

Article

Fabrication of paper sheets coatings based on chitosan/bacterial nanocellulose/ZnO with enhanced antibacterial and mechanical properties

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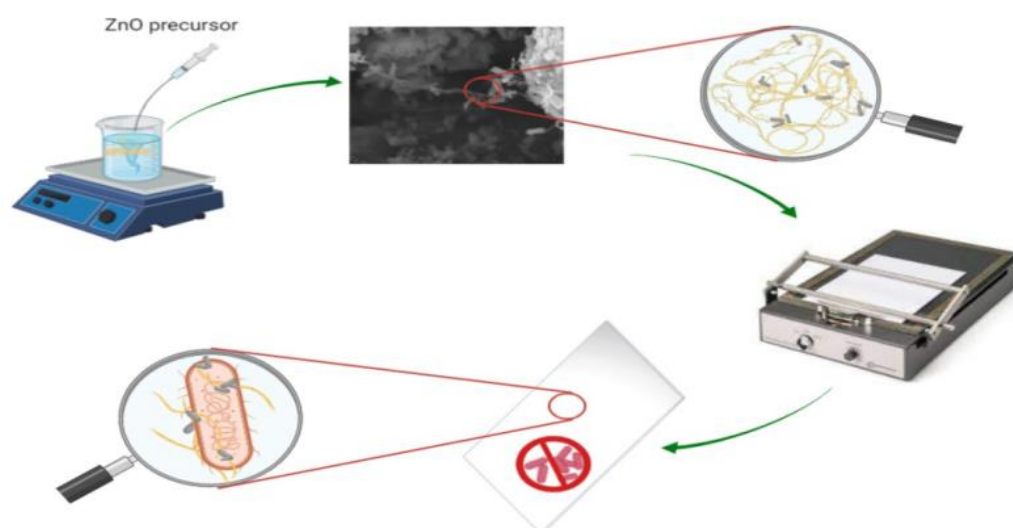
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Abstract: Here, we designed the composition of the coating of the paper sheets composed of chitosan, bacterial cellulose (nanofibres), and ZnO with boosted antibacterial and mechanical activity. We investigated the compositions with ZnO exhibiting two different sizes/shapes: (1) rods and (2) irregular sphere-like particles. The proposed processing of bacterial cellulose resulted in the formation of nanofibers. Antimicrobial behavior was tested using *E. coli* ATCC® 25922™ following ASTM E2149-13a standard. Mechanical properties of the paper sheets were measured by comparison of tearing resistance, tensile strength, and bursting strength according to ISO 5270 standard. The increased antibacterial response is assigned to the combination of chitosan and ZnO (independently of its shape and size), while the boosted mechanical behavior is due to bacterial cellulose nanofibers. Therefore, the proposed composition is an interesting multifunctional mixture for coatings in food packaging applications.



Keywords: biopolymers; paper packaging; antimicrobial activity; nanoparticles

1. Introduction

The food packaging industry is a source of tons of plastic waste every year. It raises environmental concerns about greenhouse gases emission, a growing carbon footprint, and the welfare of sea animals and birds [1,2]. Therefore, the attempts to use more eco-friendly materials, such as biopolymers, are intensified. Those materials can exhibit good properties when incorporated into the mass of cellulose used for paper production or can be applied as coatings of the paper sheets [3].

One of the biopolymers used in packaging applications is chitosan – a polysaccharide that consists of D-glucosamine and N-acetyl-D-glucosamine monomers. It is obtained as the result of deacetylation of chitin – the ingredient of the crustacean shells. Chitosan gained much recognition due to its properties such as biodegradability, non-toxicity, filmmaking, and antimicrobial potential [4].

Bacterial cellulose is a biopolymer obtained from stationary or dynamic bacterial cultures. One of the most recognized producers belongs to *Komagataeibacter* genus (previously called *Acetobacter xylinus* or *Gluconacetobacter xylinus*) [5]. Bacterial cellulose is created at the edge of the liquid-gas phases in the form of hydrogel [6]. It is characterized by significant water retention, the possibility of obtaining different shapes and good mechanical properties compared to plant cellulose due to the lack of lignin or hemicellulose in its structure. Moreover, the crystallinity of bacterial cellulose is higher than in plant cellulose, resulting in its good mechanical properties [4].

One of the most promising application areas of bacterial cellulose is the paper and textile industry, where it can be used as a reinforcing agent in the form of nanocrystals (BCNC) or nanofibers (BCNF). Bacterial cellulose nanocrystals, also called nanowhiskers, are characterized with a diameter of 4-25 nm and a length of 100-1000 nm and occur in the shape of needles [7]. They can also be obtained in the form of nanofibers which have a length of around 10-100 nm and are obtained by physical processes such as homogenization, ball milling, or sonication [8–10].

Moreover, the possibility of incorporating various nanoparticles into different biopolymers provides new opportunities for their application. In addition, nanoparticles, such as silver, copper, or zinc oxide, are used as one of the components to enhance the antimicrobial properties of the paper packaging. Zinc oxide is widely used in research concerning electronics, photocatalysis, sensors, construction materials, or medicine [11–13]. It is considered GRAS (generally recognized as safe) by the U.S. Food and Drug Administration (FDA) and can be used as a part of the food packaging [14,15]. Zinc oxide exhibits efficient antimicrobial response and is mainly incorporated into the polymer matrix as an additive.

