Kombucha-derived bacterial cellulose from diverse wastes: a prudent leather alternative

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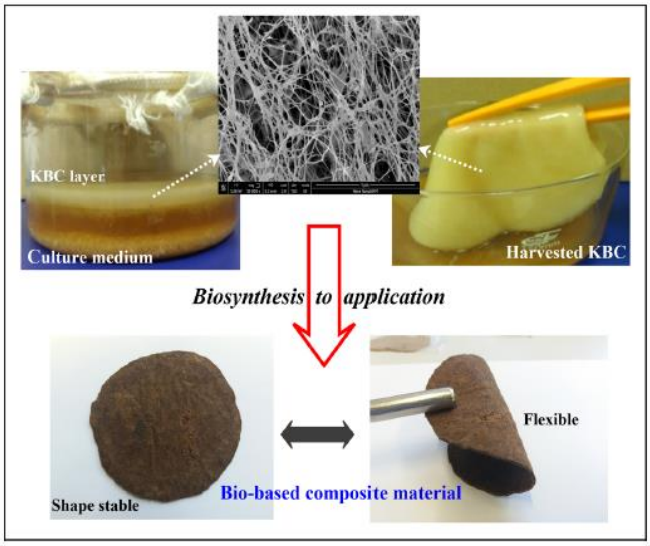
Abstract This study focuses on the production of kombucha-derived bacterial cellulose (KBC) from different modified kombucha and diverse bio-waste sources using *Komagataeibacter xylinus* for high potential towards cost-effective industrial scalability. The various nutrient media for the biosynthesis of KBC were formulated with sour whey, apple juice, and brewer's spent grains. Among the investigated nutrient sources, whey and whey/apple juice/brewer's spent grains mixture media showed superiority in the production of KBC with final yield dry weight determined as 12.59 and 12.81 g/L, respectively. The obtained KBC membranes were characterized by FTIR, TGA, SEM, and XRD analysis and results showed no significant difference in chemical structure, morphology, and thermal stability. The obtained KBC membranes were then further applied in fabrication of bio-based composites as prospective leather substitutes. The prepared bio-based composites showed good shape stability and considerable flexibility with an average tensile strenght and elastic modulus of ~ 1.69 Mpa and ~ 100 MPa, respectively. It can be inferred from the results in the present study that KBC can be used as a leather substitute in bio-textile fabrication.

Graphic abstract



Keywords: Biosynthesis • Bacterial cellulose • Biowastes • Leather-like composit

Introduction

Bacterial cellulose (BC) is an eco-friendly natural polymer produced from microbial organisms and has been widely considered as a material of the future due to its unique physicochemical and mechanical features. BC has been extensively applied in medicine as antimicrobial wound dressings (Portela et al. **2019**). In addition, this fascinating polymeric material has been used in blood vessel regeneration (Lee and Park **2017**), dental and oral implants, neural implants, urinary conduits, tympanic membrane (Lamboni et al. **2019**; Picheth et al. **2017**), bio-printing (McCarthy et al. **2019**), cosmetic ((Bianchet et al. **2020**), fabric (Yim et al. **2017**), leather (Garcia and Prieto **2019**; Ngwabebhoh et al. **2021**), textile (Fernandes et al. **2019**), paper (Skocaj **2019**), electronic devices (Xing et al. **2019**), environmental (Brandes et al. **2018**) and food packaging (Bandyopadhyay et al. **2020**; Patwa et al. **2019**). This wide application range is attributed to its three-dimensional fibrillar structure that incorporates high elasticity, durability, porosity, a high degree of crystallinity, biodegradability, non-cytotoxic and high thermal stability (Roman et al. **2019**).

Over the last decade, the biosynthesis of BC has been studied using different groups of microorganisms including *Achromobacter, Agrobacterium, Acetobac-ter, Gluconacetobacter, Rhizobium, Sarcina and Pseudomonas* (Augimeri et al. 2015; Halib et al. **2019**; Wang et al. **2019**) as well as cell-free BC synthesis via control enzyme systems (Kim et al. **2019**; Ullah et al. **2015**). The usage of microorganisms in the presence of potential nitrogen and carbon sources such as yeast extract, peptone, glycine, glucose, fructose, sucrose, mannitol and food derivatives, metabolize thereby increasing growth, development, and formation of gel-like cellulose membranes (Hussain et al. **2019**; Thorat and Dastager **2018**). Countless empirical results from Hestrin and Schramm (HS) standard medium (Hestrin and Schramm **1954**) and alternative mediums are proof of these indispensable requirements (Costa et al. **2017**; Ul Islam et al. **2017**). However, the high-cost of the nutrient sources used in the standard HS medium are one of the huge obstacles that limits commercial scalability (Azeredo et al. **2019**; Costa et al. **2017**; Garcia and Prieto **2019**; Jahan et al. **2018**; Ul-Islam et al. 2020; Ul Islam et al. **2017**). Recently, alternative sources including pineapple, apple, pomegranate, muskmelon, watermelon, tomato, orange fruits, coffee husk, sugarcane molasses, distillery effluent including waste water of noodle processing have been investigated as an efficient and comparatively low-cost nutrient sources for the production of BC (Bandyopadhyay et al. **2020**; Hussain et al. **2019**; Jozala et al. **2015**; Ul-Islam et al. **2020**). For example, BC membranes have been biosynthesized from fermented beverage also known as 'kom-bucha fungus tea'. In this process, a symbiotic microorganism culture of acetic acid bacteria and yeast (SCOBY) was used to metabolize sugars and black tea to produce BC membranes described as kombucha-derived bacterial cellulose (KBC) (Leal et al. **2018**; Villarreal-Soto et al. **2018**). According to structural analysis, the produced KBC was of high-grade and possessed similar characteristics to pristine plant cellulose. In addition, the produced KBC was confirmed to be free from contaminants such as lignin or hemicellulose and demonstrated good biological compatibility with no histologic and hematologic toxic effects on tissue cells (Jayabalan et al. **2014**). Moreover, the production process was simple, chemical-free, and only requires a short-term fermentation of sugars and tea. As such, the usage of bio-wastes from food processing industries or agriculture as nutrient source, significantly reduces the cost of BC production and also help in the direct treatment of large quantities of waste generated daily by dairy industries, which indirectly decreases environmental pollution and supports sustainability (Hussain et al. **2019**; Wang et al. **2019**).

Whey, a by-product of the dairy industry is produced in an annual capacity of 145 million tons worldwide of which 75 million tons is generated by European countries (Macwan et al. **2016**; Rosa **2018**). This by-product considered as waste contains large amounts of nutrient ingredients such as protein, water-soluble vitamins and minerals (Macwan et al. **2016**) and presently, just 50% is used to

produce bacteria culture fermentation medium, animal feed, protein isolate and concentrates (Bekatorou et al. **2019**; Macwan et al. **2016**; Revin et al. **2018**; Salari et al. **2019**; Semjonovs et al. **2017**). Thus, possesses great potential as a nutrient source for BC biosynthesis. Apple is a class of fruits that contains huge amount of sugars, proteins, fiber, vitamins, organic acids, minerals, and polyphenols (Shahidi and Alasalvar **2016**). Globally, this fruit generates large amounts of waste annually from spoilage, inventory and wild non-edible species to processing, making it highly suitable as a raw material for BC production (Bandyopadhyay et al. **2018**; Hussain et al. **2019**; Sagar et al. **2018**; Semjonovs et al. **2017**). Another potential bio-waste source includes brewer's spent grains which according to recent counts in the European countries, an estimated amount of over 9500 breweries are produced daily (Grave **2018**) where for every 5 L of beer produced, 1 kg of grains is generated as waste. This waste is rich in protein, fiber, vitamins, minerals, amino acids, phenolic compounds, oligosaccharides, and polysaccharides that may serve as suitable nutrient sources (Cooray et al. **2017**; Lynch et al. **2016**).

