CHEMICAL ENGINEERING TRANSACTIONS
VOL. 86, 2021
A publication of AIDIC
The Italian Association of Chemical Engineering
Online at www.cetjournal.it
Guest Editors: Sauro Pierucci, Jiří Jaromír Klemeš

ISBN 978-88-95608-84-6; **ISSN** 2283-9216

The future of sustainable fashion: Bacterial Cellulose Bio Textile Naturally Dyed

Claudio J. Galdino S. Jr.[1,2,3], Alexandre D. M. Medeiros[1,2,3], Julia D. P. Amorim[1,2,3], Helenise A. Nascimento[2,4], Mariana A. Henrique[4], Andréa F. S. Costa[2,5], Leonie A. Sarubbo[1,2,3*]

[1] Northeast Biotechnology Network (RENORBIO), Federal Rural University of Pernambuco, Rua Dom Manuel de Medeiros, Dois Irmãos, Recife, Pernambuco, CEP: 52171-900, Brazil
[2] Advanced Institute of Technology and Inovation (IATI), Rua Joaquim de Brito, n. 216, Boa Vista, Recife, Pernambuco CEP: 50070-280, Brazil
[3] Science and Technology Center, Catholic University of Pernambuco, Rua do Príncipe, n. 526, Boa Vista, Recife, Pernambuco CEP: 50050-900, Brazil
[4] Center for Technology and Geosciences, Federal University of Pernambuco, Cidade Universitária, s/n, CEP: 50740-540, Recife, Brazil
[5] Communications and Design Center, Federal University of Pernambuco, BR 104, Km 59, s/n -Nova Caruaru, Caruaru, Pernambuco CEP: 50670-90, Brazil
*Leonie Asfora Sarubbo (Author for correspondence)
Tel.: +55 81 21194084; fax: +55 81 21194000.
E-mail: leonie.sarubbo@unicap.br

Fashion is considered to be one of the branches of world industry that most polluted and destroys the environment. Most of the damage caused by this segment comes from the production, manufacture, and synthetic dyeing of the fabrics. Regarding this concern, the usage of natural textiles, such as cotton, wool, and linen, has been growing in an attempt to reduce these impacts. However, even though these textiles are often considered ecologically sustainable, there is still great damage regarding their extensive land utilization, with effects that are often irreversible. Among the possibility of new textile biomaterials aiming at greater sustainability of the production chain, bacterial cellulose (BC) stands out, as a biotechnological polymer that has aroused industrial interest, due to its attractive properties for the sector. BC is a biodegradable polymer produced by microorganisms; therefore, it does not pollute the environment. Thus, the present study aimed to evaluate the use of BC as a bio textile, combined with natural dyes based on Eucalyptus (*Eucalyptus globulus L.*) and Onion (*Allium cepa L.*). The obtained BC membrane had high-water retention of 98.48 %. The behavior of dyed BC was compared to that of raw cotton and linen, which were also dyed with the same methods and dyes. Post-dyeing fabrics were evaluated for fixation and X-ray diffractometry (XRD). The pure matrices obtained a higher crystallinity index than after the dyeing process, demonstrating that the dye rearranged the matrices organization, resulting in better color fixation. Through the fixation tests, it was observed that the BC can be dyed effectively, like other natural textiles. Demonstrating that microbial biocellulose can also play the role of an attractive textile for the current industry with the importance of socio-environmental awareness.
Keywords: Bacterial Cellulose; Biotechnology; Natural Dyes; Fashion; Sustainability

1. Introduction

Sustainability appears as a new way of observing the world from new and better habits of production and consumption to protect the planet's resources and generations. It is a term that can and should be used by society and the food and beverage, civil construction, textile, and fashion industries (Brenot et al., 2019).
To achieve the proposed sustainable development goals, it is essential to obtain and dispose of natural resources consciously, which requires the use of new technologies. Among those that have the potential to

contribute to sustainable development, biotechnology has a lot to offer, especially in large-scale fields for the production of beverages, cosmetics, the textile industry, construction materials, among others. There is no doubt that biotechnology is the science that will revolutionize the production and supply of materials for the markets (Rizwan et al., 2018).

Among the possibilities of supplying new biotechnological materials, the highlight is obtaining vegetable cellulose, a biopolymer produced by plants. However, the expansion of demand for vegetable cellulose derivatives, the consumption of wood as a raw material has increased, causing global environmental issues, including deforestation. Although vegetables are the largest source of cellulose, several types of microorganisms are capable of producing cellulose as an alternative source. Bacteria belonging to different genera are capable of producing cellulose, known as bacterial cellulose (BC) or biocellulose (Gomes et al., 2013; Albuquerque et al., 2020).

