FLORENTINA SEDERAVIČIŪTĖ

DEVELOPMENT AND INVESTIGATION OF BACTERIAL CELLULOSE FILM FOR THE CLOTHING INDUSTRY

SUMMARY OF DOCTORAL DISSERTATION

TECHNOLOGICAL SCIENCES, MATERIALS ENGINEERING (T 008)

Kaunas
2021

KAUNAS UNIVERSITY OF TECHNOLOGY

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This doctoral dissertation was prepared at Kaunas University of Technology, Faculty of Mechanical Engineering and Design, Department of Production Engineering, during the period of 2015–2020 and during an internship at Riga Technical University, Latvia (2017), and Tomas Bata University, the Czech Republic (2019).

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Editor: Brigita Brasienė (Publishing house "Technologija")

Dissertation Defence Board of Materials Engineering Science Field:
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The official defence of the dissertation will be held at 11 a.m. on 12th of April, 2021 at the public meeting of the Dissertation Defence Board of Materials Engineering Science Field in Dissertation Defence Hall at Kaunas University of Technology.

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The summary of dissertation was sent on 12th of March, 2021.
The doctoral dissertation is available on the internet http://ktu.edu and at the library of Kaunas University of Technology (K. Donelaičio Str. 20, 44239 Kaunas, Lithuania).

KAUNO TECHNOLOGIJOS UNIVERSITETAS

FLORENTINA SEDERAVIČIŪTĖ

BAKTERINĖS CELIULIOZĖS PLĖVELĖS APRANGOS PRAMONEI KŪRIMAS IR TYRIMAS

Daktaro disertacijos santrauka
Technologijos mokslai, medžiagų inžinerija (T 008)

2021, Kaunas

Disertacija rengta 2015-2020 metais Kauno technologijos universiteto Mechanikos inžinerijos ir dizaino fakulteto Gamybos inžinerijos katedroje ir stažuojantis Rygos technikos universitete, Latvijoje (2017) bei Tomas Bata universitete, Čekijoje (2019).

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Redagavo: Ilona Petrovė

Medžiagų inžinerijos mokslo krypties disertacijos gynimo taryba:
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Disertacija bus ginama viešame medžiagų inžinerijos mokslo krypies disertacijos gynimo tarybos posėdyje 2021 m. balandžio 12 d. 11 val. Kauno Technologijos universiteto Disertacijų gynimo salėje.

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Disertacijos santrauka išsiųsta 2021 m. kovo 12 d.
Su disertacija galima susipažinti internetinėje svetainėje http://ktu.edu ir Kauno Technologijos Universiteto bibliotekoje (K. Donelaičio g. 20, 44239 Kaunas, Lietuva).

INTRODUCTION

Research problem and relevance of the work. Nowadays, the textile and clothing industry is increasingly confronted with terms of sustainable development, sustainable consumption, or alternative materials. Over the past decade, scientists and designers have focused on environmentally friendly products, developing new materials that have exceptional functional properties and are applicable beyond traditional fields. These studies are closely linked to depleting energy resources, ecological problems, over-consumption, large amounts of waste, air pollution, etc. The textile and clothing industry is vital to the global economy; thus, it is important to use and research cleaner, renewable, and environmentally friendly materials. One of them is cellulose, which is known as the most abundant polymer on earth with the total annual biomass of about 1.5 x 1012 tons; the cellulose fibers have excellent mechanical and physical properties, but require soil, sunlight, carbon dioxide to grow, and water. Bacterial cellulose, meanwhile, is a unique material that does not require soil, sunlight, pesticides, or large amounts of water to grow. Bacterial cellulose can be extracted all year round by fermentation in a special medium that can be produced using waste from the food and beverage industry. Based on the research, it can be observed that due to its unique structure and unique properties, this material is perfectly integrated into biomedicine, paper, food, and others. Industries, projects are being developed, and this research has been carried out in order to apply bacterial cellulose film to the clothing industry.

The bacterial cellulose film could be a great alternative to address the challenges facing the textile and fashion industries. The potential of bacterial cellulose is increased by the fact that this material is not new in other fields, and based on the already known research, it can be stated that the technology of bacterial cellulose production complies with the principles of ecology and sustainability, has no negative impact on the environment, and is not harmful to human health. Bacterial cellulose is biodegradable, which is closely related to responsible and conscious consumption. The use of bacterial cellulose for the production of organic clothing would allow "as much" material to be "grown" as needed, and at the end of its useful life, such clothing would break down easily or could be used as compost, thus avoiding waste and reducing environmental pollution.

Despite the advantages of bacterial cellulose film, its application in garment manufacturing has not been extensively investigated yet. Bacterial cellulose is studied by modification or used as a raw material, but untreated bacterial cellulose has been insufficiently studied: its longevity, technological, some physical, or morphological properties, etc. are not known. In most cases, in research and ongoing projects, bacterial cellulose film is grown in standardized media, but the properties of bacterial cellulose film grown in Kombucha drink have not been extensively studied. In the last decade, Kombucha drink has become a popular commercial product, in the production of which the film formed on the surface of the drink is discarded as a by-product. In order to avoid it, this waste can be used purposefully; thus, it is important to study the characteristics of bacterial cellulose film grown in Kombucha beverage medium, the influence of chemical treatment processes on its properties, the physical properties of the film and evaluate its applicability.

The aim of the dissertation is to evaluate the possibilities of application of untreated bacterial cellulose obtained during Kombucha fermentation in the clothing industry and determine the influence of washing and chemical treatment processes on the properties of the film.

The objectives of the research:

1. To evaluate the influence of fermentation conditions, drying temperature, storage time and temperature on the properties of untreated bacterial cellulose film;
2. To determine the influence of washing methods and duration on the morphological and physical properties of bacterial cellulose film;
3. To determine the influence of 1,3-dimethylol-4,5-dihydroxyethylene urea concentration on the morphological and physical properties of bacterial cellulose film.

Scientific novelty and practical importance. Recently, the demand for organic materials has increased significantly due to the environmental reasons. Companies, designers and scientists are intensively looking for alternative sources of materials and their application in daily lives. One of the most important industries for the economy is the textile and clothing industry, where a growing interest in sustainable and environmentally friendly materials is developing; thus, it is vital to find and research materials that are less pollutant and can replace the traditional textiles used nowadays. Based on these important facts, it can be stated that the dissertation presents relevant tricks that contribute to the projects presented to the public and continue this idea. The dissertation provides a broader and more detailed analysis of the bacterial cellulose film, performed experiments that have not been performed yet, compared to the works of other scientists. The bacterial cellulose film extracted during the natural fermentation of the widely known *Kombucha* drink is unique in its production process. Bacterial cellulose is known to have excellent physical properties and could contribute to the list of sustainable materials in the textile and clothing industry. However, it is the bacterial cellulose film grown in *Kombucha* beverage medium that has not been sufficiently studied yet, and there is a lack of research and information on the characteristics of this material for its application in the clothing industry. The dissertation research provided an assessment and recommendations for the application of unprocessed bacterial cellulose to the clothing industry: the physical properties of the bacterial cellulose film were determined, in which different ingredients were added to the culture medium to expand the color gamut of the film. The whole washing process of the bacterial cellulose film was investigated as well, and the effective washing time of 8 hours using 0.5% NaOH solution was determined. It was found that after washing the bacterial cellulose film, its crystalline phase increases, as a result of which the tensile strength of the film increases 3 times, but the deformation decreases 6.5 times. Water vapor permeability is as well reduced by 82.0%. After evaluating the results of the experiments, the most efficient storage conditions of untreated bacterial cellulose film in a cool (+4.0 °C) environment and the most suitable drying temperature, taking into account the physical properties of the bacterial cellulose film (25.0 °C), the proposals for further experiments were made. The studies analyzing bacterial cellulose film treatment processes using the traditional textile chemical reagent 1,3-dimethylol-4,5-dihydroxyethylene urea, and it was found that the use of the chemical reagent restores the amorphous areas of the bacterial cellulose film, resulting in improved deformation characteristics (90.0%) and water conductivity vapor rates (80.0%).

Approbation of the research results. 5 scientific publications were published on the topic of the dissertation, of which 3 publications were published in the Clarivate Analytics Web of Science database with citation index, 1 publication in the Clarivate Analytics Web of Science database without citation index, 1 publication conference proceedings. The results of the research were published in 10 international conferences.

Structure of the dissertation. The dissertation consists of an introduction, 3 chapters, conclusions, a list of references (279 positions), a list of published scientific works, appendices. The volume of the dissertation is 110 pages; they contain 41 figures, 32 tables, 13 formulas.

CONTENT OF THE DISSERTATION

The introduction of the dissertation introduces the advantages of bacterial cellulose film as an alternative material and the issue of lack of research. The research problem and the relevance of the work are discussed; the aim of the work is formulated; the tasks of the work are set; the scientific novelty of the work, practical value and the author's contribution are discussed.

1. LITERATURE REVIEW

The first chapter of the **literature review** presents the challenges and solutions for sustainable development and sustainable use in the fashion industry, properties of alternative materials, areas of application, advantages and disadvantages, methods, properties, areas of application and limitations of bacterial cellulose production and processing.