In the present study, whey, apple fruit juice, and brewer's spent grains were used as inexpensive and eco-friendly nutrient sources for the scalable biosynthesis of KBC membranes by *Komagataeibacter xylinus* which is known as several other names, mainly *Acetobacter xylinum and Gluconacetobacter xylinus*. The morphological structure and physicochemical properties of the different KBC membranes produced were examined and compared to BC from standard HS medium. In addition, attempts towards application of KBC as an alternative leather substitute was investigated via the fabrication of bio-based composite materials and their properties evaluated as prospective nonwoven leather materials.

Materials and methods

Materials

Waste whey (supplied by Kromilk A.S, Czech Republic), brewer's spent grains (supplied by a Brewery Industry in Malenovice, Czech Republic) and apple fruits (collected from public garden parks near Tomas Bata University in Zlin, Czech Republic) were used as nutrient sources. Black tea and sunflower oil were purchased from a grocery store in Zlin, Czech Republic. D-glucose was supplied by Amersco LLC, USA. Disodium hydrogen phosphate dodecahydrate ($Na_2HPO_4.12H_2O$) and citric acid ($C_6H_8O_7$) were purchased from Penta s.r.o., Czech Republic. The strain used for the preparation of KBC known as *Komagataeibacter xylinus* (formerly called *Gluconacetobacter xylinus*) was purchased from Czech Collection of Microorganisms, Brno, Czech Republic. Sucrose, yeast extract, peptone, polycaprolactone (PCL) (Mn = 80,000 g/mol) and dichloromethane were supplied by Sigma-Aldrich (Darmstadt, Germany). Polylactic acid (PLA-4043D) was supplied by NatureWorks LLC (Ingeo®, USA). Glycerol was procured from Lachner Co. Ltd (Neratovice, Czech Republic). Polyvinyl alcohol fiber (commercial grade PVA grade: 55-88) was supplied by Kuraray Poval™ (Kamisu, Japan). All reagents were used without further purification.

Table 1 Compositions of formulated media for the production of KBC membranes

Index of medium	Compositions Whey (mL/L)	Apple juice (mL/L)	Brewer's spent grains (g/L)	Sucrose (g/L)	Black tea (g/L)
Test samples					
WST	500	–	–	100	6
AST	–	500	–	100	6
BST	–	–	100	100	6
WABST	250	250	100	100	6
Controls					
KOM	–	–	–	100	6
HS	20 g D-glucose, 5 g yeast extract, 5 g peptone, 8.6 g $Na_2HPO_4.12H_2O$ and 1.15 g Citric acid				

Table 2 Constituent components in the preparation of bio-based composites

Samples	KBC (% w/w)	Plant leaf pulp (% w/w)	PVA (% w/w)	Glycerol (% w/w)	Sunflower oil (% w/w)	PCL (% w/w)
KBC-1	25	10	15	5	10	35
KBC-2	35	10	15	5	10	25

Activation of bacterial strain

The bacterial strain Komagataeibacter xylinus (CCM3611) was preserved in the Microbiology Laboratory of the Centre for Polymer Systems, Zlin, Czech Republic. Prior to usage, the bacterial strain was cultured on HS medium (Hestrin and Schramm **1954**) at 30 °C for a period of 3 days for activation. The bacteria strain was then inoculated in the various nutrient media for the production of KBC.

Biosynthesis of kombucha-derived bacterial cellulose (KBC)

In brief, sour whey was prepared in the pH range of 5.5-5.8 and kept at a temperature between 4 to 6 °C. Apple fruits were crushed using a juice blender (Guzzanti GZ 020, Italy) and the juice extracted (solution pH 3.8-4.0). Brewer's spent grains were pureed using a Nutribullet blender (N17-0908 machine, USA). Black tea was soaked in 200 mL of boiled distilled water for 10 min to obtain the extract. Subsequently, five different culture media were prepared following the mixture compositions in **Table 1.** Typically, each of the cultured media was contained in a round jar of 150 mm diameter followed by sterilization at 121 °C in an autoclave for 15 min. The different sterilized media were then cooled to room temperature and 1% (v/v) suspension of *Komagataeibacter xylinus* (CCM 3611) with an approximate initial bacteria density of 1.78 x 10^{10} CFU/mL (as prepared in 2.2.1) was added separately. The mixtures were masked using cleaned textile materials and statically incubated at 30 °C for 15 days. After the incubation stage, the cultured KBC and BC (as control) membranes on the surface of each medium were harvested, treated with 0.5% (w/v) NaOH for 1 h, washed with distilled water to neutral pH, and freeze-dried at — 110 °C for 24 h. The obtained freeze-dried samples were then stored in a desiccator for further analyses.

Fabrication of bio-based composite material (Bioleather)

In order to depict the potential of KBC as a suitable component for leather substitute production, two different composite samples of varying KBC ratios were formulated as shown in **Table 2**. Glycerol, maple plant leaves pulp (MLP), epoxidized sunflower oil, polyvinyl alcohol fiber, and KBC were mixed using a micro ball mill (Lab Wizz 320, Laarmann Group, Netherlands) for 1 min at room temperature. According to some previous studies, the blending of PCL with other biodegradable polymers like polylactic acid and PVA (Akos et al. **2013**; Bartnikowski et al. **2019**; Ngwabebhoh et al. **2020**; Saba et al. **2017**) generates good surface adhesion between the bio-filler particulates and the polymer matrix. In addition, components such as glycerol and epoxy vegetable oils (Peng et al. **2020**; Wang et al. **2020**; Yin et al. **2021**; Yu et al. **2020**) have proven to significantly enhanced structural flexibility, mechanical stability and to a lesser extend hydrophobicity of composite materials. Thus, in the present study the ball mill mixture was blended with 20% w/v PCL dissolved in dichloro-methane to form a uniform paste. The mixtures were then compressed under a press force of 5 to 20 kN using 2 mm thick stainless molds to form bio-based composite sheets. The samples were subsequently cured at 120 °C for 10 min, and air dried for two days to constant weight. The samples were then stored for further evaluations.

Determination of produced KBC and pH value of the nutrient media

In order to assessed the final dry weight of produced KBC membranes per volume of nutrient medium used, the harvested membranes were washed with distilled water, oven-dried at 40 °C to constant weight, and weighted. The final dry weight of KBC was then determined using the following formula:

$$\text{Final weight} = \frac{\text{Dry amount of cultured cellulose(g)}}{\text{Volume of medium used(L)}} \quad (1)$$

The pH values of the different nutrient media used were determined at the beginning and end of the bacteria cultured time using an electronic handheld pH meter (Lovibond pH meter-445R, USA).

Scanning electron microscopy

The freeze-dried cellulose membranes and prepared composite materials were examined for their surface morphology structures under a scanning electron microscope (SEM, FEI^{TM}, Brno, Czech Republic) at an accelerating voltage of 5 kV. Prior to analysis, the JEOL JFC 1300 Auto Fine coater (Tokyo, Japan) was used to gold coat the samples' surface to enhance conductivity.