Many studies have utilized the above-mentioned components in the fabrication of films or paper coatings. Salari *et al.* [4] used BCNC, chitosan, and silver nanoparticles to fabricate nanocomposites in the form of film. It expressed good barrier and mechanical properties and exhibited antimicrobial activity. George and Siddaramaiah [16] formulated a composite based on BCNC and gelatine, which resulted in the enhancement of mechanical properties compared to pristine gelatine film. Moreover, George *et al.* [17] noted higher thermal stability and better mechanical properties of BCNC-poly(vinyl alcohol) composite in comparison to the film from the pure polymer. In turn, Viana *et al.* [18] obtained films based on pectin and bacterial cellulose in the form of BCNF. The films were characterized with better mechanical properties and water resistance than pristine pectin films. Yadav *et al.* [19] produced chitosan-based films containing ZnO nanoparticles and gallic acid that exhibited good antimicrobial and mechanical properties and antioxidant behavior. Some authors prepared coated paper samples. Divsalar *et al.* [20] prepared a paper coated with chitosan, ZnO, and nisin that showed antibacterial activity. Prasad *et al.* [21] obtained papers with ZnO and soluble starch. The samples exhibited improved whiteness, oil absorbency, and excellent antifungal and UV-protecting properties.

Here, we focused on the fabrication of paper coating based on chitosan, bacterial cellulose, and ZnO to induce antimicrobial response and enhance mechanical properties

with respect to the pristine paper. The processing of bacterial cellulose sheets was performed via lyophilization, ball milling, sonication, and re-lyophilization. It resulted in the formation of bacterial cellulose nanofibers (BCNF). These nanostructures were later decorated with ZnO particles in two different shapes: rods and irregular sphere-like particles. Therefore, the morphology and their performance when used in paper coatings were efficiently tuned.

2. Results

2.1. Characterisation of the composites and coatings

SEM analysis allowed to observe the change in bacterial cellulose structure after 1-hour sonication. The process resulted in partial separation of the nanofibers. The comparison of non-sonicated, agglomerated bacterial cellulose and the sonicated sample is presented in Fig. 1. Moreover, SEM enabled the preliminary verification of the ZnO synthesis on the bacterial cellulose surface. Micrographs in Fig. 2 present the morphology modification of the nanocomposites affected by different temperatures applied during metal oxide deposition: room temperature (a) and 80 °C (b). The noticeable differences were noted, such as the size and the shape of the ZnO particles. ZnO synthesized at room temperature is characterized by nanometric size and non-uniform structure. Moreover, the particles were agglomerated. However, the shape of ZnO synthesized at 80 °C can be described as elongated rods with a diameter ~200 nm and length 1 µm.

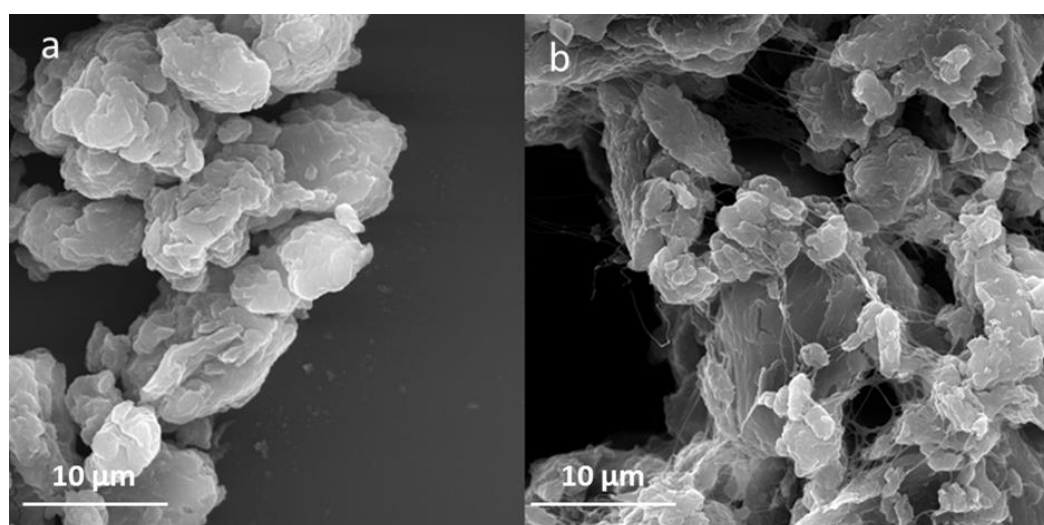


Figure 1. SEM micrographs of bacterial cellulose (a) before and (b) after sonication

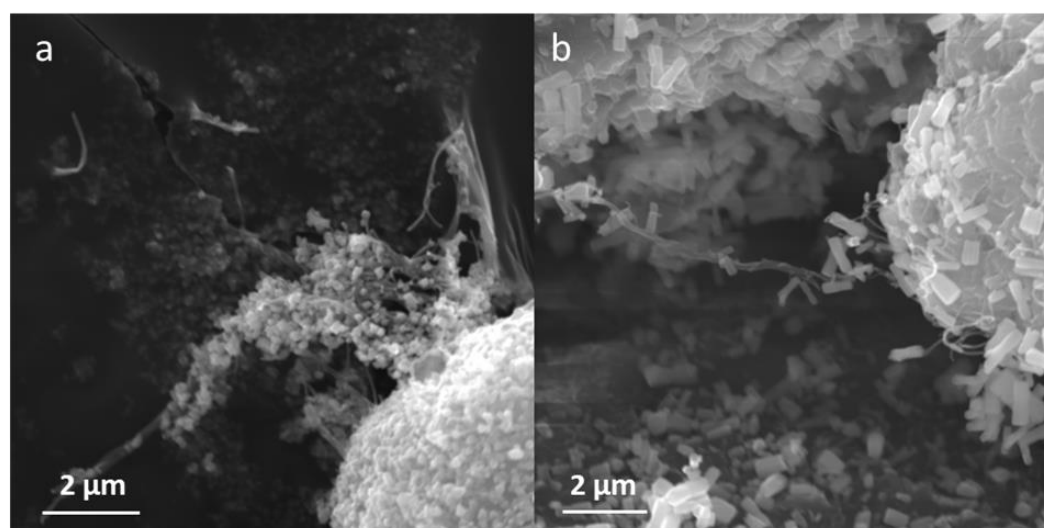


Figure 2. SEM micrographs of bacterial cellulose modified with ZnO (a) at the room temperature; (b) at 80°C