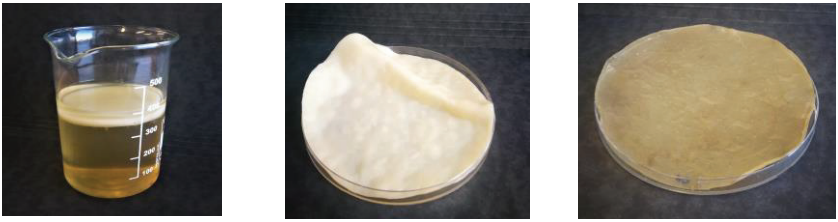
2. RESEARCH METHODOLOGY

The second chapter, **methodological part**, provides information about the research object, cultivation culture, cultivation medium and conditions, technological treatment conditions and regimes.

Experimental materials

A bacterial cellulose film formed during the fermentation of the beverage by a symbiosis of acetic acid bacteria and yeasts is shown in Figure 1.

BC film growth medium is prepared from 1 l of boiled water, 4 g of green tea (100.0% Ceylon green tea “Impra royal elxir green”, Sri Lanka), 100 g of sucrose (Dansukker, “Panevėžys plus sugar”, Lithuania) and 100 ml 6.0% apple cider vinegar (apple cider vinegar “Extra line”, Lithuania). *Kombucha* tea mushroom (MB Arbatologija, Lithuania) is placed in a culture medium at a temperature of 20.0 °C. The fermentation is carried out in a standard environment (temperature 20.0 ± 2.0 °C and relative humidity 65.0 ± 5.0%). After 7 days, a floating, gel-like film of material (Fig. 1 a) forms on the surface of the growth medium, the thickness of which increases with increasing growth time.



a b c

Fig. 1. Bacterial cellulose membrane: a) during growing process, b) in wet state, c) in dry state

In order to investigate the color change possibilities of BC film, the film was grown by the same method, but the composition of the growth medium was changed by introducing a color-giving component (Table 1).

Table 1. BC film growth medium

Ingredients	Amount	Code of samples		
		BC_T$_{KA}$	**BC_T$_{ZA}$**	**BC_T$_{BS}$**
Boiled water	1000 ml	x	x	x
White sugar	100 g	x	x	x
6.0% apple cider vinegar	100 ml	x	x	x
Green tea	4 g		x	x
Kombucha tea mushroom	1 piece	x	x	x
Rosehip tea*	4 g	x		
Beetroot juice**	600 ml			x
Lemon juice***	40 ml			x

* Rosehip Petal Tea, Sudan.
** Beetroots, Lithuania.
*** Lemons, Argentina. The juice was pressed by the author with professional multifunctional citrus fruit juicer "ZYLE ZY503CJ".

In order to evaluate the efficiency of the sucrose source, BC film was grown by the same method, but the sucrose source was changed in the growth medium as shown in Table 2.

Table 2. Sucrose sources used in BC film growth medium

Code of samples	**Sucrose source**
BC_S$_{BC}$	White sugar
BC_S$_{RC}$	Brown sugar*
BC_S$_{FR}$	Fructose**
BC_S$_{GL}$	Glucose***

* Brown cane sugar "Alvo", Mauritius.
** Crystallized fructose "Alvo", Turkey.
*** Glucose "Skanėja", Lithuania.

For testing, the BC film is prepared by washing it to remove organic impurities. Initially, the samples were washed under running tap water and treated with two different methods, i.e., deionized water and a weak alkaline solution (Table 3). Deionized water and alkaline solution were changed after 1, 2, 4, and 8 hours. After the washing procedure, the samples are removed from the solutions, washed with deionized water, and blotted dry with a paper towel to remove excess moisture.

Table 3. BC film washing methods

Code of samples	**Washing method**
BC_PD_{H2O}	BC washed with deionized water in a water bath with shaker for up to 24 hours at 30 ± 0.2 °C at 120 rpm
BC_PD_{NaOH}	BC washed with 0.5% NaOH in a water bath with shaker for up to 24 hours at 30 ± 0.2 °C at 120 rpm

Following the washing procedure, the samples were treated with 1,3-dimethylol-4,5-dihydroxyethylene urea (DMDHEU), commercially known as Arkofix (ARKOFIX NZK LIQ M1000, Switzerland), which is used to finish traditional textiles. The BC washed samples were soaked for 8 hours (10 minutes in the case of BC film shredding) in the prepared solution: 100 g / l DMDHEU and 15% magnesium chloride (MgCl2) (Sigma-Aldrich) from DMDHEU concentration, which was used as a catalyst. DMDHEU concentrations varied by 5.0%, 10.0%, and 20.0% (Table 4).

Table 4. BC film treatment methods

Code of samples	**Washing**	**Treatment**	**Drying**
BC_PDA_5	BC washed with 0.5% NaOH in a water bath with shaker for up to 24 hours at 30 ± 0.2 °C at 120 rpm	5.0% DMDHEU and 15.0% $MgCl_2$, from DMDHEU concentration, for 8 hours	Laboratory oven, 25.0±1.0 °C + 4 minutes at 150.0 °C
BC_PDA_{10}	BC washed with 0.5% NaOH in a water bath with shaker for up to 24 hours at 30 ± 0.2 °C at 120 rpm	10.0% DMDHEU and 15.0% $MgCl_2$, from DMDHEU concentration, for 8 hours	Laboratory oven, 25.0±1.0 °C + 4 minutes at 150.0 °C
BC_PDA_{20}	BC washed with 0.5% NaOH in a water bath with shaker for up to 24 hours at 30 ± 0.2 °C at 120 rpm	20.0% DMDHEU and 15.0% $MgCl_2$, from DMDHEU concentration, for 8 hours	Laboratory oven, 25.0±1.0 °C + 4 minutes at 150.0 °C

Table 5. Method for drying BC samples

Code of samples	Drying method
BC_NN	-
BC_ND$_{25}$	Laboratory oven, 25.0±1.0 °C, up to 30 hours
BC_ND$_{50}$	Laboratory oven, 50.0±1.0 °C, up to 10 hours
BC_ND$_{75}$	Laboratory oven, 75.0±1.0 °C, up to 5 hours

After washing and treatment procedures, BC samples were dried by convection drying (Table 5).

Experimental methodology

Prior to the BC film determination experiments, all samples were stored for 24 hours under standard climatic conditions according to the standard LST EN ISO 139: 2005 [1].

The thickness *h* of each bacterial cellulose sample was measured with a digital indicator DPT 60, with an accuracy of 0.01 mm.

The mass *m* of the wet and dry BC samples is measured on a Kern EG-N laboratory balance, to the nearest of 0.001 g.

The thickness of the BC film fibers was measured using SEM images with CorelDraw X7 software.

The values of diameter *d* (mm) of the samples in 4 places were measured using a ruler accurate to 0.5 mm, and their average was determined.

In order to determine the color of the BC film specimens, the specimens were scanned with an EPSON Perfection V370 Photo scanner at a color image resolution of 600 dpi. After calculating the RGB values of each sample, the colors that corresponded to the calculated RGB values of each sample were described using Color Schemer Online V2.

For the BC film sample bonding experiments, the samples were prepared by the following methods.

For thread splicing experiments, the specimens were sewn on a Juki DDL-888 industrial sewing machine with black polyester thread Amann Saba C80, no. 0348, class 1 overlay seam with parameters: needle number - 90 Nm, stitch density - 4 cm^{-1}, seam width - 10 mm. The tensile characteristics of these specimens were determined in accordance with LST EN ISO 13935-1: 2001 [2].

The samples were bonded using adhesive (Table 6). BC specimens prepared according to the conditions described above were joined by an interlocking joint with an overlap size of 10 mm. An adhesive made of potato starch (Aloja-Starkelsen, Latvia) was used to join the samples. When the samples were bonded without adhesive, the wet BC samples were stacked on top of each other that the edges overlap by 10 mm. Pressures of 0.02 MPa and 0.06 MPa were used to join the samples.

Table 6. BC film sample bonding parameters

Code of samples	**Drying temperature, *T***	**Drying time, *t***	**Group I**	**Group II**	**Group III**
			Pressure, MPa	**Pressure, MPa**	**Pressure, MPa**
BC_JS$_{25}$	Laboratory oven, 25.0±1.0 °C	24 h.	0.00	0.02	0.06
BC_JS$_{50}$	Laboratory oven, 50.0±1.0 °C	8 h.	0.00	0.02	0.06
BC_JS$_{75}$	Laboratory oven, 75.0±1.0 °C	6 h.	0.00	0.02	0.06

The hardness of BC samples was determined according to the LST EN ISO 868: 2003 standard [3] with a Mitutoyo 811-332-10 hardness tester (KTU, MIDF) according to Shore, measuring scale A.

Water vapor transmittance rate (WVTR) and water vapor permeability (WVP) were tested in a desiccator according to ASTM E96 / E96M-10 [4].

The water absorption capacity (WAC) was tested according to ISO 20158: 2018 [5].

The wettability of BC film samples was evaluated by measuring the contact angle (CA) using a PG-3 pocket goniometer according to ISO 15989: 2004 [6].