Fourier Transform Infrared Spectroscopy

The freeze-dried cellulose membranes and prepared composite materials were analyzed for their chemical structures via Fourier Transform Infrared Spectroscopy (FTIR) using a Nicolet iS5 spectrometer (Thermo Scientific, USA) attached with an attenuated total reflectance mode (iD5-Ge-ATR) assembly. The samples were scanned at a 4.0 cm^{-1} resolution using 64 scans in the wavenumber range of 400-4000 cm^{-1}.

Thermal analysis

The thermogravimetric analysis (TGA) curves of the freeze-dried cellulose membranes and prepared composite materials was recorded using TGA Q500 (TA Instruments, USA). The samples were heated from 25 to 600 °C under a nitrogen atmosphere at a flow rate of 40-60 mL/min and a heating/cooling rate of 10 °C/ min.

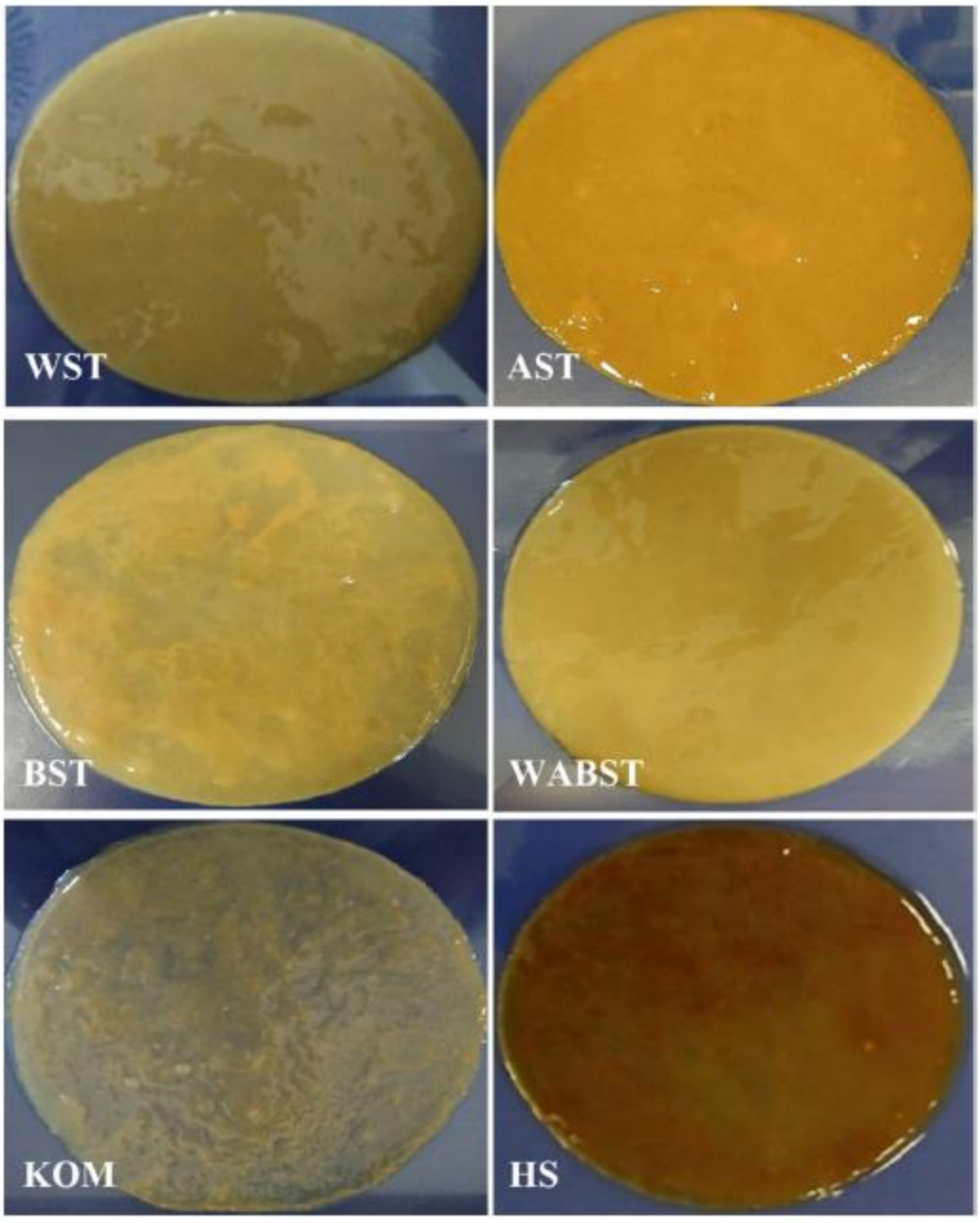


Fig. 1 Images showing physical appearance of cultured KBC membranes from the different nutrient media after 15 days

X-ray diffraction analysis

The crystalline structure of the freeze-dried cellulose membranes and other materials was measured by X-ray diffraction (XRD) analysis method on a Mini Flex^{TM} 600 X-ray diffractometer (Rigaku, Japan). Prior to analysis, a scan blank run was initially performed and used as the baseline. The samples (approximately 0.5 g) were tightly pressed to obtain films and were mounted onto a quartz substrate. The scans were performed in the range of 5°-45° at a scanning speed of 5°/min using a foil filtered CuKα radiation (λ = 0.1542 nm) at 40 kV voltage and a current of 15 mA. The divergence slit was maintained at 0.1 ° throughout the experiment. Iterations were repeated until the maximum F-number was obtained. During all measurements, the F-number was > 10,000 counts, which corresponds to an R^2 value of 0.997. The crystalline index (%) of analysed samples was calculated by dividing the diffractogram area due to crystalline region by the total area of the original diffractogram using the formulation below.

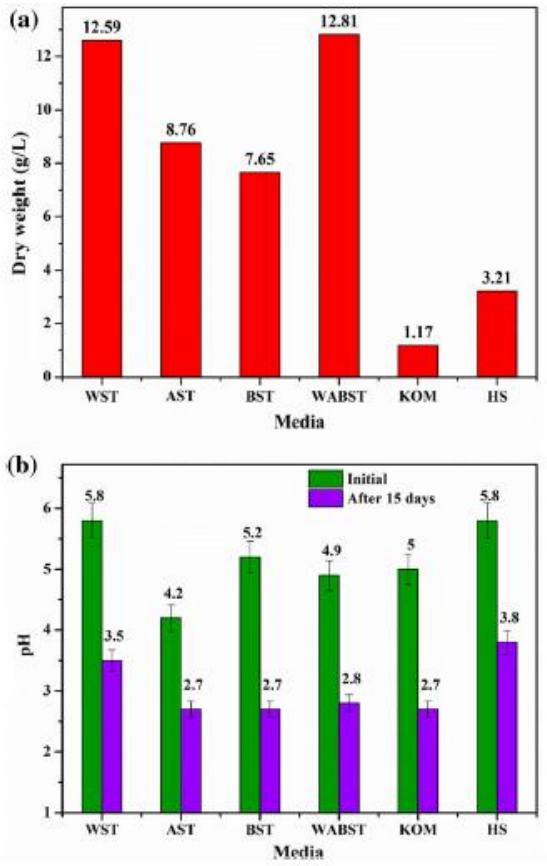


Fig. 2 a Dry weight of produced KBC membranes and b observed variation of pH in nutrient media (before and after the formation of KBC)

$$\text{Crystallinity index\%} = \frac{\text{Area of all the crystalline peaks}}{\text{Area of all the crystalline and amorphous peaks}} \text{x} 100 \quad (2)$$

Pore volume airflow rate Measurement and mechanical analysis of composite material

The porosity of the prepared biocomposites was investigated by measuring the pore volume in terms of airflow rate using a Bendtsen N3500 model porosity tester following the ISO 5336-2013 standard. Prior to analysis, the samples were cut in diameters of 40 mm and under constant pressure drop air was passed through a given area of the sample. The airflow rate per unit area was then recorded.