Moreover, SEM analysis was applied to observe the surface of the coated paper sheets (Fig. 3). However, this method did not allow the comprehensive comparison, probably due to the low content of the filler (10% w/w in relation to dry mass of chitosan) and sputtering of the samples with chromium that precluded the differentiation of the metallic and non-metallic structures. The only example of the probable identification of ZnO particles in BCson80ZnO composites is shown in Fig. 3 (indicated with red arrows).

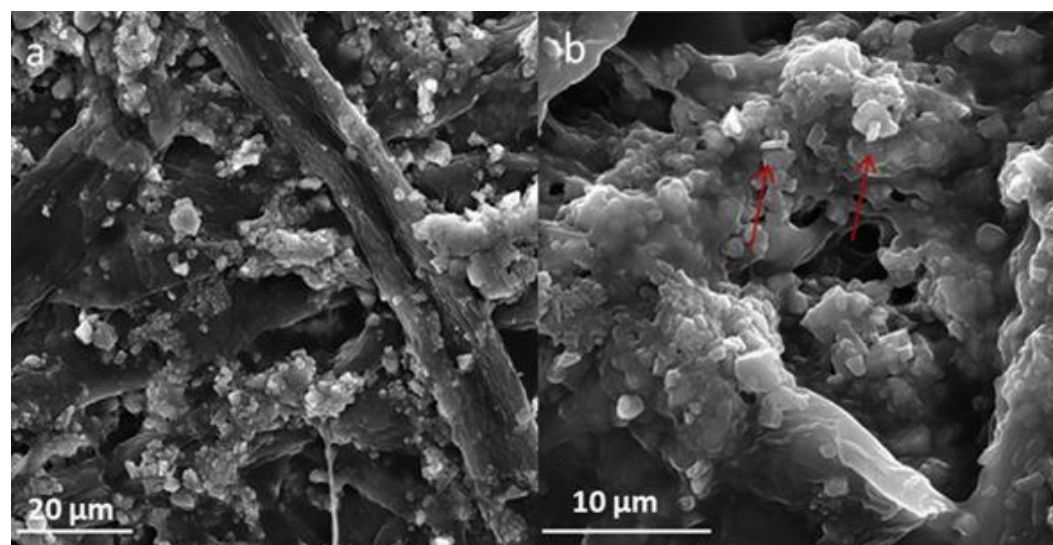


Figure 3. SEM micrographs of papers coated with (a) chitosan; (b) BCson80ZnO

TEM analysis allowed to confirm the successful fabrication of the bacterial cellulose-ZnO composites and the detailed analysis of their morphology. Fig. 4 presents the images of all obtained composites. BCsonTZnO composite consisted of agglomerated ZnO particles in the network of relatively well-separated nanofibers of bacterial cellulose. On the other hand, BCson80ZnO composite contained ZnO long rods placed on the fibers. TEM analysis confirmed that sonication induced bacterial cellulose nanofibers fabrication. Moreover, elemental mapping of the obtained composites was carried out to assess the location of the elements in the samples. It confirmed the presence of carbon (C-K), oxygen (O-K), and zinc (Zn-K and Zn-L) (Fig.5).

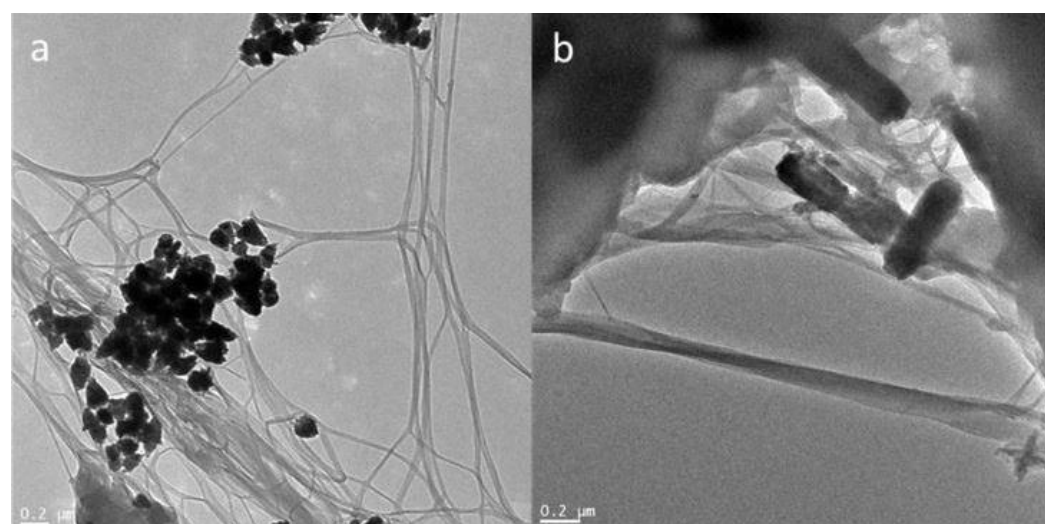


Figure 4. TEM micrographs of bacterial cellulose-ZnO composites (a) BCsonTZnO; (b) BCson80ZnO