When testing the moisture content of BC samples according to ISO 585: 1990 [7], the mass of wet samples was measured at different time intervals of the wash cycle (0, 0.5, 1, 4, 8, and 24 hours).

The tensile characteristics of BC film were determined using a TINIUS OLSEN H10 KT tensile testing machine according to LST EN ISO 527-3: 2018 [8].

Shear strength characteristics were determined according to LST EN 1465 [9].

A scanning electron microscope (SEM) was used for morphological studies of BC film and performed with FEI Quanta 200 FEG (KTU, Institute of Materials).

The qualitative analysis of the chemical structure of BC film was performed using Fourier transform infrared spectroscopy (FTIR).

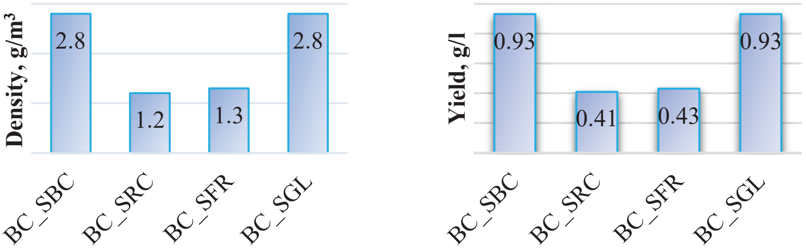
The thermal characteristics of the BC film were investigated by differential scanning calorimetry (DSC) using a Universal V4.1D heat analyzer (TA Instruments, USA).

An X-ray diffractometer (XRD) Bruker SMART X2S analysis was used to describe the crystalline and amorphous regions of the BC film as well as the degree of crystallinity.

3. RESULTS OF INVESTIGATIONS

3.1. Analysis of the properties of untreated bacterial cellulose film

Figure 2 shows the results of the density and the yield of the BC film grown using a different source of sucrose in the growth medium.



a b

Fig. 2. BC film density (a) and yield (b) using different sucrose sources in the culture medium

The highest values of density and yield of the BC film obtained during the fermentation were obtained in the growth medium using white sugar (BC_SBC) and glucose (BC_SGL), and the lowest using brown sugar (BC_SRC) and fructose (BC_SFR).

3.1.1. Bacterial cellulose fermentation medium and color

As shown in Figure 3, the density of green tea (BC_T$_{ZA}$) in the medium and the amount of BC material grown were the highest at 9.2 g/m^3 and 0.42 g/l, respectively. For the preparation of the medium using rosehip tea (BC_T$_{KA}$), the density index was 4.1 g/m^3; the amount of grown material (yield) was lower 0.29 g/l. When trying to grow BC film using beetroot juice (BC_T$_{BS}$), the density of BC film was the lowest 3.5 g/m^3, and the yield was 0.15 g/l. The results showed that in this experiment, lower values of BC content were obtained after the introduction of rosehip tea and beetroot juice into the growth medium comparing with the usual medium with green tea.

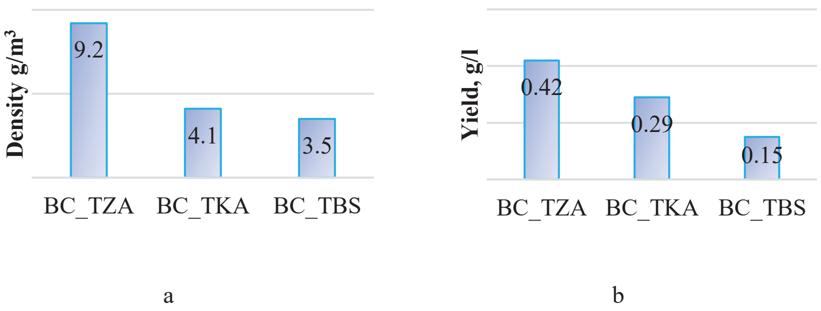


Fig. 3. BC film density (a) and yield (b) indicators when grown in different media

The thermograms of the DSC experiment show (Fig. 4) that different BC film growth media influence the structure of the grown material. The change in endothermic reaction peaks from 114.59 °C (BC_T$_{BS}$) to 164.11 °C (BC_T$_{ZA}$) can be attributed to the higher crystalline areas discussed later, obtained by growing BC film in the conventional medium with green tea without colourful additives.

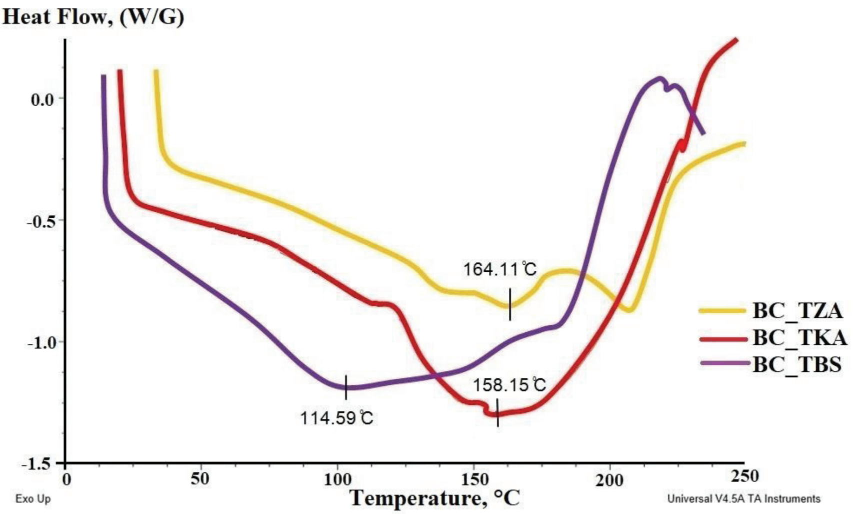


Fig. 4. DSC thermograms for samples BC_T$_{ZA}$, BC_T$_{KA}$, and BC_T$_{BS}$

Table 7. Mechanical characteristics of BC film

Parameter	BC_T_{BS}	BC_T_{KA}	BC_T_{ZA}
Tensile strength σ, MPa	9.08±4.68	11.86±3.79	19.14±2.98
Strain ε, %	9.90±3.27	23.80±2.86	34.80±5.34
Tensile modulus E, MPa	1.21	1.09	1.78

From the tested specimens, the lowest tensile strength was determined for BC_T_{BS} specimens (9.08 MPa) and the highest for BC_T_{ZA} specimens (19.14 MPa), for which the elongation values were the highest as well 34.80% (Table 7). The comparison of results with other studies confirmed that BC film is a sufficiently strong and elastic material, but the introduction of color additives to the culture medium may impair both its strength and the deformation properties.

3.1.2 Influence of storage conditions and duration on the properties of bacterial cellulose film

It has been investigated that the change in tensile strength, depending on the storage conditions of the BC film (Table 8), decreases uniformly when the samples are stored at 23 ± 1 °C. The initial tensile strength of the specimens was 79.23 MPa, which decreased to 24.37 MPa in 10 days, to 21.79 MPa in 20 days, and decreased to 15.62 MPa in 30 days. Thus, the strength of the samples decreased by around 80.0% within 30 days. Meanwhile, BC film samples were stored in a cool environment at +4 °C; the tensile strength decreased as well, but not that significantly, i.e., to 58.63 MPa in 10 days, to 63.10 MPa in 20 days, and to 30.92 MPa in 30 days. When the specimens were stored in a cool environment for 30 days, their tensile strength was reduced by 61.0%.

Table 8. Tensile characteristics of tested specimens

Code of samples	Tensile strength σ, MPa	Elongation ε, %
BC_SS_{K}	79.23±3.54	23.85±4.21
BC_SS_{N10}	24.37±4.85	6.01±5.01
BC_SS_{N20}	21.79±2.48	3.85±2.65
BC_SS_{N30}	15.62±3.95	4.21±1.23
BC_SS_{S10}	58.63±2.93	15.72±3.56
BC_SS_{S20}	63.10±5.96	17.10±4.89
BC_SS_{S30}	30.92±3.84	8.40±2.78

As it can be seen, the BC film retains its mechanical properties better when stored in a cool environment.

3.1.3. Determination of characteristics of bacterial cellulose film bonding

When sewing BC film samples, the sewing machine did not always manage to obtain high-quality compounds: the layers of material slipped and stretched. The average tensile strength of the class 1 overlay was 14.8 MPa, and the elongation was 40.9%. It has been observed that BC film specimens were missing at the seam joint, indicating that the suture compromised the integrity of the material. This indicates that the BC film is weaker than the seam, but the material deforms better.

The highest shear strength was when the specimens had been dried at 50 °C (BC_JS_{50}). When the samples had been dried at 25 °C, 75 MPa (BC_JS_{25}) was determined, and at the highest temperature of 75 °C, the shear strength of the dried samples was the lowest (BC_JS_{75}). It cannot be said that high drying temperatures are unsuitable for obtaining reliable adhesion of compounds, because samples tht had been dried at high temperatures were in many cases lacking in adhesion.