In order to investigate the mechanical properties of the fabricated bio-based composite materials, tensile testing was performed using Instron 5567 (Instron, USA) under a static load of 5 kg and a crosshead speed of 10 mm/min at room temperature (25 °C). The test was conducted following the ASTM D882 standards and the tensile strength as well as elongation at break as a function of displacement by force applied were determined. In addition, the storage modulus (E'), loss modulus (E'') and damping factor (tan delta) of the materials were analyzed using a DMA Q-800 dynamic mechanical analyzer (TA instruments, Delaware, USA) in the temperature sweep range from — 25 to 150 °C at a heating rate of 3 °C/min.

Statistical analysis

OriginLab software version 9.0 was used for statistical analysis. Analysis of variance (ANOVA) was applied for statistical evaluation and experimental results displayed as Mean ± Standard error where p <0.05 is determined as statistically significant.

Results and discussion

KBC production

Figure 1 depicts the cellulose membranes produced after 15 days from the different culture media. As observed, the colors of cultured KBC and BC membranes varied from light yellow-brown to dark brown, which is typical for bacterial cellulose, which is attributed to the composition of the culture medium. In both HS and investigated kombucha-based nutrient media, the development of cellulose membranes increased gradually with the increase of biomass of microorganism and cultured time/incubation period. The harvested cellulose membranes from the different test media (WST, AST, BST, WABST, KOM and HS) formed gel-like membrane layers on the top of their individual nutrient medium with thickness in the range of 0.5 ± 0.2 to 1 ± 0.3 mm. This confirms that all types of bio-waste sources used as a source of nutrient, supported cellulose synthesis by *Komagataeibacter xylinus*.

Figure 2a shows the obtained dry weight results of produced KBC from the various investigated media. Following the deduced results, whey containing media (WABST and WST) showed superiority in the amount of cellulose produced (12.831 and 12.593 g/L), respectively. However, the other nutrient media produced higher amounts of cellulose compared to HS (3.206 g/L) and KOM (1.171 g/L) media as controls.

Table 3 Comparison of produced cellulose with similar previous studies

Inoculum (Bacteria)	Medium	Incubation period (days)	Dry weight (g/L)	Reference
G. xylinus	Whey pretreatmented with β—Galactosidase	14	3.55	Salari et al. (2019)
K. rhaeticus	Whey was diluted and pretreatmented with β—Galactosidase	14	6.55	Semjonovs et al. (2017)
K. medellinensis	Whey without pretreatment	10	2.37	Molina-Ramirez et al. (2018)
K. rhaeticus	Apple juice diluted to initial sugar concentration of 20 g/L	14	9.49	Semjonovs et al. (2017)
K. rhaeticus	Hestrin and Schramm medium (HS) added glycerol	15	8.70	Thorat and Dastager (2018)
G. xylinus	Standard fructose medium (50 g/L fructose, 5 g/L peptone, 5 g/L yeast extract, 2.6 g/L Na_2HPO_4, 1.15 g/L critic acid)	15	2.20	Tian et al. (2018)
Kombucha symbiotic microorganisms	Black tea, glucose	20	13.30	Sharma and Bhardwaj (2019)
K. xylinus	Whey, sucrose, black tea (WST)	15	**12.59**	This study
K. xylinus	Whey, apple and brewed spent grain extract, sucrose, black tea (WABST)	15	**12.81**	This study
K. xylinus	Apple fruit extract, sucrose, black tea (AST)	15	**8.76**	This study
K. xylinus	Brewer's spent grains extract, sucrose, black tea (BST)	15	**7.64**	This study

The bold values represent the dry weight values to show the difference with other previous studies

The possible reason for the outstanding KBC production from whey medium compared to the others and previous reports (Jozala et al. **2015**; Molina-Ramirez et al. **2018**; Salari et al. **2019**; Semjonovs et al. **2017**), could be attributed to the efficient nutrient combination of whey and black tea. Nutrients such as protein (e.g. albumins, globulins), vitamins, amino acids, organic acids (e.g. lactic, succinic, propionic), caffeine and theophylline (Leal et al. **2018**; Macwan et al. **2016**) play great roles to enhance matter in bacterial cell respiration processes. For example, lactate stimulates the development of cells at an early stage of the culture process (Semjonovs et al. **2017**) due to succinic acid located in the tricarboxylic acid (TCA) cycle (Salari et al. **2019**; Semjonovs et al. **2017**) or caffeine and theophylline as stimulators of cellulose production by prevented bis-(3',5')-cyclic diguanosine monophosphate (c-di-GMP) in the cellulose synthesis cycle of bacteria (Sharma and Bhardwaj **2019**). As such, after adding the different bio-wastes in the media, the carbon source of the media improved the abundance glucose, fructose, galactose, arabinose, xylose, and lactose (Lynch et al. **2016**; Macwan et al. **2016**; Shahidi and Alasalvar **2016**; Xiros and Chris-takopoulos **2012**), which in tend caused a dramatic increase in the KBC biosynthesis process. Hence, this promotes the efficiency of *Komagataeibacter xylinus* in the presence of excess carbon sources.

It should be noted that the change of pH value of the culture medium also plays a critical role in cellulose growth, production ability, and membrane structure formation (Salari et al. **2019**; Ul Islam et al. **2017**). The optimum pH for cellulose membranes production normally depends on the variety of bacteria strains used but to some extent, the growth activity is also related to the slight acidic pH range (approximately 4-7) (Azeredo et al. **2019**; Wang et al. **2019**).

