properties of Composites with Varying Hair Filler Content

Fig. 1 illustrates the trend of tensile strength (UTS) and elastic modulus as a function of the filler-to-matrix mass fraction. As shown in the figure, increasing the mass fraction of hair particle filler tends to reduce the tensile strength of the composite material. The lowest tensile strength was obtained for the composite with a filler-to-matrix mass fraction of 45%:55% (sample A5), with a value of 5.16 ± 0.52 MPa. This decrease may be associated with reduced load transfer efficiency between the bacterial cellulose matrix and the hair particle filler at higher filler contents. Similar behavior was reported by Pangestu et al. [16] who observed that the tensile strength and impact strength of mask-waste-based composites decreased with increasing bamboo fiber content.

Nevertheless, the tensile strength of the green composite obtained in this study remains relatively high and comparable to several previous studies. For instance, Bale et al. [17] reported that the highest tensile strength of a polyester-based composite reinforced with nylon fibers was achieved at a fiber volume fraction of 60%, with a tensile strength of 7.07 MPa. This comparison indicates that the use of bacterial cellulose as a matrix contributes positively to the mechanical performance of the composite, although the effect of higher filler fractions on tensile strength still requires further investigation. Therefore, controlling the filler fraction, improving mixture homogeneity, and enhancing interfacial bonding through the use of suitable binding agents are important aspects to be addressed in future studies.

In addition to tensile strength, the elastic modulus also varied with changes in filler fraction, reflecting the influence of the filler on the stiffness of the composite material. An increase in elastic modulus at certain filler fractions indicates enhanced material stiffness, whereas a decrease at higher filler contents suggests reduced filler effectiveness in reinforcing the matrix. This behavior can be attributed to limitations in interfacial bonding and non-uniform filler distribution. Based on the results presented in Fig. 1, the elastic modulus increased with filler addition up to a mass fraction of 25%. However, similar elastic modulus values were observed at filler mass fractions of

15% and 25%, as well as between those at 35% and 45%. This trend may be related to non-uniform filler dispersion and possible void formation within the composite structure, which can influence the mechanical response of the material. Nevertheless, these results indicate that the incorporation of hair particle filler is still effective in enhancing the elastic modulus compared to the composite without filler (100% matrix, sample A1).

The elastic modulus of the composite with a filler-to-matrix mass fraction of 0%:100% (sample A1) was 44.11 ± 3.02 MPa and increased significantly to 130.46 ± 4.94 MPa after the addition of 15% filler. For other filler mass fraction variations, the elastic modulus values were also higher than those of the composite composed of 100% bacterial cellulose matrix, indicating that the incorporation of hair particle filler contributes to an increase in composite stiffness. This result is consistent with the findings of Pangestu et al. [16], who reported that the addition of 8% bamboo fiber increased the elastic modulus of mask-waste-based composites from 129.33 MPa to 246.00 MPa. This similar trend suggests that the presence of filler acts as a reinforcing phase that restricts matrix deformation, thereby increasing the stiffness of the composite material.

The internal morphology of bacterial cellulose–based composites reinforced with hair particle filler was examined to evaluate the internal structure by observing the fracture surfaces of the composite specimens after tensile testing. Three composite samples were selected for this analysis: a composite consisting of 100% bacterial cellulose matrix (sample A1), the composite exhibiting the highest tensile strength at a filler-to-matrix mass fraction of 15%:85% (sample A2), and the composite with the lowest tensile strength at a filler-to-matrix mass fraction of 45%:55% (sample A5). The morphological analysis produced two-dimensional images that represent the internal morphology of the composite materials at the microscale.

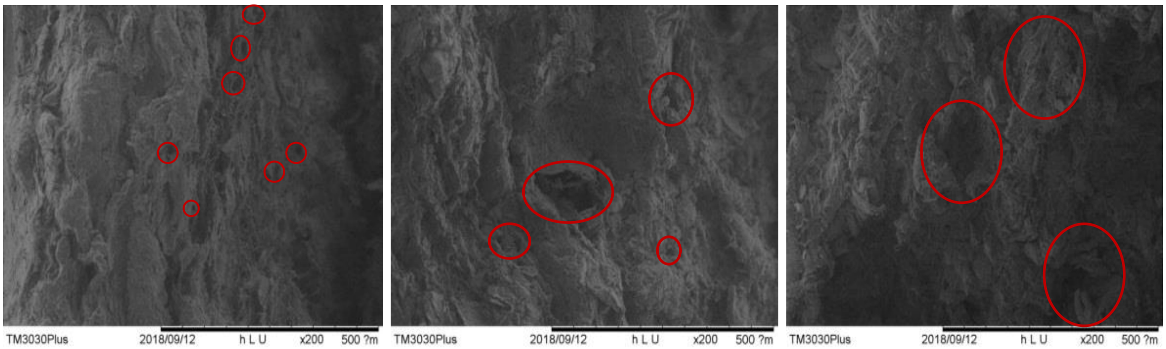

Fig. 2. SEM images showing the internal morphology of composites at filler-to-matrix mass fractions of (a) 0%:100%, (b) 15%:85%, and (c) 45%:55% (200× magnification).