When evaluating the elongation at break of these specimens, it has been observed that the higher is the drying temperature, the lower is the elongation at break. When dried at 25 °C, the elongation at break was 28.0%, at 50 °C, it was 12.0%, and for samples dried at 75 °C, it was 9.0%. This confirms the results described above that the film loses its deformation properties at high temperatures.

The mechanical characteristics were evaluated as well for the specimens, which were bonded using different pressure. Figure 5 shows that the maximum shear strength of the compounds (30.61 MPa) was determined for the specimen joints, using 0.06 MPa pressure and drying at 25 °C, which is almost 6 times higher than for the specimen joints without pressure.

The lowest shear strength was found in all the cases for compounds formed without the use of pressure, but even the lowest pressure allows the formation of compounds with significantly higher shear strength, leading to the conclusion that pressure has a positive effect on bonding, and the BC film adheres more reliably. Comparing the samples with the previous studies, it has been observed that although high temperature affects the mechanical properties of the BC film negatively, the values of the bond strength increase almost 6 times with the use of pressure.

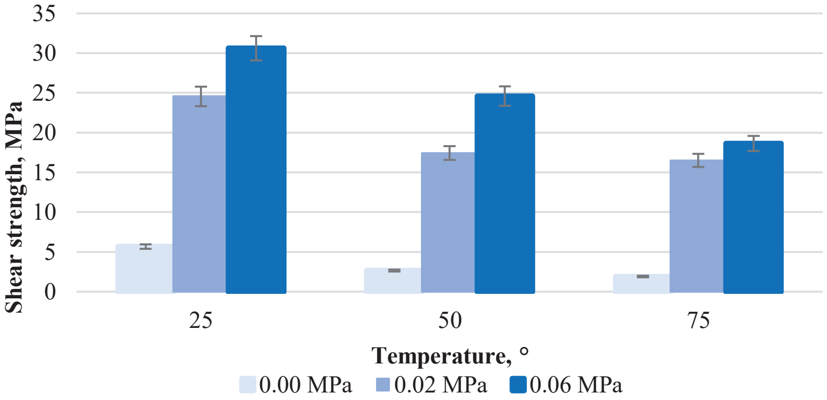


Fig. 5. Influence of pressure on shear strength of BC film bonding

The maximum shear strength of BC film specimens dried at 50 °C was 43.37 MPa (BC_JA$_{50}$), using a natural adhesive to join the BC film specimens. The results show that low temperature is not suitable for the specimens, as the strength of the compound with the adhesive layer is determined to be the lowest (8.06 MPa); these specimens of the compounds break rapidly through the joint. At 75 °C, the specimens were joined tightly enough; it breaks not through the joint, but when the BC film is disintegrated. Thus, the compounds that are stronger than the formed material have been created.

It has been observed that the higher is the temperature at which the samples are dried, the worse are the deformation properties of the compounds. Although the adhesive bond in the compound was not strong at 25 °C, the deformation properties remained best, and the elongation at break was 14.26%, it decreased to 13.28% at 50 °C, and 12.94% at the temperature of 75 °C. The elongation results of the compound confirmed that the mechanical properties of the BC film deteriorated at high temperatures, and the film became brittle, although the adhesive bond in the compound that had been formed at 50 °C was reliable and had the highest tensile strength.

3.1.4. Influence of drying temperature on bacterial cellulose film properties

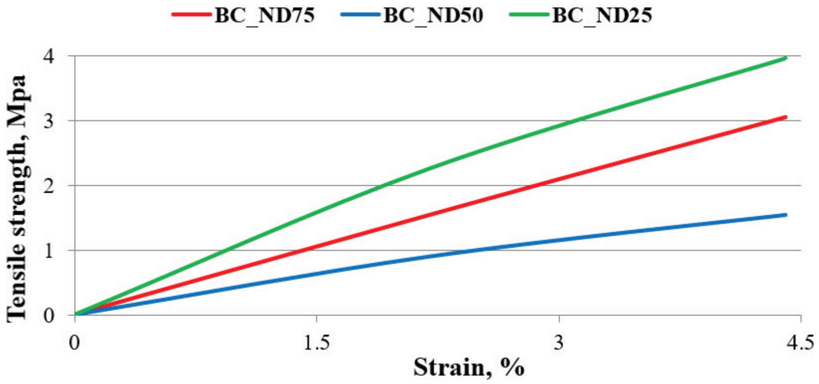
The change in the thickness of the samples was evaluated during the drying process (Table 9). An average reduction in thickness of 87% was recorded when drying until the sample gained a constant mass. The drying of the samples at room temperature decreased the most by 97%, drying at 25 °C by 84.0%, at 50 °C by 89.0%, and at 75 °C by 80.0%.

Table 9. Influence of drying process on BC film mass, geometric changes, and hardness

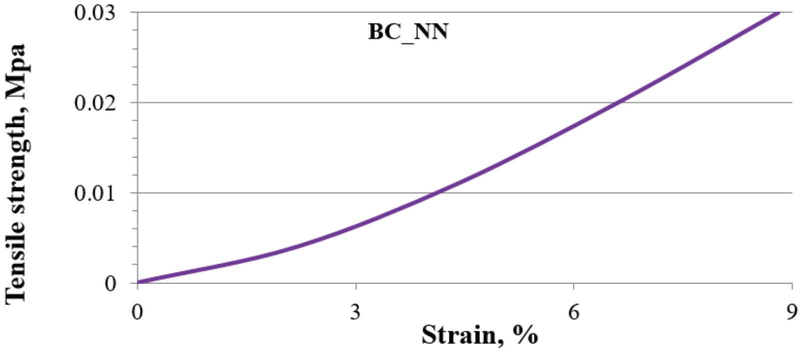
Parameter	**BC_ND$_{25}$**	**BC_ND$_{50}$**	**BC_ND$_{75}$**
Drying temperature, *t* °C	25.0 ± 1	50.0 ± 1	75.0 ± 1
Mass of the wet sample, g	141.32	190.76	152.33
Mass of the dry sample, g	120.82	175.12	134.50
Change in mass, %	-85.5±2.63	-91.80±4.25	-88.3±2.85
Thickness of the wet sample, mm	0.43	0.53	0.79
Thickness of the dry sample, mm	0.36	0.47	0.63
Change in thickness, %	-83.9±6.85	-88.60±3.78	-79.6±4.25
Diameter of the wet sample, mm	14.60	17.70	15.40
Diameter of the dry sample, mm	14.59	17.69	15.39
Shrinkage, %	0.04±0.32	0.05±0.45	0.03±0.14
Hardness (Shore A), %	41.0±2.34	46.0±1.36	61.0±3.48

The shrinkage of the longitudinal dimensions of the specimens (diameter was measured in the case of round specimens) during drying was recorded as well, which was insignificant, i.e., 0.02%, when the specimens were dried at room temperature, 0.04% when drying at 25 °C, 0.05% by drying at 50 °C, and 0.03% by drying at 75 °C. It has been observed that the higher is the temperature at which the samples are dried, the more uneven the surface of the samples is fixed after drying.

3.1.5. Dependence of mechanical characteristics on BC drying temperature



a



b

Fig. 6. BC tensile strength and strain curves of initial zone: (a) BC specimens dried at different temperatures, (b) wet specimens

In Figure 6 (p. 18-19), the initial zone of tensile strength and strain curves is presented, which shows the behavior of the material under the influence of low tensile forces. The tensile strength values of the wet specimens (BC_NN) were found to be the lowest of all the tested specimens, i.e., 0.4 MPa, but the elongation value of the gel-like material was high (37.9%), and the low modulus of elasticity was 0.40. The samples dried at 25 °C and 50 °C as well have low modulus of elasticity and are continuous, ranging from 18.8% to 23.9% in the absence of elongation. Dried specimens have much higher tensile strength values than the wet specimens, i.e., 17.30 MPa (BC_NN) and 27.9 MPa (BC_ND$_{25}$). BC_ND$_{75}$ specimens dried at 75 °C showed the lowest tensile strength, and the lowest elongation at 12.80 MPa and 4.5%, respectively, these specimens have the highest hardness due to high temperature drying.

3.1.6. Evaluation and recommendations for the application of untreated bacterial cellulose for the clothing industry

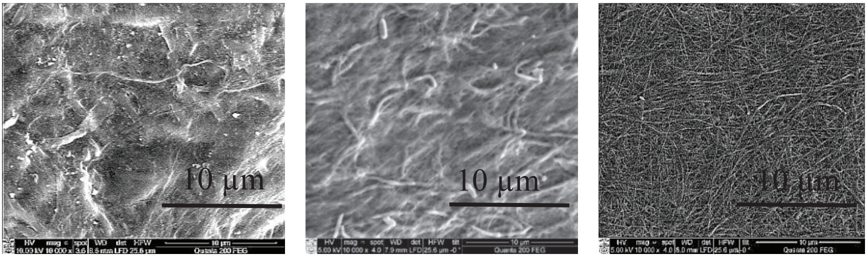
Based on the results of the previous studies, it is recommended to ferment the bacterial cellulose film samples using white sugar and green tea to ensure the most efficient film growth and obtain a purer form of cellulose, to choose cool (+4 °C) conditions for storage of prepared specimens to maintain the positive properties of the film for longer than under standard room conditions. It is advised to not use threaded bonding of bacterial cellulose films for untreated film, but consider leaching impurities in the bacterial cellulose film that interfere with quality thread bonding. It is recommended to use 0.06 MPa pressure when gluing the specimens together, select temperature up to 50 °C when bonding samples with the adhesive. It is recommended that samples of bacterial cellulose film should be dried at 25 °C to preserve its mechanical properties.