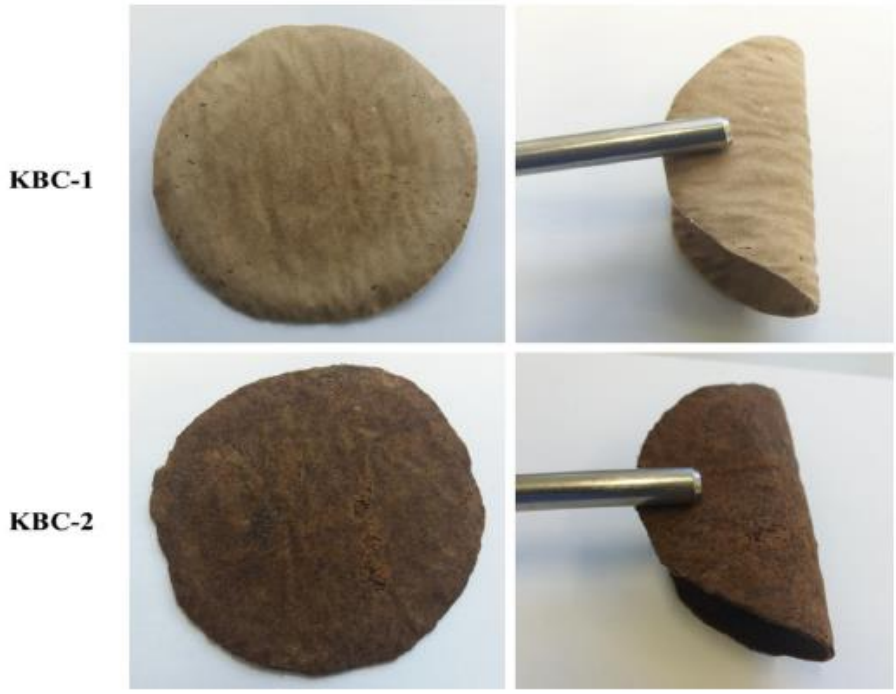


Fig. 3 Physical appearance of prepared bio-based composite materials

Figure 2b shows that the pH of different investigated media gradually decreased to acidic conditions at the end of the fermentation period. This decrease in pH of the culture media is attributed to the conversion of glucose and sucrose to acids such as gluconic and acetic acid (Jayabalan et al. **2014**; Leal et al. **2018**; Sharma and Bhardwaj **2019**; Wang et al. **2019**), which reduces the pH of various media at the end of the culture process. This decrease in pH tends to induce lack of oxygen in the cultured media leading to inhibition of cellulose growth and due to low activity of *Komagataeibacter xylinus* (Azeredo et al. **2019**; Leal et al. **2018**; Thorat and Dastager **2018**; Ul Islam et al. **2017**). Based on this, a favorable remedy may depend on the amount of added nutrients in the various media. That is the more nutrients are added into the medium, the slower pH decreases, which possibly could explain the superiority in KBC production achieved from media that contained whey.

Following a more detailed consideration, the lowest weighted results of produced cellulose membranes were obtained from KOM medium that contained just sucrose and black tea as the only carbon and nitrogen source. This low result may be due to priority activity of *Komagataeibacter xylinus* strains towards using glucose, fructose and galactose more than sucrose (Salari et al. **2019**; Thorat and Dastager **2018**). Another reason may be attributed to the low quantity of nutrients from black tea as compared to expensive chemicals used in HS medium. On the other hand, it is worth noting that the use other nutrient sources such as brewer's spent grains for the production of BC produced impressive results (7.65 g/L). Prior to preparation of culture medium, brewer's spent grains did not undergo any extraction or pretreatment process, which presented the simplicity of this method and the amount of cellulose produced was over 2 times higher than that cultured from standard HS medium. However, the inherent nutrient content of brewer's spent grains may be inadequate or not completely drained into the nutrient medium to completely stimulate strong cellulose biosynthesis capacity by *Komagataeibacter xylinus* compared to that observed for whey or apple juice extracts. Notwithstanding, the results obtained proved extremely feasible when compared to previous cellulose production studies as shown in **Table 3**. Overall, the highest amount produced in the present study

(12.59 g/L) was in close comparison to that reported by Bekatorou et al. **(2019)** as18.90 g/L. This confirms the high potential of cellulose production using the kombucha fermentation method.

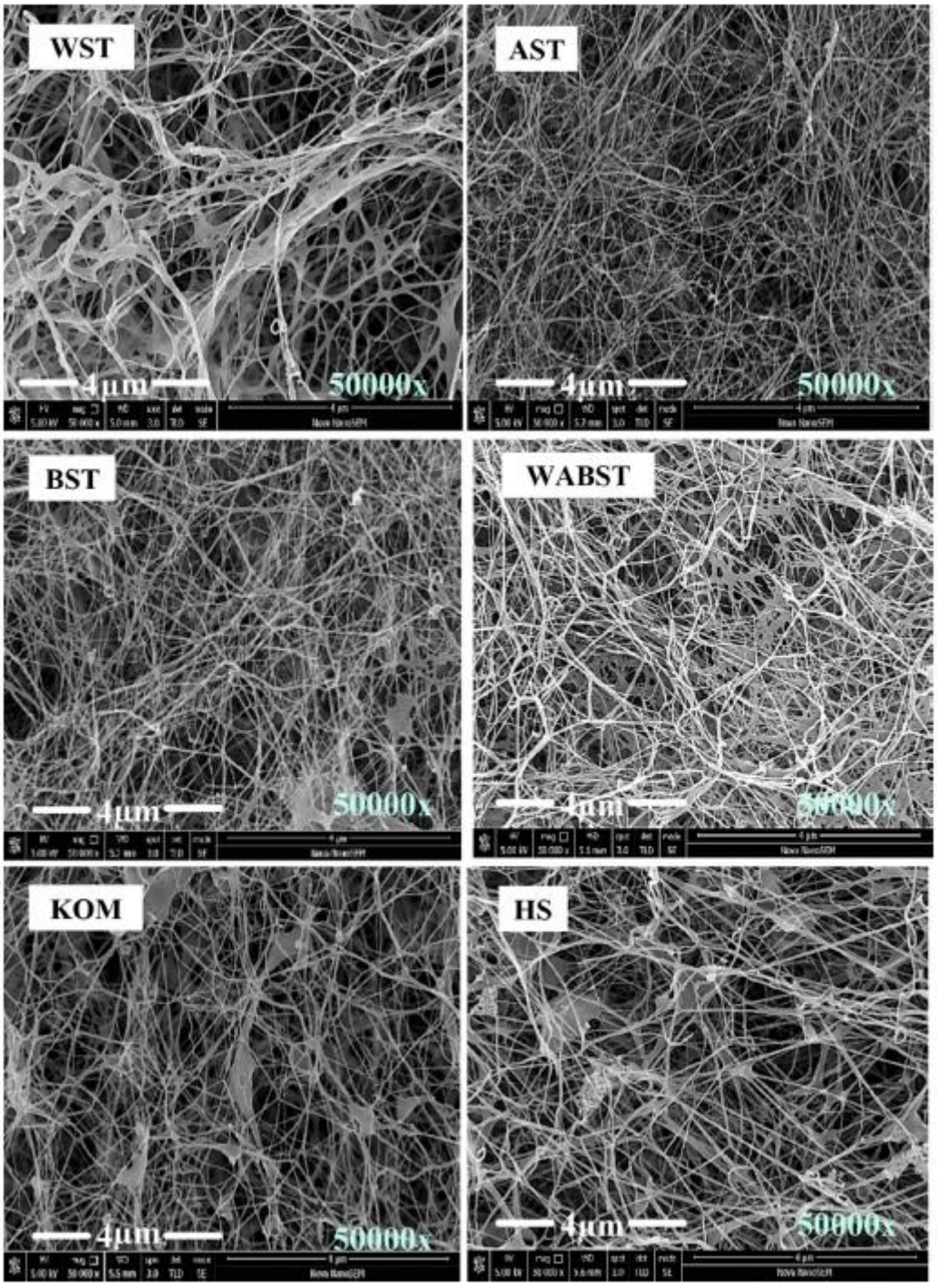


Fig. 4 SEM micrographs of cultured KBC membranes after 15 days at 50,000 x magnifications

Preparation of bio-based composite material (Bioleather)

The physical appearance of two prepared bio-based composite materials is depicted in **Fig. 3** Their thickness was within 1-2 mm and as can be seen, both of them were less stiff, considerably tough and flexible with opaque light brown (KBC-1) and dark brown (KBC-2) colors attributed to the amount of incorporated KBC in the materials.

SEM analysis

Figure 4 shows the SEM images of dried cellulose membranes from the different test media (WST, AST, BST, WABST, KOM and HS). According to observed morphologies, all KBC membranes depicted close similarity to the structure of conventional cellulose membranes from HS medium. In addition, the SEM micrographs showed no significant difference among the dimensions of obtained KBC nanofibers from the different investigated nutrient media. Based the obtained results, the fiber diameter for KBC membranes were in the range of 21 to 87 nm as compared to KOM medium and BC produced from HS medium that were between 45 to 93 nm. Generally, KBC consist of three-dimensional network spongy structures formed via random assembly of the rod-shaped nanofiber bundles and following the microstructural analyses of the images, provides evidence of strong interfacial adhesion between the KBC fibers and their dispersion. Similar cellulose fiber structures and dimension have also been investigated and reported by Salari et al. **(2019)**, Bekatorou et al. **(2019)**, Sharma and Bhardwaj **(2019)**.