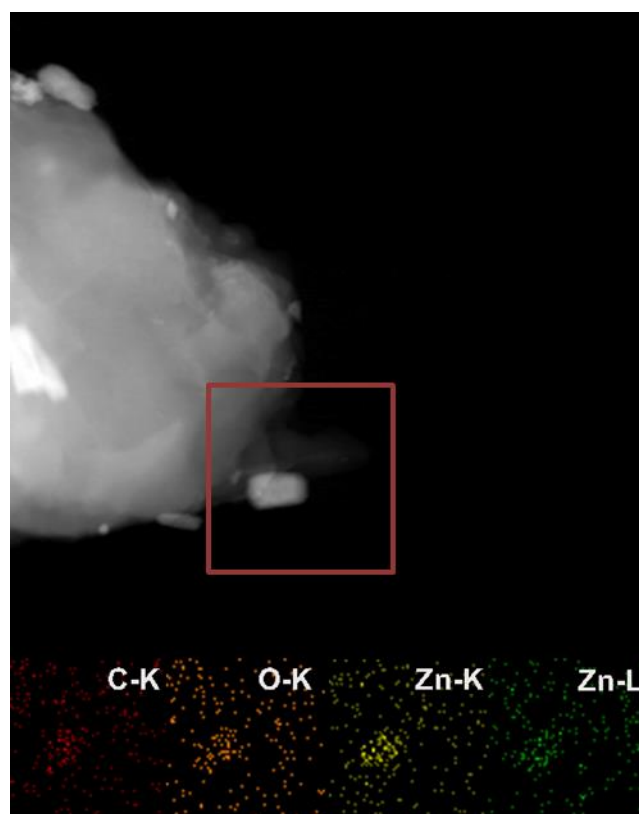


Figure 5. Elemental analysis of BCson80ZnO composite

Figure 6 presents X-ray diffraction patterns taken for bacterial cellulose composites. The patterns revealed that the obtained samples exhibit a typical crystalline form of cellulose I. For each sample, major diffraction peaks at 14.6° , 16.7° , and 22.6° corresponding to the crystallographic plane of (1 $\bar{1}0$), (110), and (200), respectively, could be recognized. However, a number of additional peaks in all nanocomposites samples can be observed. Sharp and intense Bragg reflections are positioned at 31.05° , 34.58° , 36.48° , 47.68° , 56.78° , 63.07° , 66.53° , 68.11° , 69.29° . These reflections are well-matched with the usually reported signals of ZnO wurtzite hexagonal structure attributed to (100), (002), (101), (102), (110), (103), (200), (112), and (201), respectively (in accordance with standard PDF card no. 01-079-0207). The obtained XRD pattern of BCsonTZnO displays few additional diffraction peaks in the range of 20° – 65° . These comparatively weak signals are assigned to zinc hydroxide, which is an intermediate product during the formation of ZnO in alkaline solution. Zn(OH)₂ can only be found in the sample prepared at room temperature.

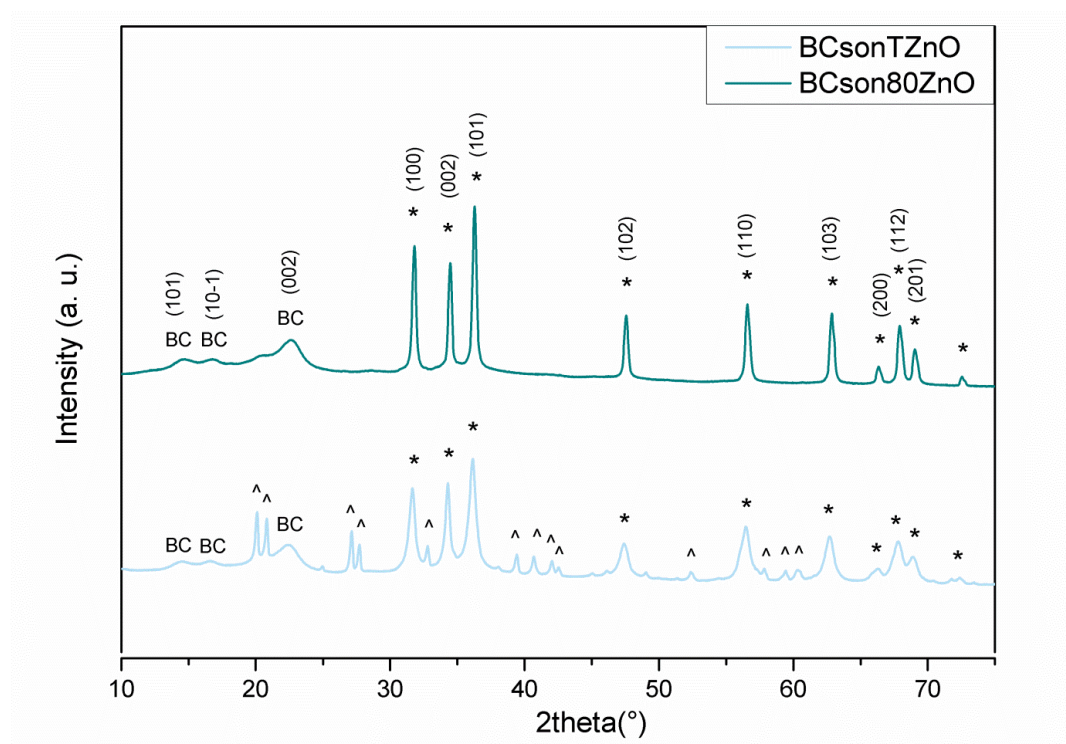


Figure 6. XRD analyses of the obtained bacterial cellulose-ZnO. * - ZnO, BC – bacterial cellulose, ^ - Zn(OH)₂