Consideration should be given to studies in order to control the thickness of the bacterial cellulose film and its distribution in the sample. Further experiments and cultivation of bacterial cellulose film were carried out according to the prepared recommendations.

Assessing the results, it can be concluded that untreated bacterial cellulose film is not suitable for use in the clothing industry, its properties are not stable and vary depending on many factors, as it is an organic material; therefore, further research is needed to control the properties of the film better and investigate the influence of the respective processes on the properties of the BC film.

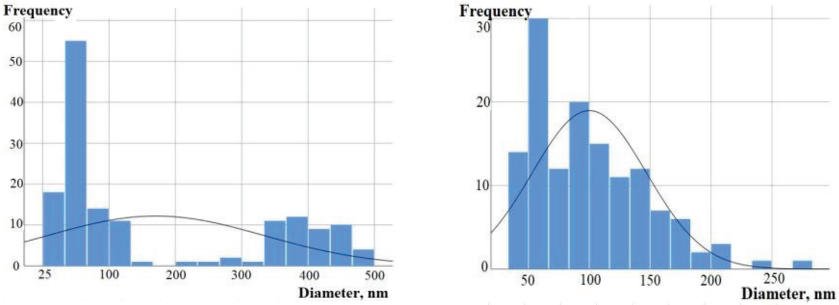
3.2. Influence of purification on the properties of bacterial cellulose film

The structure of the dried BC film samples was analyzed by electron scanning microscopy (SEM) (Fig. 7). In the images magnified 10,000 times, a fibrous structure of the BC film with micropores can be observed. As shown in Figure 7 a, the structure of the unwashed sample is unclear due to the above binders (usually sucrose), and the thickness of the cellulose fibers cannot be determined or measured. The structure of BC_PD$_{H_2O}$ samples was cleaner and clearer, but unwashed bacteria are clearly visible, and by-product residues (Fig. 7 b), when measuring fiber thickness, it can be observed that the diameter varies in a wide range of 50–500 nm (Fig. 8 a). The BC_PD$_{NaOH}$ sample shows a tridimensional (3D) network of disordered cellulose fibers with a diameter of around 100 nm (Fig. 8 b) (Fig. 7 c).



a b c

Fig. 7. SEM images of BC film samples: a) BC_ND, b) BC_PD$_{H_2O}$, c) BC_PD$_{NaOH}$



a b

Fig. 8. Cellulose fiber diameter measurements for BC film samples: a) BC_PD$_{H2O}$, b) BC_PD$_{NaOH}$

The XRD results of the untreated sample showed three diffraction peaks at 14.5°, 16.79°, and 22.65° (Fig. 9), which are generally assigned to the regions of the crystallographic plane amorphous region (101), amorphous region (010), and crystalline region (200) and characterizes type I cellulose [10]. The peaks in the washed samples were recorded for the BC_PD$_{H2O}$ sample at 14.24°, 22.47° and the BC_PD$_{NaOH}$ sample at 14.68°, 22.92°, which are assigned to the cellulose phases 1α and 1β, respectively. After washing the amorphous parts of the fiber, no peaks were recorded. The analysis of the X-ray diffractogram results showed that both washing methods did not significantly affect the structure of the BC film, and no conversion from cellulose I to cellulose II was observed.

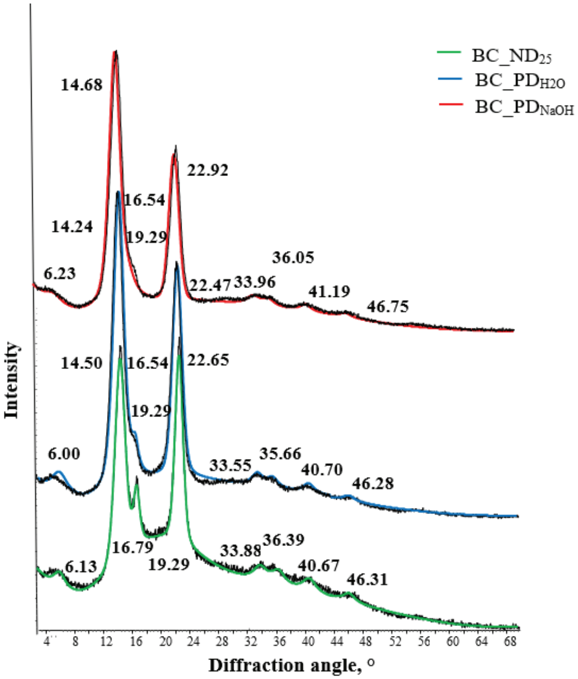


Fig. 9. X-ray diffractogram for samples BC_ND$_{25}$, BC_PD$_{H2O}$, and BC_PD$_{NaOH}$

The influence of washing on the mechanical behavior of the specimens was evaluated according to the tensile characteristics. As it can be seen from Table 2, the elongation of the unwashed specimen (BC_ND$_{25}$) is high 45.18%, but the strength of the material is lower (tensile strength is 27.91 MPa, and elastic modulus is 44 MPa), compared to the purified specimens. The values of tensile strength of the purified samples are similar (75.75 MPa BC_PD$_{H2O}$ and 73.71 MPa BC_PD$_{NaOH}$) and almost 3 times higher than the BC_ND$_{25}$ samples. However, the elongation values of these samples were obtained much lower than BC_ND$_{25}$ (8.36% BC_PD$_{H2O}$ and 5.47% BC_PD$_{NaOH}$, respectively).

The results confirmed that the tensile strength as well as the tensile modulus (950 MPa BC_PD$_{H2O}$ and 1069 MPa BC_PD$_{NaOH}$) increased significantly, and the BC film lost deformation properties after the washing procedure. This may be related to the removal of amorphous areas from the BC film membrane (as confirmed by the results of XRD and FTIR experiments), which may increase the tensile strength of the film, but reduce the deformation properties of the film.

3.3. Influence of 1,3-methylol-4,5-dihydroxyethylene urea on the properties of bacterial cellulose film

The infrared spectrum of the samples treated with the chemical reagent (DMDHEU) became less intense at 3344 cm^{-1}, 2899 cm^{-1}, and 934 cm^{-1}. The main changes after the treatment process can be observed at three peaks: a significant increase at 1698 cm^{-1}, which is attributed to the occurrence of NC = O groups, a significant increase at 1470 cm^{-1}, which is attributed to the occurrence of NH groups, and a significant increase at 1239 cm^{-1} that can be assigned to CO or CN groups [11] (Fig. 10). A strong absorption range at the 1698 cm^{-1} peak indicates an increase in the amount of carbonyl groups in the DMDHEU reagent [12]. Two N-methylol groups present in the DMDHEU chemical finishing reagent are capable of reacting with the hydroxyl groups of the BC cell wall polymer. After treatment, the number of waves of this peak decreased from 1731 cm^{-1} to 1698 cm^{-1}. This can be explained by the hydrolysis of ether groups in condensed DMDHEU reagent. These three main peaks lead to important changes in the chemical structure of the BC samples. Based on the obtained results, it can be stated that DMDHEU chemical reagent was detected in the FTIR curves, and cross-links were formed between the BC film and DMDHEU agents of the chemical reagent.

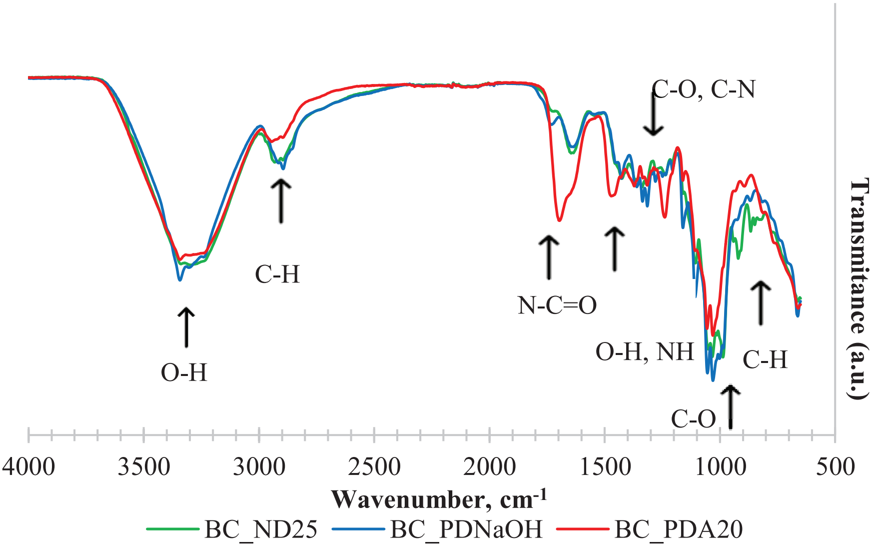


Fig. 10. FTIR reflectance spectrum of samples BC_ND$_{25}$, BC_PD$_{NaOH}$, and BC_PDA$_{20}$

Based on the results of the XRD analysis, it can be stated that the crystallinity of the samples treated with the chemical reagent DMDHEU was 10.0% (Table 10), i.e., 5 times smaller than the washed BC film samples (BC_PD_{NaOH}) and 3 times smaller than the untreated samples (BC_ND_{25}), indicating that the amorphous phase predominates in the samples after the treatment with chemical reagent.