Fig. 2 presents the SEM micrographs of the fractured surfaces of the green composites observed at a magnification of 200× with a scale bar of 500 μm. Fig. 2a shows the morphology of the composite composed of 100% bacterial cellulose matrix. The microstructure appears relatively compact with only a few small voids, with estimated diameters generally below approximately 50–100 μm, indicating a fairly homogeneous internal structure of the matrix. This morphological condition is consistent with the tensile test results, in which the composite with 100% bacterial cellulose matrix exhibits the highest tensile strength of 11.54 ± 0.56 MPa.

In contrast, the composite containing 15% hair particle filler (Fig. 2b) exhibits a higher number of voids with larger estimated diameters ranging approximately from 100 to 200 µm, which indicates the presence of local heterogeneities in the composite structure. These voids, highlighted by the red circles in the micrograph, can act as stress concentration sites that initiate microcracks during tensile loading, thereby contributing to the observed reduction in tensile strength.

A more pronounced morphological change is observed in the composite containing 45% filler (Fig. 2c). The SEM micrograph shows a greater number of voids with larger sizes, some of which exceed approximately 200 µm, suggesting a less compact internal structure at higher filler content. The presence of these larger voids indicates increased internal defects and potential discontinuities at the matrix–filler interface, which can reduce the efficiency of stress transfer between the bacterial cellulose matrix and the hair particle filler.

In natural fiber–reinforced composites, void formation and non-uniform filler dispersion are commonly reported to reduce mechanical performance because they interrupt the load transfer path across the matrix–filler interface [18]. Therefore, the morphological observations in Fig. 2 support the tensile test results, where increasing filler content is associated with a reduction in tensile strength.

The functional groups in the composite materials were analyzed using FTIR spectroscopy to understand the influence of hair particle filler addition on the chemical characteristics of the composites. The results of this analysis are presented as FTIR spectra, as shown in Fig. 3. The spectra were then qualitatively analyzed to identify the functional groups present in each composite sample.

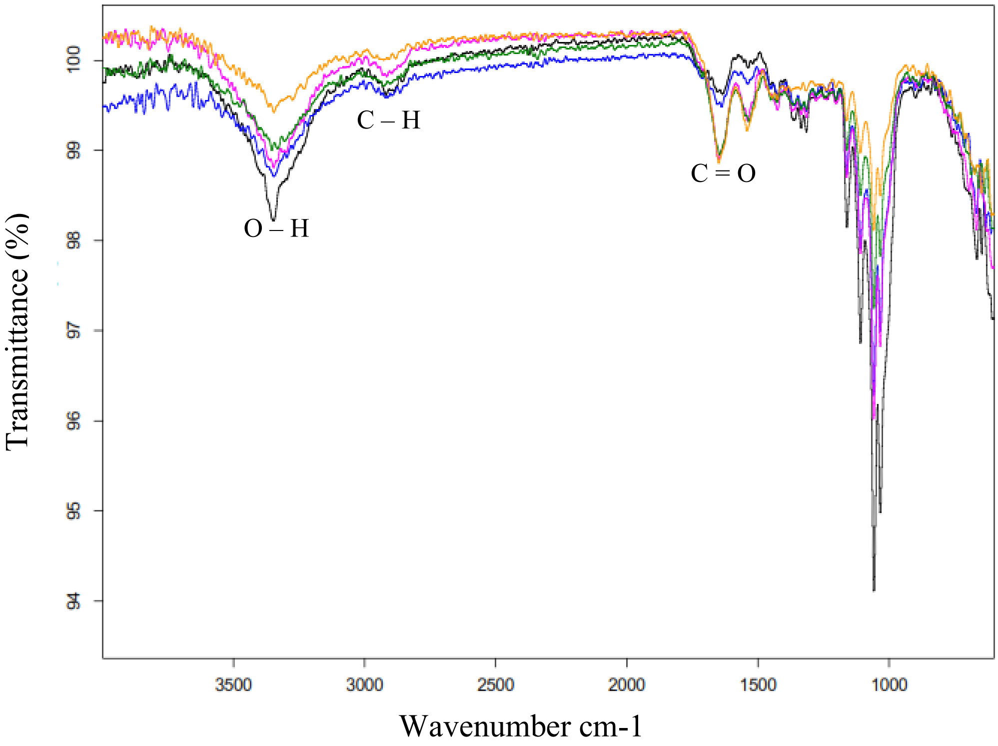

Fig 3. FTIR spectra of composite samples with different filler-to-matrix mass ratios.