Figure 7 shows FT-IR spectra of two representatives of bacterial cellulose in respect to the pristine bacterial cellulose sample. All spectra contain the main bands: at 3432 cm⁻¹ attributed to the O-H stretching, 2921 cm⁻¹ and 2853 cm⁻¹ characteristic for C-H stretching of CH₂ and CH₃ groups, 1165 cm⁻¹ assigned to C–O–C antisymmetric bridge stretching of 1,4-β-d-glucoside and 1053 cm⁻¹ corresponding to bending of the C–O–H bond of carbohydrate. The band centered around 1626 cm⁻¹ is due to the O-H bending of adsorbed water. It is present in all samples, and its intensity is the highest for the pure BC sample, indicating the hydrophilic character of the material. There are some differences between BC and all obtained composites. Subtle changes can be observed in the low-wavenumber region, where the bands characteristic of Zn-containing groups have been observed (400-500 cm⁻¹). The results may be related to the strong chemical interaction between cellulose and ZnO phases. Spectra obtained for the composites show broadening of the bands in the region 3200-3600 cm⁻¹, probably due to the rearrangement and increase in the hydroxyl group content. Moreover, the signals in the 2850-2950 cm⁻¹ region have become more distinctive but less intense after the functionalization process.

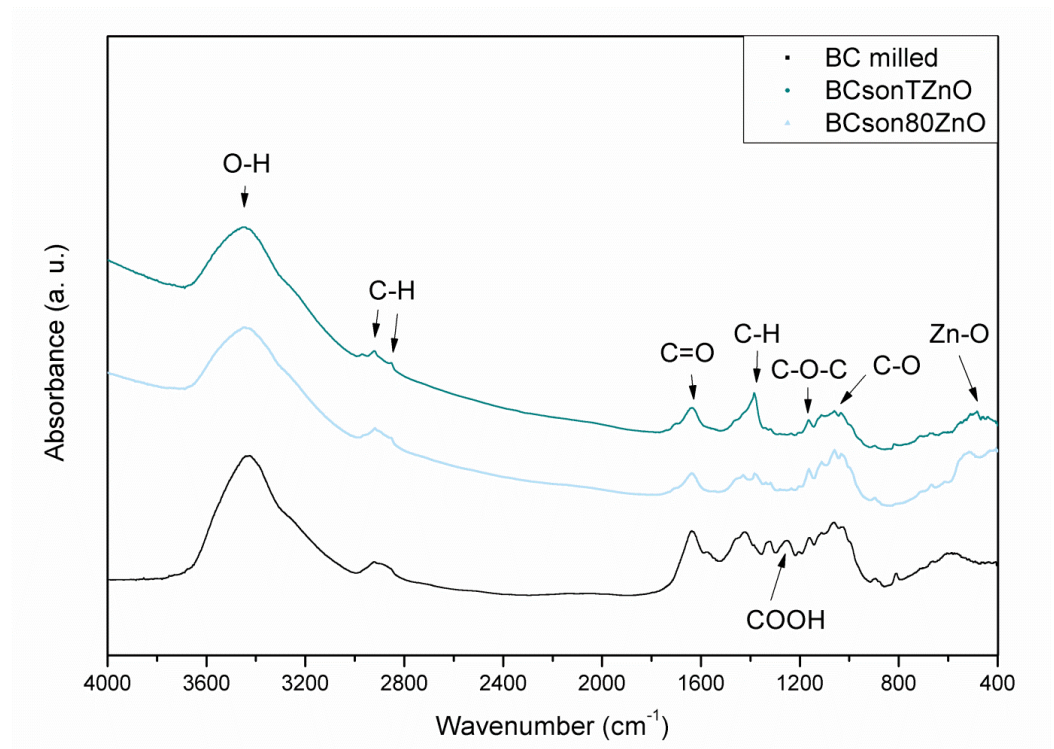


Figure 7. FT-IR analyses of the obtained bacterial cellulose-ZnO composites

2.2. Antibacterial properties

The results of antibacterial properties are presented in Fig. 8. The antimicrobial assay showed a 96.36% reduction of *E. coli* titer after incubation with chitosan-coated paper and 100% of reduction with both papers coated with fabricated chitosan-bacterial cellulose-ZnO composites. In the case of pristine paper, the titer of bacteria was increased by 70%. In this study, it is crucial to point out that there is no shape/size effect of ZnO on the antimicrobial response.