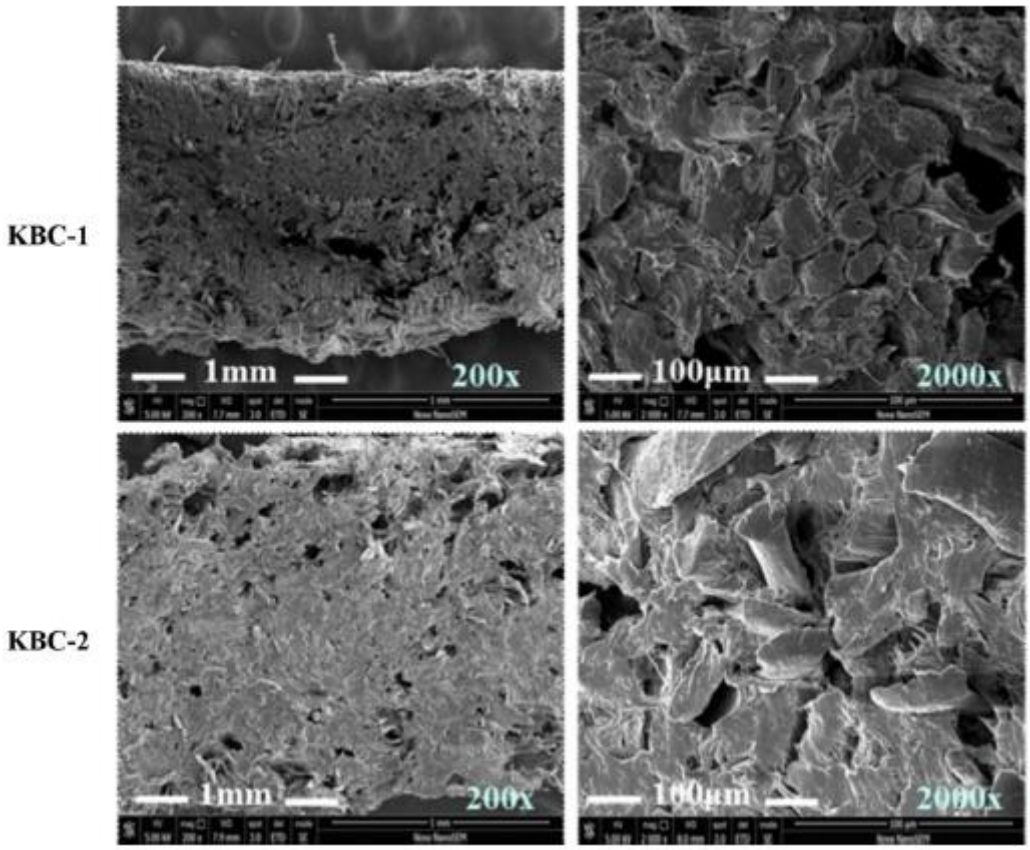


Fig. 5 a SEM micrographs of prepared bio-based composite materials (KBC-1 and KBC-2) at 200 and 2000 x magnifications

For the bio-based composite materials prepared, SEM analyses were performed to evaluate their structure and porosity. **Figure 5** depicts the crosssection morphology of both KBC-1 and KBC-2. As observed, the composites possessed a tight compacted structure that demonstrated a homogenous blending without particle agglomeration. It is clear that the biofillers were randomly dispersed in the matrix polymer indicating that the processing conditions used in the blending and molding steps were suitable. Pores were randomly arranged throughout the matrix of the prepared materials. Furthermore, the evaluation results with regards to the presence of free volume in the two samples (KBC-1 and KBC-2) showed close similarities in pore volume airflow rate determined as 782 ± 22 and 820 ± 43 mL/min, respectively. This analysis indicates that the bio-based composite materials prepared possess a high extent in porosity and breathable properties.

FTIR analysis

Figure 6 shows the FTIR spectra of investigated cellulose membranes and bio-based composite materials. According to cellulose prepared from HS medium, the peaks at 3390, 2895, 1405, 1159, 1068 cm^{-1} were ascribed to the characteristic bands of cellulose structure as previously reported, designated respectively to O-H stretching (3300 to 3400 cm^{-1}), C-H stretching (2800 to 2900 cm^{-1}), OH bending and C-O-H stretching (1430 to 1660 cm^{-1}), C-O-C stretching (1160 cm^{-1}) and C-O stretching (1035 to 1060 cm^{-1}) (Salari et al. **2019**; Sharma and Bhardwaj **2019**; Yim et al. **2017**).

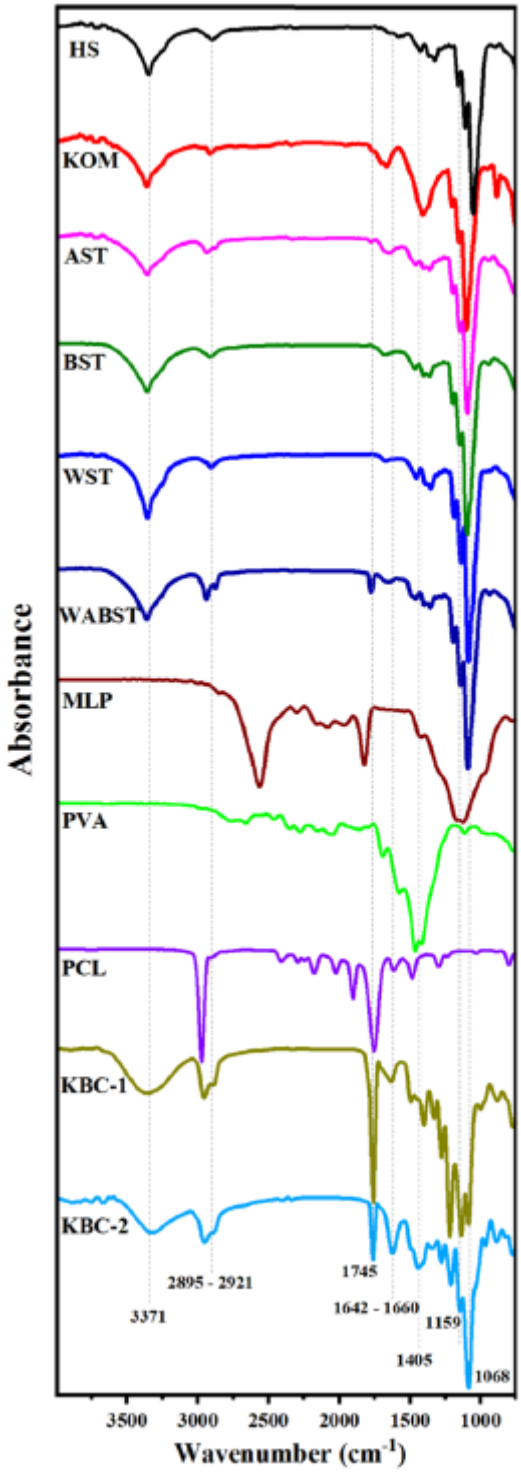


Fig. 6 FTIR spectra of KBC membranes and prepared biobased composite materials

Similarly, the spectra of KBC from the different nutrient media showed characteristic peaks at 3365 to 3431 cm^{-1} relating to O-H stretching vibration, 2921 cm^{-1} attributed to C-H stretching, 1642 to 1660 cm-1 ascribed to O-H bending, 1159 cm^{-1} for C-O-C, and 1065 cm^{-1} describing C-O vibration. All the spectra confirmed that the biopolymers produced by *Komagataeibacter xylinus* CCM 3611 were all pure cellulose. Close observation showed that except for KBC produced from the traditional kombucha