The IR transmittance spectra of the samples are presented in Fig. 3. The figure shows the characteristic wavenumbers corresponding to the functional groups present in the five synthesized composite variations, namely sample A1 (0:100%) in black, sample A2 (15:85%) in blue, sample A3 (25:75%) in green, sample A4 (35:65%) in pink, and sample A5 (45:55%) in yellow. All samples exhibit similar spectral profiles with characteristic bands appearing at nearly identical wavenumbers, although variations in transmittance intensity are observed. These differences in transmittance mainly reflect compositional variations arising from different filler-to-matrix mass fractions, rather than the formation of new functional groups [13,14]. The similarity of the spectral positions indicates that the incorporation of hair particle filler does not significantly alter the fundamental chemical structure of the bacterial cellulose matrix.

A broad band observed at approximately 3346 cm^{-1} corresponds to the stretching vibration of hydroxyl (O–H) groups originating from bacterial cellulose. With increasing hair particle filler content, the transmittance in this region increases slightly, indicating a reduction in the relative contribution of hydroxyl groups from the cellulose matrix. The broad nature of this band also suggests the presence of hydrogen-bonding interactions between cellulose hydroxyl groups, keratin functional groups, and citric acid molecules within the composite structure [5,19]. Another absorption band appears at around 2930 cm^{-1}, corresponding to the stretching vibration of aliphatic C–H groups, which are present in both cellulose and keratin structures. The transmittance in this region slightly increases with increasing filler content, indicating a reduction in the relative intensity of C–H absorption from the cellulose matrix as the proportion of hair particles increases.

Additionally, a transmittance decrease observed around 1650 cm^{-1} becomes more pronounced with increasing filler fraction. This band is attributed mainly to the amide I (C=O) vibration characteristic of keratin proteins in human hair, indicating that the increased absorption in this region primarily originates from the compositional contribution of keratin rather than the formation of new carbonyl groups [20]. If extensive esterification between citric acid and cellulose had occurred, an additional ester carbonyl band would typically appear around 1720–1740 cm^{-1} [4,21]. However, no distinct new band is clearly observed in this region, suggesting that esterification reactions within the composite are limited.

The region between 1500 and 1000 cm^{-1}, known as the fingerprint region, shows several overlapping bands associated with the structural vibrations of cellulose and keratin. Bands in the range of approximately 1160–1050 cm^{-1} are commonly assigned to C–O–C and C–O stretching vibrations [13,22] of the cellulose backbone. Although these bands overlap and cannot be individually resolved, their presence confirms the contribution of the polysaccharide network within the composite.

Overall, the FTIR spectra indicate that the incorporation of hair particle filler mainly modifies the transmittance intensity of characteristic bands due to compositional differences between bacterial cellulose and keratin, while the primary chemical structure of the matrix is not significantly altered. Thus, the interactions within the composite are predominantly governed by physical interactions such as hydrogen bonding rather than the formation of new covalent bonds [18,20]. These observations are consistent with the mechanical and morphological results, which highlight the influence of interfacial interactions and filler dispersion on the composite properties.

4. Conclusions

This study demonstrates that the incorporation of hair particle filler affects the mechanical and structural characteristics of bacterial cellulose–based composites. Increasing the filler fraction generally leads to a decrease in tensile strength and an increase in elastic modulus, with the composite containing a 15%:85% filler-to-matrix ratio (sample A2) showing the highest tensile strength of 9.64 ± 1.08 MPa and an elastic modulus of 130.46 ± 4.94 MPa within the investigated compositions. Morphological observations indicate that higher filler contents are associated with the presence of more visible voids, which may contribute to the reduction in mechanical performance. FTIR transmittance spectra confirm that the main functional groups (O–H, C–H, and C=O) remain present in all samples, while variations in band transmittance mainly reflect compositional differences between bacterial cellulose and keratin rather than the formation of new functional groups. However, this study has several limitations, including the absence of quantitative porosity analysis and statistical significance testing, which should be addressed in future research to provide a more comprehensive understanding of the composite behavior.

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