Table 10. XRD parameters of BC samples

Parameter	**BC_ND_{25}**	**BC_PD_{NaOH}**	**BC_PDA_{20}**
Crystalline area	9203.55	17495.85	223.71
Amorphous area	22420.28	19794.98	2098.88
Degree of crystallinity, %	29.10	46.91	9.63

The purification process of the BC material is one of the most important factors in improving the tensile strength of BC material (Fig. 11). The tensile strength of the samples increased by 52.0% after the purification with NaOH solution. Unfortunately, washed BC material lost its ability to deform, and strain decreased by 93.0%. It can be related to the removal of impurities and sugars or amorphous parts from the cellulose network; BC fibrils obtained a better interaction with one another, as it has been noticed after FTIR analysis.

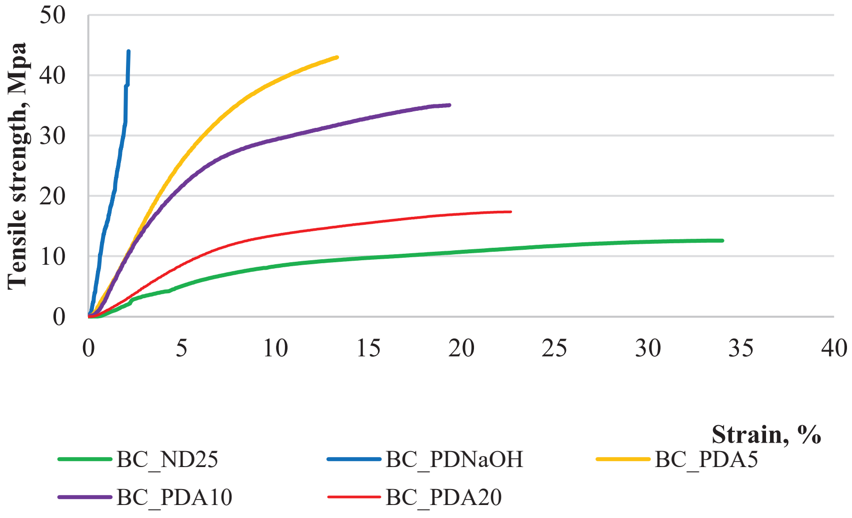


Fig. 11. Tensile strength and strain curves for BC samples

The loss of tensile strength due to the presence of DMDHEU was 4% when treated with 5.0% of DMDHEU, 8.0% when treated with 10.0% of

DMDHEU, and 49.0% when treated with 20.0% of DMDHEU, compared to the purified samples. SEM and FTIR analysis have affirmed that applied treatment changed the structure of BC material by creating the cross-linking of cellulose, and the deformational properties of the material were improved. Since 15.0% concentration of catalyst (Mg^{2+}) was the same in all solutions with DMDHEU, the tensile strength reduced with the increase of DMDHEU treatment. It might be explained: DMDHEU molecules can penetrate within the cellulose structure, cross-link with the hydroxyl groups in the amorphous region and thus reduce the slipping of cellulose chains.

The thickness of dry samples after the washing process is 74.0% lower than that of dry but unwashed samples (Table 4). The reason for this, as shown by the results of the studies obtained by SEM, FTIR, and mechanical characteristics, may be the efficient washing of impurities from the BC film. After the treatment of the BC film with DMDHEU chemical reagent and drying of samples, it has been observed that the thickness of the samples increases with the increasing concentration of the used solution. At a DMDHEU concentration of 5.0%, the thickness of the samples increases by 34.0%, with 10.0% by 162.0%, and with 20.0% by 293.0%.

Table 11. Thickness and water barrier properties of BC samples

Samples	Thickness, mm	WVTR, $g/h \times m^2$	WVP, $g \times mm/m^2 \times kPa \times h$	WAC, %	Contact angle, °
BC_ND_{25}	0.674	9.864	0.049	92.92	63.41±3.24
BC_PD_{NaOH}	0.178	7.408	0.009	171.23	87.60±3.84
BC_PDA_5	0.238	18.82	0.034	89.98	77.40±2.76
BC_PDA_{10}	0.466	21.365	0.075	18.98	68.56±3.59
BC_PDA_{20}	0.700	20.669	0.108	18.08	63.52±4.16

As shown in Table 11, the WVP values of the washed BC samples decreased by 82.0%, compared to the unwashed BC film samples. These values varied from 0.049 $g \times mm/m^2 \times kPa \times h$ to 0.009 $g \times mm/m^2 \times kPa \times h$. Water vapor first penetrates through amorphous areas of cellulose; thus, the absence of amorphous areas can reduce WVP. The XRD results confirmed the absence of amorphous areas in the washed BC film samples, which supports the hypothesis that the alkaline solution used to wash the BC film eliminated the amorphous areas. Increasing the concentration of DMDHEU chemical reagent shows that the values of WVP increase as well, and in the case of 20.0% DMDHEU, even become higher than before the BC washing process. The results of the study show that washing the samples in NaOH solution improves the water resistance of the BC film matrix. One reason may be that cellulose fibers form new bonds with the oxygen groups of NaOH, and this strong structure can reduce the dispersion of water molecules in the sample. In samples using chemical treatment, water molecules can easily diffuse in H bonds with OH groups in polymer chains and form new bonds with the DMDHEU reagent matrix [13]. In the FTIR experiment, it was shown that chemical treatment of BC film samples with DMDHEU reagent reproduces amorphous areas of the BC sample; therefore, this phenomenon has an effect on the increase of WVP values. Due to the changed morphology of the samples with chemical treatment, it can be observed that the water vapor transmission rate (WVTR) increased significantly from 7.4 g/h×m^2 (BC_PD$_{NaOH}$) to the average of 20.0 g/h×m^2 for BC_PDA$_{20}$ samples.

Hydrophilic/hydrophobic character of the BC material was evaluated by the measurement of the contact angle (CA) of water on the surface (Table 11). It can be stated that the purification process was very effective for increasing the CA; after that, the purification parameter became 38.0% higher comparing to the untreated sample. Although with the higher concentration of DMDHEU, CA lowered from 77.40º (with 5% of DMDHEU) to 68.56º (with 10% of DMDHEU), and to 63.52º (with 20% of DMDHEU), which indicate the hydrophilic character of the treated BC material, which may depend on the appearance of the chemical treatment reaction with OH groups in BC material and the susceptibility of these groups in the amorphous regions within the BC chains along with the appearance of the functional groups, containing nitrogen after the treatment with DMDHEU. The purification of the samples improved WAC values up to 171%, or it was about 1.8 times higher than in the untreated samples. Obviously, the use of DMDHEU reagent for BC film treatment affected water absorption capacity. It has been observed that the decrease in VAG values of BC film samples may be related to the concentration of DMDHEU in the chemical reagent, where the values varied from 89.98% to 18.08%, respectively. Obviously, the addition of DMDHEU had an influence on water absorption capacity. The decreased WAC values of BC samples depended on the amount of DMDHEU treatment, and it changed from 90.0% to 18.0%.

Recommendations for further bacterial cellulose testing

The experimental studies confirmed a positive effect of chemical reagent on the properties of the BC film; therefore, the influence of the storage conditions and duration of the treated sample on the physical and morphological properties of the film should be investigated in order to apply it in the clothing industry.