medium (KOM), the band between 3300 to 3400 cm^{-1} and 1068 cm^{-1} in the other KBC samples were more intense than that for BC from HS medium. This indicated that KBC from KOM media possessed stronger intermolecular hydrogen bonds. However, the displayed results are also attributed to the participation of the waste biological sources that increased the presence of impurities, causing a slight change in the FTIR spectrums of the KBC membranes from KOM media as compared to the FTIR spectrum of cellulose from HS medium. For the FT-IR analysis results of the prepared bio-based composite materials, the cellulose characteristic peaks from KBC and maple leave pulp were observed. In addition, the peak at 1745 cm^{-1} was ascribed to the C=O stretching vibration in neat PCL polymer, which was also observable in the KBC-1 and 2 blends. However, some of the peaks did not appear due to the effect of alkali treatment or thermal pressing that may have altered some chemical groups relating to hemicellu-lose, methyl, methylene, methoxy, or alcohol and carboxylic (Laaziz et al. **2017**; Reddy et al. **2016**).

TGA measurements

Figure 7 presents the thermogravimetric degradation curves of the cultured KBC membranes and bio-based composite materials following the percentage weight loss versus temperature in the studied range from 25 to 600 °C. The cellulose membranes obtained from the different culture media exhibited two phase degradation except for KBC from WST medium that showed an extra stage at 243.5 °C **(Fig. 7a**). This extra degradation phase may be related to the supplementary residues of whey in the cultured medium, which altered the molecular weight, crystallinity, and orientation of the nanofibers (Bagewadi et al. **2020**; Bekatorou et al. **2019**; Dorame-Miranda et al. **2019**). In general, the thermal stability of KBC produced from KOM media was higher than 200 °C and quite similar to that of cellulose obtained from HS medium. The first degradation phase from 25 to 230 °C was attributed to the weight loss of water molecules in the polymer matrix. In essence, this stage depicts no major difference in moisture content between the KBC samples. The second degradation phase occurred in the temperature range from 330 to 360 °C, which showed a significant decrease in mass ascribed to scissor and decomposition of the KBC polymer chains. The weight loss at this stage may be further related to the degradation, depolymerization, dehydration, or decomposition of the structural compositions of cellulose. **Figure 7b** shows the TG and DTG plots of the prepared bio-based composite materials. Based on obtained results, three degradation peaks were determined at 174 and 163 °C, 301 and 272 °C, and 369 and 361 °C for KBC-1 and KBC-2, respectively. According to observed results, the addition of KBC with increased the thermal stability of the blended materials, which may be attributed to inherent nature of KBC relating to poor heat-resistant components such as hemicelluloses and lignin (Jaya-balan et al. **2014;** Roman et al. **2019**; Villarreal-Soto et al. **2018**). However, an in-depth research is necessary to accurately determine the most effective KBC concentration for mixing with the different substrates toward optimizing the thermal stability of the materials prepared.

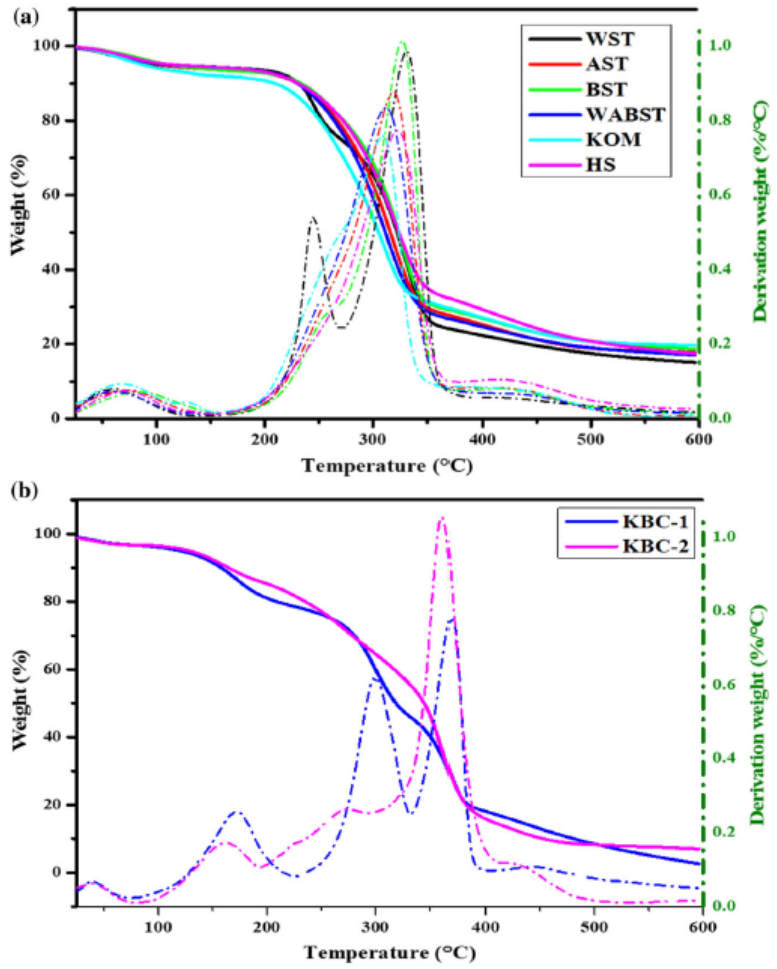


Fig. 7 TG and DTG plots of a KBC membranes obtained from different nutrient medium (WABST, BST, AST, WST, KOM, HS) and b prepared bio-based composite materials

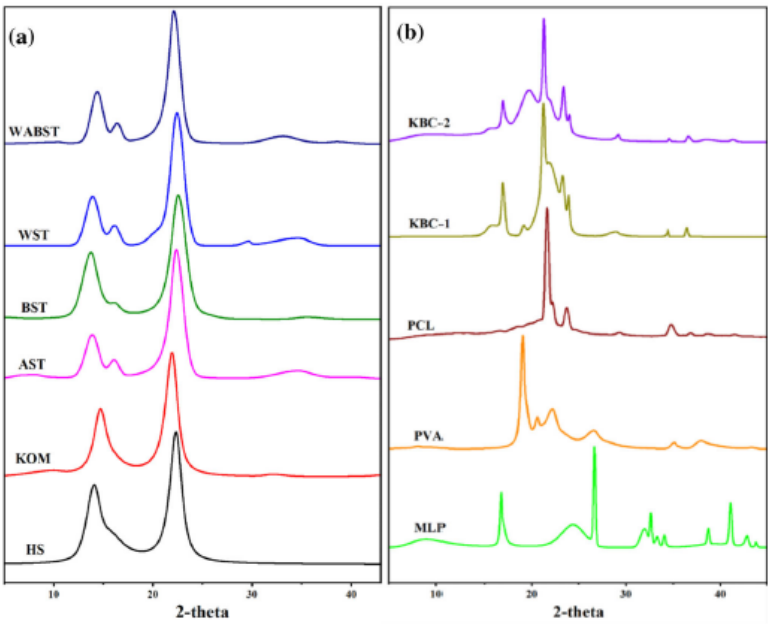


Fig. 8 X-ray diffractograms of a KBC membranes and b prepared bio-based composites