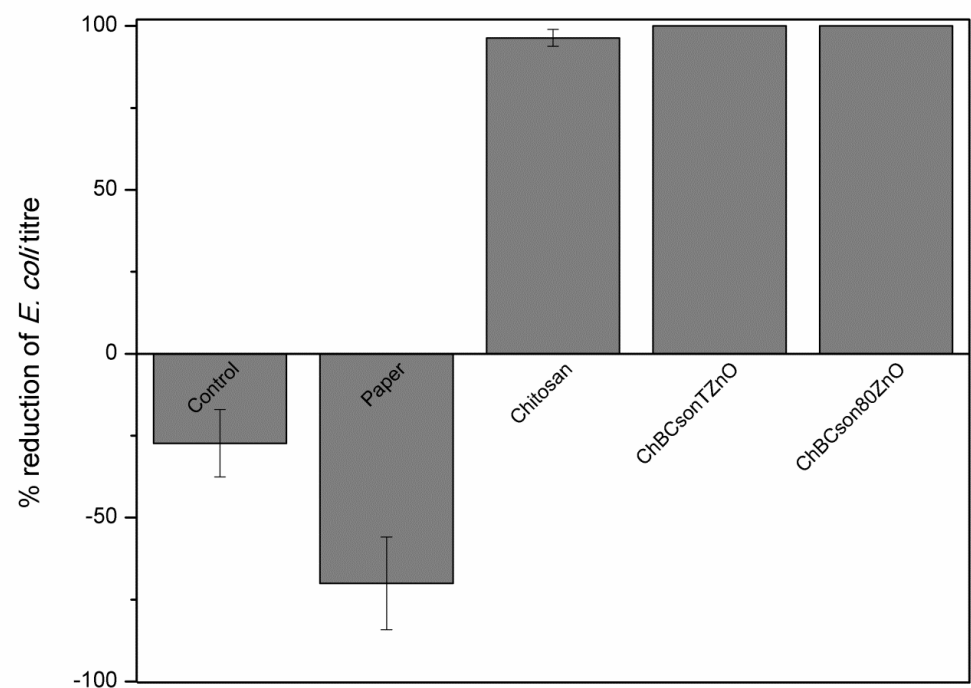


Figure 8. Antibacterial properties assay according to ASTM E2149-13a standard (chitosan – paper coated with chitosan, ChBCsonTZnO – paper coated with chitosan, sonicated bacterial cellulose and ZnO synthesized at the room temperature, ChBCson80ZnO - paper coated with chitosan with sonicated bacterial cellulose and ZnO synthesized at 80°C)

2.3. Mechanical properties

To verify the effect of the various coating on the mechanical properties of the non-coated and coated paper sheets were subjected to mechanical tests. The tests evaluated the tensile strength, tearing strength, and bursting strength. Because of the slight changes in weight of the coated paper sheets, all the collected data are presented as strength index, which is a preferable factor to compare the samples with different grammages. Table 1 summarizes the obtained results indicating enhancement of mechanical parameters of the coated samples in respect to the pristine paper is observed. The tensile strength value of the coated paper sheets has increased by ~ 10 % compared to the reference sample. Tearing strength value was improved by 15 % in the machine direction and even more in cross-machine direction – 25 %. The bursting strength value of the coated paper sheets also increased by 18%. Interestingly, both composite-coated samples have shown similar mechanical performance indicating that the shape of ZnO does not affect it. It is more induced by the presence of nanocellulose and chitosan.

Table 1. The effect of the different coatings on the mechanical properties of paper

	Pristine paper	Chitosan	ChBCsonTZnO	ChBCson80ZnO
Tensile index [N m/g]	40.4	43.18	44.00	44.74
Tear index (MD) [mN m ² /g]	5.69	6.54	6.54	6.52
Tear index (CD) [mN m ² /g]	5.97	8.28	6.78	6.78
Burst index [kPa m ² /g]	1.37	1.57	1.50	1.61

3. Discussion

Our study indicated that fabricated paper coated with composites based on chitosan, bacterial cellulose, and ZnO exhibited enhanced antibacterial activity and mechanical performance. The antibacterial effect may be explained by the analysis of the antimicrobial properties of chitosan and ZnO. In the case of chitosan, its properties are associated with many factors, e.g., pH of the solution, molecular weight and degree of acetylation, or the surface of contact with the cells [4,23]. There are several mechanisms of antimicrobial activity of chitosan described in the literature [24,25]: (1) the first one is explained as an electrostatic attraction of positively charged chitosan and negatively charged components on the surface of bacterial cells, (2) another proposed mechanism is based on the ability of chitosan to penetrate the cell, bind with DNA and disturb the transcription and translation processes. An effect of such action is the disorganization of protein synthesis crucial for sustaining cells' homeostasis. The last (3) mechanism relies on the chelation of metal ions by amino groups of chitosan. Such effects are observed in pH above six since amino groups become unprotonated and the pair of electrons are donated to metal. It results in the creation of metal complexes. Many authors examining the antimicrobial activity of chitosan or its composites used the agar diffusion method and obtained different, sometimes not coherent, results [4,23,26]. Almasi *et al.* and Sani *et al.* [23,26] confirmed the antimicrobial activity of chitosan. However, Salari *et al.* [4] did not. Salari *et al.* [4] explained the research outcome by suggesting that the problems with diffusion through agar strongly limit chitosan's antimicrobial activity in the form of non-soluble film. At the same time, they underlined that it is probably due to the fact that chitosan activity is limited by the surface of contact with microorganisms. According to

ASTM E2149-13a standard, pieces of analyzed materials are suspended in a buffer that is constantly stirred. Therefore, the contact surface with bacteria is greater than in the agar diffusion method and is probably a reason for the high reduction of bacterial titer. The antimicrobial properties of the prepared chitosan solution could also be increased by the sonication process conducted to obtain uniform dispersion in acetic acid solution. Applied chitosan was characterized with a high deacetylation degree (75-85%) and medium molecular weight (190,000-310,000 Da). According to Czechowska-Biskup *et al.* [27], sonication of chitosan may lead to a decrease in molecular weight. Qin *et al.* [28] suggested this may result in higher antimicrobial activity.