CONCLUSIONS

1. The influence of used sucrose source on the yield and density of grown bacterial cellulose has been determined. The most efficient bacterial cellulose film grows in the medium using white sugar (BC_S_{BC}) and glucose (BC_S_{GL}): in both cases, the same BC film density (2.8 g/m^3) and yield (0.93 g/l) were determined. By changing the composition of the culture medium, the color of the BC film can be changed, but the formation of BC in the medium using green tea is more efficient (BC_T_{ZA} density 9.2 g/m^3 and yield 0.42 g/l), compared to the samples in which rosehip tea was used in the culture medium (BC_T_{KA} density 4.1 g/m^3 and yield 0.29 g/l) or beetroot juice (BC_T_{BC} density 3.5 g/m^3 and yield 0.35 g/l).
2. The storage conditions and duration have been found to affect the properties of untreated bacterial cellulose film. Bacterial cellulose samples at 23 ± 1 °C temperature and 60.0% humidity reduce the tensile strength and strain values of samples by 80.0% over 30 days (BC_SS_{N30}), whereas the storage of samples at 4 ± 1 °C at 80.0% humidity decrease the tensile strength and strain values of samples by 61.0% (BC_SS_{S30}). Thus, the BC film retains its mechanical properties better when stored in a cool environment.
3. The process of sewing BC film has been complicated by undesirable properties of the raw material such as tack, ductility, and wrinkling. The formed thread compounds have been obtained stronger than the BC film (tensile strength 14.8 MPa, strain 40.9%). When forming BC film bonding by self-adhesion, it has been found that the drying temperature affects the strength of the bond, and the values of the shear strength of the compound increase 6 times when the pressure of 0.06 MPa is used to join the samples. For the bonding of BC film samples using adhesive, it has been observed that the compounds formed at higher temperature (50 °C) are stronger than the bonded material and have better adhesive interaction: the shear resistance of the compounds is 43.37 MPa (BC_JA_{50}).
4. The parameters of the drying process were found to affect the physical properties of the untreated BC film. The drying of the BC film at lower temperatures helps to preserve the deformation properties of the material: at 25 °C, the elongation at break was 23.9% (BC_ND_{25}), at 50 °C, 18.8% (BC_ND_{50}), while the drying of the BC film at higher temperatures (75 °C), the material becomes hard (hardness (Shore A) 61.0%), and the worst deformation properties (elongation at break 4.5%) are determined (BC_ND_{75}).
5. The purification procedure of the BC film with 0.5% sodium hydroxide solution for 8 hours was found to be effective in washing away undesired fermentation impurities from the BC structure, which consists of a three-

dimensional network of ~ 100 nm diameter cellulose filaments (BC_PD_{NaOH}). The washed BC film is safe for contact with human skin (pH 7.2). The results of morphological studies confirm that after washing the BC film, the crystalline phase of the material increases from 29.0% to 47.0%, which changes the mechanical characteristics of the film: tensile strength increases 3 times, elongation decreases 6.5 times. After leaching sucrose from the amorphous areas of the BC film, the water vapor permeability in the samples decreases by 82.0%.

6. The treatment of the BC film with 1,3-dimethylol-4,5-dihydroxyethylene urea revealed cross-linking in the structure of the material, and a positive effect of the process on the physical properties of BC film was observed: using 20.0% 1,3-dimethylol-4,5-dihydroxyethylene urea, the elongation at break of bacterial cellulose samples increases by 90.0% (BC_PDA_{20}), compared to the washed BC film. Morphological experiments have shown that the use of a chemical reagent restores the amorphous areas of the bacterial cellulose film, resulting in a 92.0% improvement in water vapor permeability.

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REZIUMĖ

Tiriama problema ir darbo aktualumas

Šiuo metu tekstilės ir aprangos pramonė vis dažniau susiduria su darnios plėtros, tausojančio vartojimo ar alternatyvių medžiagų terminais. Paskutinį dešimtmetį mokslininkai ir dizaineriai skiria daug dėmesio aplinką tausojantiems produktams, kuria naujas medžiagas, kurios pasižymi išskirtinėmis funkcinėmis savybėmis ir yra pritaikomos ne tik tradicinėse srityse. Šie tyrimai glaudžiai susiję su senkančiais energetiniais resursais, ekologinėmis problemomis, pertekliniu vartojimu, dideliais atliekų kiekiais, oro tarša ir t. t. Tekstilės ir aprangos pramonė yra labai reikšminga pasaulio ekonomikai, todėl svarbu naudoti ir tyrinėti švaresnes, atsinaujinančias ir draugiškas aplinkai medžiagas. Viena iš jų – celiuliozė, kuri žinoma kaip gausiausias polimeras žemėje, kuris sudaro apie 1,5 x 10^{12} tonų bendros metinės biomasės, o celiulioziniai pluoštai pasižymi puikiomis mechaninėmis ir fizikinėmis savybėmis, tačiau jiems užauginti reikalingas dirvožemis, saulės šviesa, anglies dioksidas bei vanduo. O bakterinė celiuliozė yra unikali medžiaga, kuriai užauginti nereikalingas dirvožemis, saulės šviesa, pesticidai ar didelis vandens kiekis. Bakterinę celiuliozę galima išgauti visus metus fermentuojant specialioje terpėje, kuri gali būti gaminama panaudojant maisto bei gėrimų pramonės atliekas. Remiantis moksliniais tyrimais galima pastebėti, kad dėl išskirtinės sandaros ir unikalių savybių ši medžiaga puikiai integruojama į biomedicinos, popieriaus, maisto ir kt. pramones, vystomi projektai ir atliekami tyrimai, siekiant pritaikyti bakterinės celiuliozės plėvelę ir aprangos pramonei.

Bakterinės celiuliozės plėvelė galėtų būti puiki alternatyva sprendžiant tekstilės ir mados pramonės problemas. Bakterinės celiuliozės potencialą didina tai, kad ši medžiaga nėra naujiena kitose srityse ir, remiantis jau žinomais tyrimais, galima teigti, kad bakterinės celiuliozės gavimo technologija atitinka ekologijos ir tvarumo principus, neturi neigiamos įtakos aplinkai bei yra nekenksminga žmogaus sveikatai. Bakterinė celiuliozė pasižymi biologiniu skaidumu, tai glaudžiai susiję su atsakingu bei sąmoningu vartojimu. Jos naudojimas ekologiškos aprangos gamybai leistų užauginti tiek medžiagos, kiek reikia, o pasibaigus eksploatacijos laikui tokia apranga lengvai suirtų ar galėtų būti naudojama kaip kompostas, taip nesukurdama atliekų ir mažindama aplinkos taršą.

Nepaisant bakterinės celiuliozės plėvelės privalumų, jos naudojimas aprangos gamybai dar nėra plačiai ištirtas. Bakterinė celiuliozė yra tiriama ją modifikuojant ar naudojant kaip žaliavą, tačiau neapdorota bakterinė celiuliozė tyrinėta per mažai: nežinomas jos ilgaamžiškumas, technologinės, kai kurios fizikinės ar morfologinės savybės ir pan. Dažnai moksliniams tyrimams bei vykdomiems projektams bakterinės celiuliozės plėvelė užauginama standartizuotose terpėse, tačiau užaugintos *kombučios* gėrime jos savybės plačiai neištirtos. Pastarąjį dešimtmetį *kombučios* gėrimas tapo populiarus komercinis produktas, kurį gaminant paviršiuje susidaranti plėvelė, šalutinis produktas, išmetama į atliekas. Siekiant to išvengti, šias atliekas galima būtų tikslingai panaudoti, todėl svarbu ištirti bakterinės celiuliozės plėvelės, užaugintos *kombučios* gėrimo terpėje, charakteristikas, cheminio apdorojimo įtaką jos savybėms, fizikines savybes bei įvertinti jos pritaikymo aprangai gaminti galimybes.

Darbo tikslas – įvertinti neapdorotos bakterinės celiuliozės, gautos *kombučios* fermentacijos metu, pritaikymo aprangos pramonei galimybes ir nustatyti plovimo bei cheminio apdorojimo įtaką plėvelės savybėms.

Darbo uždaviniai:

1. Įvertinti neapdorotos bakterinės celiuliozės plėvelės fermentavimo sąlygų, džiovinimo temperatūros, saugojimo trukmės ir temperatūros įtaką jos savybėms;
2. nustatyti plovimo metodų ir trukmės įtaką bakterinės celiuliozės plėvelės morfologinėms ir fizikinėms savybėms;
3. nustatyti 1,3-dimetilolio-4,5-dihidroksietilenkarbamido koncentracijos įtaką bakterinės celiuliozės plėvelės morfologinėms ir fizikinėms savybėms.