XRD analysis

X-ray diffraction was used to evaluate the crystalline structure as well as the change in crystallinity of the culture cellulose membranes and prepared bio-based composite materials. As shown in **Fig. 8a**, two peaks near ≈14.2° and ≈22.5° corresponding to Miller indices of (100) and (110) for cellulose Iα (French **2014**) were observed in all surveyed mediums and the standard HS medium. The (010) peak at about 16.2° is also visible on the AST, BST, WST, AND WABST patterns. The results obtained in this study were in close agreement with similar previous studies by Smarak Bandyopadhyaya et al. **(2018)** and Dorame-Miranda et al. **(2019)**. The crystallinity percentages for cellulose produced from the different media were obtained as 63.9%, 51.7%, 52.8%, and 59.7% for WST, BST, AST, and WABST, respectively, compared to the control samples determined as 51.6% for KOM and 65.8% for HS. The difference in the crystallinity degree for the various media was as a result of variation in the culture medium compositions or supplement-residues of bio-waste sources.

For the bio-based composite materials, the characteristic peaks of the pristine components are observed to be present in the prepared composites in **Fig. 8b**. The characteristic peaks of PCL (≈21.5°, ≈23.6°) (Borjigin et al. **2013**), PVA (≈ 19.2°) (Maet al. **2016**), and cellulose from MLP or KBC (≈ 14.2-16.8°, ≈ 22.5-23.9°) (Salari et al. **2019**; Yim et al. **2017**) are clearly displayed. Their insignificant difference is attributed to the effect of the pretreatment components, heat pressing process, or the combination of substrates causing a change in the material's structure Furthermore, similar results were observed in one of our previous studies that investigated the preparation of biocomposites from bacterial cellulose and plant leave pulp as potential non-woven fibrous materials (Ngwabebhoh et al. **2020**).

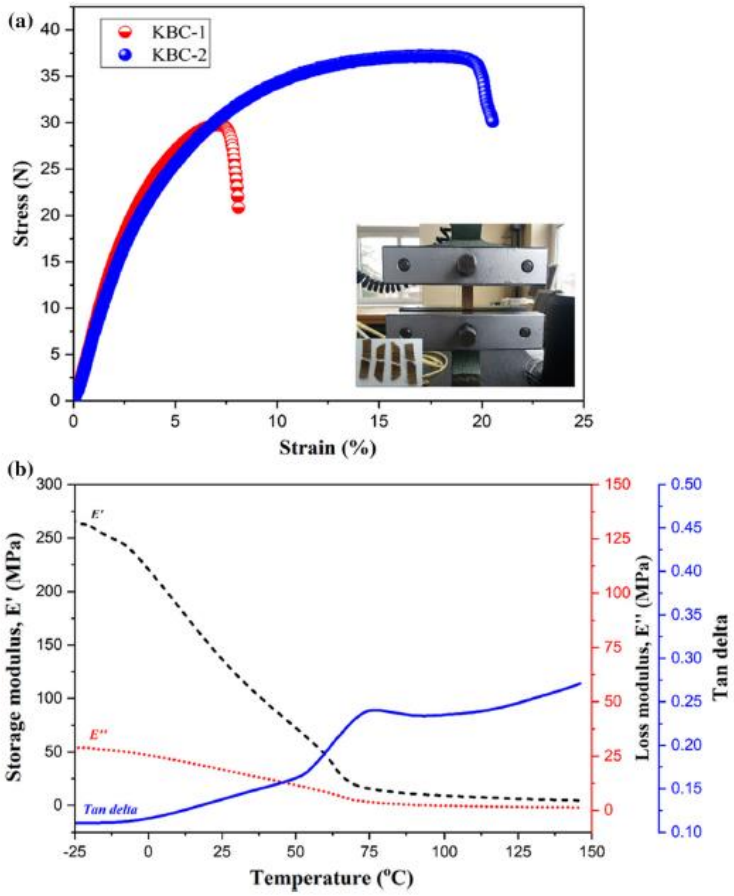


Fig. 9 Mechanical measurements of a tensile tests (KBC-1 and KBC-2) and b DMA thermograph of KBC-2 bio-based composite material

Mechanical analysis of bio-based composite material (Bio-leather)

The mechanical properties of the prepared bio-based composite material were based on analysis of tensile strength and elongation at break, as well as elastic and loss modulus as a function of temperature. **Figure 9a** shows the tensile analysis of the fabricated bio-based composite materials. An average tensile strength of 1.69 ± 0.33 MPa, tear strength of 25.44 ± 0.12 N/ mm and an elongation at break of 14.54% was recorded for KBC-2 as compared to KBC-1 which showed a lower value of tensile strength of 1.33 ± 0.16 MPa, tear strength of 20.41 ± 0.09 N/ mm and an elongation at break of 8.04%. As observed, an increase in KBC led to an increase in the strength of composite matrix. The higher tensile strength displayed by KBC-2 compared to KBC-1, was attributed to the reinforcing capacity cellulose incorporated into the material's matrix, which allows for the particulates, and polymer matrix components to be more compatible by forming good bonding interactions, leading to an improved 3D structure with enhanced mechanical strength (Sangregorio et al. **2019**). In addition, the increase in elongation may be attributed to the increase in interfacial interactions between the fibers and polymer matrix, which enhanced stress transfer efficiency of the interface. The principal reason for preparing such material in the present study was to demonstrate the potential prospects of KBC as a possible alternative for usage in artificial leather production with good textural appearance, tensile strength, flexibility and shape stability.

Figure 9b depicts the DMA analysis of KBC-2 as a function of temperature by measuring the storage (E') and loss (E'') modulus values. As observed, the value E' gradually decreased with increasing temperature from — 25 to 150 °C, which was attributed to the weakening of existing in the network assembly of the composite structural matrix. The further decrease in E' at a temperature greater than 60 °C, corresponded to the relaxation transition phenomena of the material's matrix. The Tg value of the material was determined in the range of 60 to 75 °C, which can be related to the Tg values of some of the components, such as PVA and PLA. The average E' and E'' of the KBC bio-based composite sample at body temperature (37 °C) were determined as 100.80 ± 0.71, and 15.16 ± 2.08 MPa, respectively. The low value determined was indicative that the prepared KBC composite material was flexible and less stiff, associated with the incorporation of KBC into the blend matrix and to a lesser extent the effect of other used components. The tan delta represents the loss factor and describes the damping within the composite material. The maximum peak in the tan delta curve is related to the damping contribution from the material's matrix and fibers, which falls in the range of the composite material Tg value, implying there is a strong contribution connecting the fibers and their adhesion with the matrix, probably also related to the heterogeneity in the KBC/biodegradable polyester ratio (Saba et al. **2016**).

Conclusions

Sour whey, apple juice, brewed spent grains were extensively effective in improving the production of KBC membranes with characteristics showing insignificant difference compared to cellulose produced from standard HS medium. KBC content used for the fabrication of bio-based composite materials showed positive effect in terms of tensile strength, leading to enhance elastic modulus or shape stability. The materials produced were considerably flexible demonstrating its potential and suitable component for the preparation of leather alternative in the fashion industry. Though more research investigation is still required, we believe that the cellulose (KBC) produced by the microorganism *Komagataeibacter xylinus* represents a good production source since it enhances sustainability, simplicity, and cost-effectiv-ity of the process to transform waste into useful products.

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