The bactericidal properties of zinc oxide were described by many authors and are a well-known phenomenon [29,30]. The antimicrobial activity of zinc oxide nanoparticles is hypothesized to be based on three main mechanisms: 1) the release of Zn^{2+} ions, 2) interaction of nanoparticles with bacteria which may result in cell disruption, 3) production of reactive oxygen species (ROS) [31,32]. In the presented research, the size/shape of the particles was assessed by transmission electron microscopy. The size of ZnO particles was estimated as nanometric in the case of ZnO synthesized at room temperature, while ZnO synthesized at 80°C was characterized with micrometric dimensions. On that basis, we hypothesize that the most probable antimicrobial mechanism of the fabricated coatings was acting through zinc ions released to the buffer used in the conducted assay. Such a process is connected to the amphoteric nature of ZnO in water solutions. The ions bind with thiol groups of the enzymes involved in cell respiration, which disorganizes their functions and leads to cell death [31,32]. The assay of antimicrobial activity of the composites revealed that the combination of chitosan and ZnO was more effective than chitosan alone and lead to 100% reduction of the bacterial titer.

The benefits of using nanocellulose in the improvement of mechanical properties have already been addressed in the literature [33]. The addition of cellulose nanostructures having a high surface to volume ratio improves the formation of hydrogen bonding within cellulose pulp which increases density boosting the mechanical performance of paper. Tanpichai *et al.* [34] reported significant enhancement of mechanical properties of the paper composed of 50 wt% nanofibrillated cellulose (NFC). Tensile strength and strain were 10-fold and 3-fold higher than those of the paper without NFC, respectively. Jin and his coworkers [35] have designed paper coating using nanofibrillated cellulose as a coating agent. The results of their studies revealed that increased NFC addition led to the enhancement of tensile strength, which was achieved as a function of the NFC addition in the paper coating system. They demonstrated that 0.03 % addition of NFC improved the tensile strength of coated paper by approximately 2.5 %

Recent studies have also found the viability of chitosan as a paper coating not only for the implementation of antibacterial features but also to improve its mechanical properties. This enhancement of mechanical properties might be due to the fact that pores within the paper can be filled with polymer molecules. Zakaria *et al.* [36] have coated A4 paper with 2 wt% of chitosan solution and reported some changes in paper strength and toughness in comparison to uncoated samples. The burst and tensile strength values for the coated paper have increased by 9% and 6%, respectively. Moreover, it has been reported that chitosan adheres well to the fiber surfaces causing the formation of bridges between inter-fiber distances [37]. Thus coating paper with the mixture of chitosan and nanocellulose can benefit from the formation of bonds between cellulose fibers and nanocellulose fibrils. Therefore, exploiting the additive effect of bacterial cellulose nanofibers and chitosan for the mechanical properties reinforcement is possible.

4. Materials and Methods

Zinc acetate dihydrate ($\text{Zn}(\text{CH}_3\text{COO})_2 \times 2\text{H}_2\text{O}$), acetic acid, and medium molecular weight chitosan (190,000-310,000 Da, 75-85% deacetylated) were purchased from Sigma Aldrich (Poland). Sodium hydroxide (NaOH) was supplied from Chempur (Poland).

4.1. Bacterial cellulose growth, processing and functionalization with ZnO

Bacterial cellulose was produced by *Komagataeibacter xylinus* ATCC® 53524™. The culture was conducted for seven days at 30°C in S&H 1717 ATCC medium (bactopeptone 5 g/L, yeast extract 5 g/L, Na₂HPO₄ 2.7 g/L, citric acid 1 hydrate 1.15 g/L). After sterilization, the aqueous solution of mannitol (20% w/w) was added in the amount of 20 g/L, and the medium was buffered to pH = 5. Next, the inoculum was transferred to the new portion of the sterile medium and cultivated in a stationary culture (1.2 L) on the polypropylene trays for the next eight days at 30°C. After the incubation time, the sheet of bacterial cellulose was harvested, rinsed thoroughly with distilled water, and incubated in 0.1M NaOH solution at 80°C for 30 minutes to remove the residual cells. After that, the sheet was rinsed again with distilled water to remove the residue of NaOH.

The bacterial cellulose processing was based on the lyophilization of the whole sheet, ball milling (1 hour), sonication for 1 hour, and re-lyophilization. This approach resulted in bacterial cellulose nanofibers (BCNF) formation due to mechanical processing.

The synthesis of the zinc oxide particles was conducted directly on the bacterial cellulose samples according to the modified method by Ali *et al.* [22]. The calculated ratio of the bacterial cellulose:ZnO was 1:2. Briefly, 750 mg of the bacterial cellulose was inserted into 250 mL of the distilled water and stirred with a magnetic stirrer (300 rpm, 30 minutes) to obtain the homogenous dispersion. Next, Zn(CH₃COO)₂ × 2H₂O was added to the flask and stirred for the next 0.5 h. After that, 0.1M NaOH was introduced using a vacuum pump with the flow 3 mL/minute (Programmable Microfluidics Syringe Pump, NE-1002X). After this step, the obtained pellet was washed with ethanol, and the residues were rinsed with distilled water and filtered on Whatman filter (0.2 µm) until pH=7 was reached. Then the samples were dried at 60 °C.

4.1. Preparation of chitosan composites

The chosen concentration of the chitosan solution was 1.5% w/v in 1% v/v water solution of the acetic acid based on the preliminary attempts to coat a sheet of paper. The solution was placed on the magnetic stirrer (300 rpm) and sonicated for 15 mins until the homogeneous suspension was obtained. The final composites were prepared by adding 10% w/w (in relation to the dry mass of chitosan) of bacterial cellulose modified with ZnO into the chitosan solution. The composites were stirred with a magnetic stirrer for 30 minutes and sonicated (15 minutes) to obtain the homogenous dispersion of the modified bacterial cellulose particles. The final concentrations of bacterial cellulose and chitosan equaled 6.77% and 3.33%, respectively. Coating of the kraft paper sheets (grammage 90 g/m², Arctic Paper S.A., Poznan, Poland) was conducted with a coating machine (RK K Control Coater, UK) by the distribution of the nanocomposite on the surface of the paper sheet with a squeegee with a coating thickness of 12 µm. Afterward, the coated papers were air-dried. All the samples are listed in Table 2.