Mokslinis darbo naujumas ir praktinė vertė

Pastaruoju metu dėl aplinkosauginių priežasčių ekologiškų medžiagų paklausa labai išaugo. Įmonės, dizaineriai bei mokslininkai intensyviai ieško alternatyvių medžiagų šaltinių bei jų pritaikymo būdų kasdieniam gyvenimui. Ne išimtis ir viena iš svarbiausių pramonės šakų ekonomikai – tekstilės ir aprangos pramonė, kurios susidomėjimas tvariomis ir draugiškomis aplinkai medžiagomis vis auga, todėl itin svarbu ieškoti ir tyrinėti medžiagas, dėl kurių būtų mažiau teršiama aplinka ir kurios galėtų pakeisti iki šiol naudojamą tradicinę tekstilę. Remiantis šiais svarbiais faktais galima teigti, kad disertacijoje pateikti aktualūs tyrimai, kurie prisideda prie visuomenei pristatytų projektų ir tęsia šią idėją. Disertacijoje pateikiama platesnė ir išsamesnė bakterinės celiuliozės plėvelės analizė, įvykdyti iki šiol neatlikti eksperimentai, kurie lyginami su kitų mokslininkų darbais. Bakterinės celiuliozės plėvelės, išgaunamos plačiai žinomo *kombučios gėrimo* natūralios fermentacijos metu, gamyba yra unikali. Žinoma, kad bakterinė celiuliozė pasižymi puikiomis fizikinėmis savybėmis ir galėtų prisidėti prie tvarių tekstilės ir aprangos pramonės medžiagų sąrašo. Tačiau būtent *kombučios gėrimo* terpėje užauginta bakterinės celiuliozės plėvelė dar nėra gerai ištirta ir trūksta tyrimų bei informacijos apie jos charakteristikas, kad būtų galima pritaikyti aprangos pramonei. Disertacijos tyrimų metu pateiktas neapdorotos bakterinės celiuliozės pritaikymo aprangos pramonei įvertinimas ir rekomendacijos: nustatytos fizikinės bakterinės celiuliozės plėvelės savybės, į kurios auginimo terpę buvo įterpiami skirtingi ingredientai, norint praplėsti plėvelės spalvinę gamą. Taip pat ištirtas bakterinės celiuliozės plėvelės plovimas ir nustatyta 8 valandų efektyvaus plovimo trukmė, naudojant 0,5 % NaOH tirpalą. Nustatyta, kad išplovus bakterinės celiuliozės plėvelę didėja jos kristalinė fazė, dėl to plėvelės stipris tempiant didėja 3 kartus, tačiau deformacija mažėja 6,5 karto. Laidumas vandens garams taip pat mažėja 82,0 %. Įvertinus eksperimentų rezultatus nustatytos efektyviausios neapdorotos bakterinės celiuliozės plėvelės saugojimo sąlygos vėsioje (+4 °C) aplinkoje bei tinkamiausia džiovinimo temperatūra, atsižvelgiant į plėvelės fizikines savybes (25 °C), pateikti tolesnių eksperimentų siūlymai. Disertacijos rengimo metu atlikti aktualūs tyrimai, analizuojantys bakterinės celiuliozės plėvelės apdorojimą, naudojantys tradicinį tekstilės cheminį reagentą 1,3-dimetilolio-4,5-dihidroksietilenkarbamidą, ir nustatyta, kad cheminis reagentas atstato bakterinės celiuliozės plėvelės amorfines sritis, dėl to gerėja deformacinių charakteristikų (90,0 %) ir laidumo vandens garams rodikliai (80,0 %). **Darbo aprobacija.** Disertacijos tema paskelbtos 5 mokslinės publikacijos, iš jų 3 publikacijos paskelbtos mokslinės informacijos instituto duomenų bazės „Clarivate Analytics Web of Science“ leidiniuose su citavimo indeksu, 1 publikacija mokslinės informacijos instituto duomenų bazės „Clarivate Analytics Web of Science“ leidiniuose be citavimo indekso, 1 publikacija konferencijų pranešimų medžiagose. Tyrimų rezultatai paskelbti 10 konferencijų pranešimų medžiagose.

Darbo sandara ir apimtis. Disertacija sudaryta iš įvado, 3 skyrių, išvadų, literatūros sąrašo (279 įrašai), publikuotų mokslinių darbų sąrašo, priedų. Disertacijos apimtis – 103 puslapiai, juose pateikta 41 paveikslas, 32 lentelės, 13 formulių.

IŠVADOS

1. Nustatyta naudojamo sacharozės šaltinio įtaka užaugintos bakterinės celiuliozės kiekiui ir tankiui. Efektyviausiai bakterinės celiuliozės plėvelė auga terpėje naudojant baltą cukrų (BC_S_{BC}) ir gliukozę (BC_S_{GL}) – abiem atvejais nustatytas vienodas BC plėvelės tankis (2,8 g/m^3), ir kiekis (0,93 g/l). Keičiant auginimo terpės sudėtį galima keisti BC plėvelės spalvą, tačiau BC formavimasis terpėje naudojant žalią arbatą yra efektyvesnis (BC_T_{ZA} tankis 9,2 g/m^3, kiekis 0,42 g/l) lyginant su bandiniais, kurių auginimo terpėje buvo naudojama kinrožių arbata (BC_T_{KA} tankis 4,1 g/m^3, kiekis 0,29 g/l) ar burokėlių sultys (BC_T_{BC} tankis 3,5 g/m^3, kiekis 0,35 g/l).
2. Nustatyta, kad saugojimo sąlygos ir saugojimo trukmė daro įtaką neapdorotos bakterinės celiuliozės plėvelės savybėms. Bakterinės celiuliozės bandinius laikant 23,0 ±1,0 °C temperatūroje, kaip drėgmė 60,0 %, per 30 dienų bandinių

stiprumo ir ištįsos vertės sumažėja 80,0 % (BC_SS_{N30}), kai tuo tarpu, bandinius laikant 4,0 ± 1,0°C temperatūroje, kai drėgmė 80,0 %, bandinių stiprumo ir ištįsos vertės sumažėja 61,0 % (BC_SS_{S30}). Taigi, BC plėvelė geriau išlaiko mechanines savybes saugoma vėsioje aplinkoje.

3. BC plėvelės siūlinio jungimo procesas buvo komplikuotas dėl nepageidaujamų neapdorotos medžiagos savybių tokių, kaip lipnumas, tąsumas bei raukšlėjimasis. Suformuoti siūliniai junginiai gauti stipresni už BC plėvelę (stipris tempiant 14,8 MPa, ištįsa 40,9 %). Suformuojant BC plėvelės junginius klijavimo būdu, nustatyta, kad džiovinimo temperatūra turi įtakos junginio stiprumui, o bandinių jungimui panaudojus 0,06 MPa slėgį junginio atsparumo šlyčiai vertės padidėja 6 kartus. BC plėvelės bandinių jungimui naudojant gamtinį adhezyvą – kleisterį, pastebėta, kad aukštesnėje temperatūroje (50 °C) suformuoti junginiai yra stipresni už jungiamą medžiagą ir pasižymi geresne adhezine sąveika - junginių šlyties atsparumas yra 43,37MPa (BC_JA_{50}).

4. Nustatyta, kad džiovinimo proceso parametrai daro įtaką neapdorotos BC plėvelės fizikinėms savybėms. BC plėvelės džiovinimas žemesnėje temperatūroje padeda išsaugoti medžiagos deformacines savybes: džiovinant 25 °C temperatūroje nustatyta trūkimo ištįsa 23,9 % (BC_ND_{25}), 50 °C temperatūroje – 18,8 % (BC_ND_{50}), kai tuo tarpu, BC plėvelę džiovinant aukštesnėje temperatūroje (75 °C), medžiaga tampa kieta (kietumas (Šoras A) 61,0 %) ir nustatomos blogiausios deformacinės savybės (trūkimo ištįsa – 4,5%) (BC_ND_{75}).

5. Nustatyta, kad 8 valandų BC plėvelės plovimas 0,5% natrio šarmo tirpale yra efektyvus - išplaunamos nepageidaujamos fermentacijos priemaišos iš BC struktūros, kurią sudaro trimatis ~100 nm skersmens celiuliozės gijų tinklas (BC_PD_{NaOH}). Plauta BC plėvelė yra saugi kontaktui su žmogaus oda (pH 7,2). Morfologinių tyrimų rezultatai patvirtina, kad išplovus BC plėvelę, medžiagos kristalinė fazė padidėja nuo 29,0 % iki 47,0 %, dėl ko keičiasi plėvelės mechaninių charakteristikų rodikliai – stipris tempiant padidėja 3 kartus, ištįsa sumažėja 6,5 karto. Iš BC plėvelės amorfinių sričių išplovus sacharozę, laidumas vandens garams bandiniuose sumažėja 82,0 %.

6. Nustatyta, kad apdorojant BC plėvelę 1,3-dimetilol-4,5-dihidroksietilenkarbamidu, medžiagos struktūroje susidaro skersiniai ryšiai ir pastebima teigiama proceso įtaka BC plėvelės fizikinėms charakteristikoms: panaudojus 20,0 % 1,3-dimetilol-4,5-dihidroksietilenkarbamido, bakterinės celiuliozės bandinių ištįsos tempiant rodiklis lyginant su plauta BC plėvele padidėja 90,0 % (BC_PDA_{20}). Morfologinių eksperimentų metu buvo įrodyta, kad cheminio reagento naudojimas, atstato bakterinės celiuliozės plėvelės amorfines sritis, ko pasekoje 92,0 % pagerėja ir laidumo vandens garams rodikliai.

PADĖKA

Visų pirma dėkoju savo mokslinei vadovei doc. dr. Jurgitai Domskienei už žinias, pagalbą, nesuskaičiuojamas konsultacijas, tikėjimą, palaikymą ir įgytą patirtį visais studijų metais.

Dr. Paulei Bekampienei už visapusišką pagalbą, skatinimą ir bendradarbiavimą, kurie padarė didelę įtaką mano tobulėjimui.

Dr.Ilzei Baltinai, Dušan'ui Kimmer'ui ir jų komandoms už pagalbą ir rūpestį stažuočių metu.

Dr. Juditai Puišo, kolegoms, maisto bei tekstilės institutų darbuotojams ir visiems su kuriais teko konsultuotis, kurie negailėjo savo laiko, vertingų pastabų bei patarimų.

Noriu nuoširdžiai padėkoti savo artimiausiems žmonėms už paramą ir skatinimą tobulėti, o labiausiai savo sūnui už supratimą ir palaikymą siekiant savo tikslų.

UDK 677.46(043.3)

SL344. 2021-03-02, 2,5 leidyb. apsk. l. Tiražas 50 egz. Užsakymas 58.
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