Table 2. The list of the tested samples

Sample abbreviation	Description of the sample
P	Non-coated paper
Ch	Paper coated with chitosan solution
ChBCsonTZnO	Paper coated with chitosan solution with the addition of the BCNF modified with ZnO synthesized at room temperature
ChBCson80ZnO	Paper coated with chitosan solution with the addition of the BCNF modified with ZnO synthesized at 80°C

4.3. Characterization

SEM analysis was conducted to assess the morphology of the bacterial cellulose and obtained ZnO particles. The samples were prepared on carbon tape and sputtered with chromium. The voltage of the electron beam equaled 20 kV (VEGA3 TESCAN). TEM analysis was carried out to precisely assess the size of the obtained ZnO particles and the efficiency of the sonication of the bacterial cellulose. The samples were sonicated and placed on a copper grid and incubated at 60°C to remove the water. The electron beam voltage used was 200 kV (Tecnai F30). The additional elemental analysis was run to identify the elements of the composites. XRD allowed the confirmation of the presence of the bacterial cellulose and ZnO particles in the samples prior to the insertion into chitosan solution. The powdered sample was placed on the rack, and the measurements were conducted in the range of 10-75° with the copper lamp as a radiation source ($K\alpha_1 = 1.54056 \text{ \AA}$) (X'Pert Philips Diffractometer). FT-IR method (Nicolet 6700 FT-IR) was used to identify the characteristic groups of the used components. The samples were prepared by forming tablets with KBr. The measurements were conducted in the range of 4000-400 cm^{-1} .

4.4. Antibacterial properties

The assessment of the antimicrobial properties of the coated paper was conducted according to ASTM E2149-13a standard (Standard Test Method for Determining the Antimicrobial Activity of Antimicrobial Agents Under Dynamic Contact Conditions). The bacterium was kept at -20°C in a trypticase soy broth (TSB) medium containing 10% glycerol. Before the study, the microorganism was revived on trypticase soy agar (TSA) and incubated at 37°C for 24 h. For experiments, bacteria were inoculated to falcon-type tubes (50 mL) containing 30 mL of TSB and incubated overnight at 37°C on a rotary shaker for 18 hours. Then, the cultures were diluted in phosphate buffer (0.3 mM KH_2PO_4), and the obtained inoculum was used for the experiments. One gram of each paper sample was weighed on the analytical scale and inserted into sterile Erlenmeyer flasks. Next, 50 mL of the inoculum was added, the solution was mixed by hand, and 100 μL of each sample was collected for the plating. All the samples were incubated at room temperature for 1 hour with shaking (150 rpm). Afterward, 100 μL of the cultures were collected for plating on plate count agar (PCA) in two repetitions. The plates were incubated at 37°C for 24 hours. After the incubation, the colonies were counted, and the reduction percentage was calculated in comparison to the control.

4.5. Mechanical properties

Mechanical properties of functionalized and non-functionalized paper samples were investigated. For comparative purposes, non-coated paper sheets were also tested and used as a reference sample. The mechanical tests were carried out at the Arctic Paper company in the air-conditioned room (23°C, 50% of the humidity level) according to ISO 5270 standard. Tearing resistance was determined using Elmendorf apparatus (Lorentzen & Wettres). The tensile strength measurements were conducted on the automatic tensile tester (Messmer Büchel, K465, Veenendaal, Netherlands), and the bursting strength was tested on the bursting strength tester (Messmer Büchel).

5. Conclusions

In summary, we fabricated the paper sheets coated by the composites based on chitosan, bacterial cellulose (nanofibres), and ZnO with enhanced antibacterial and mechanical properties. The experimental conditions allowed to tune the shape/size of ZnO: rods and irregular sphere-like particles, respectively. However, no size/shape effect on antimicrobial and mechanical response was detected. Boosting of mechanical properties has been assigned to the bacterial cellulose nanofibers formation during coating preparation. In addition, excellent antimicrobial activity was observed thanks to the co-application of chitosan and ZnO. Therefore, we assume that the designed paper coated with chitosan-bacterial cellulose-ZnO composites may be a promising composition in food packaging applications.

Author Contributions: Conceptualization, Joanna Jabłońska, Magdalena Onyszek and Ewa Mijowska; Formal analysis, Maciej Konopacki, Adrian Augustyniak, Rafał Rakoczy and Ewa Mijowska; Funding acquisition, Rafał Rakoczy and Ewa Mijowska; Investigation, Joanna Jabłońska, Magdalena Onyszek, Maciej Konopacki and Adrian Augustyniak; Methodology, Joanna Jabłońska, Magdalena Onyszek, Maciej Konopacki, Adrian Augustyniak and Ewa Mijowska; Project administration, Rafał Rakoczy and Ewa Mijowska; Resources, Rafał Rakoczy and Ewa Mijowska; Supervision, Rafał Rakoczy and Ewa Mijowska; Visualization, Joanna Jabłońska and Magdalena Onyszek; Writing – original draft, Joanna Jabłońska, Magdalena Onyszek, Maciej Konopacki, Adrian Augustyniak, Rafał Rakoczy and Ewa Mijowska; Writing – review & editing, Joanna Jabłońska, Magdalena Onyszek, Maciej Konopacki, Adrian Augustyniak, Rafał Rakoczy and Ewa Mijowska.

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Conflicts of Interest: Authors declare that they have no conflicts of interest.

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