Biocellulose has unique properties, such as biodegradability, biocompatibility and renewable character, resulting in a wide possibility of applications in the textile area, and several other technological domains, such as packaging, filter membranes, cosmetics, among others (Costa et al., 2017; Albuquerque et al., 2020; Amorim et al., 2020; Galdino et al., 2019; Galdino et al., 2020).

The potential of biocellulose goes beyond its existing applications, especially in large-scale production, using low-cost raw materials, including special textile materials, such as functional materials and even active packaging (Albuquerque et al., 2020). "Environmentally friendly" fashion or "sustainable clothing" is a global concern for both producers and consumers. BC is a renewable raw material that can serve as the basis for obtaining "green clothing" (Costa, Rocha & Sarubbo, 2017).

Finding sustainable raw materials and developing viable, conscious processes that reduce environmental impacts and have competitive advantages is a major challenge for the textile industry. Besides, the colors found in nature are crucial for the acceptance of textiles, adding aesthetic and symbolic value. With this in mind, the incorporation of the use of natural dyes for textile dyeing can be considered an extremely viable alternative

Natural plant dyes are considered safe for nature due to their non-toxic, non-cancerous, and biodegradable characteristics. Its use has enormous advantages, such as abundance in the environment and simple extraction methods (Boutrup & Ellis, 2019; Sharma, 2019).

As a result, the use of biotechnology and materials such as bacterial cellulose in industrial processes can have a positive influence on companies and consumers. The current trend of using biomaterials in fashion and clothing products is growing and offers perspectives on sustainable design that assimilates science and technology, aiming at social well-being and innovation. Fashion designers should look at biotechnology as a tool to be used as part of the creative process, and BC and natural dyeing as useful tools in designing their projects to better serve the garment industry in terms of quality innovative clothing, and increasingly trying to create textiles whose life cycle adapts to the needs of the increasingly conscious consumer (Carvalho & Santos, 2015; Costa, et al., 2019).

In this work, BC will be produced using *Camellia sinensis*, combined with natural dyes from Eucalyptus (*Eucalyptus globulus L.*) and Onion (*Allium cepa L.*) which will be tested after the purification process and will have their performance compared to natural textiles (raw cotton and linen). This work intends to show that bio textiles such as with BC, can serve as a basis for sustainable textiles that the fashion industry seeks. The combination of biodegradable and non-polluting products, such as vegetable dyes and microbial cellulose membranes, has great chances of being new biotechnological products that meet the needs of the market, which seeks safe and ecological options.

2. Materials and Methods

2.1 BC producing microorganisms and means of maintenance

For the production of the BC membrane, the microorganisms present in the Symbiotic Culture of Bacteria and Yeast (SCOBY) were used. According to Villarreal-Soto et al. (2018), the microbiological composition present in the consortium presents bacteria and fungi. Among the microorganisms, acetic acid bacteria (*Gluconobacter sp, Acetobacter sp.*), Lactic acid bacteria (*Lactococcus sp.* and *Lactobacillus sp.*), and yeasts (*Zygosaccharomyces bailii, Saccharomyces cerevisiae, Schizosaccharomyces pombe, Brett)* are found. The production medium consists of 10 g / L green tea (*Camellia sinensis*), 50 g / L of sucrose, 1.15 g / L of citric acid at pH 6.

2.2 Cultivation conditions, washing, purification, and yield of the BC membrane

BC's production in the culture medium was carried out by transferring 10% of a pre-inoculum containing the microorganisms present in the consortium in suspension to the 500 ml Schott flasks, containing 350 ml of the liquid tea medium. Cultivation was carried out statically at 30 ° C for 14 days. The cleaning of the membranes was carried out by washing in running water and purified by immersion in a 0.1 M NaOH solution for 1 h to eliminate the retained cells and to achieve color uniformity. Subsequently, the membranes were weighed to determine the yield.

2.3 Percentage of Water Retention (PWR)

The PWR is linked to the moisture content of the biomaterial, this analysis will determine its capacity to adsorb and fix the dyes. Five wet membranes of BC were weighed and dried in the oven to completely remove the water to constant weight. Then the PWR was obtained using Eq. (1):

$$PWR(\%) = \frac{Mean\ of\ the\ wet\ weights - Mean\ of\ the\ dry\ weights}{Mean\ of\ the\ wet\ weights} \quad (1)$$

2.4 Dyeing of BC membranes, raw cotton (RC), and linen (LI)

2.4.1 Dye extraction

For the extraction of the dye present in the plant extracts of *Eucalyptus globulus L.* (50 g of dry leaves of the plant) and *Allium cepa L.* (50 g of the bulb bark) consisted in an infusion at room temperature, in a solution composed of 1,0 L of deionized water and 0.25 L of 70 % ethanol. After 24 hours, the infusions were boiled for 30 minutes and filtered to remove the vegetable peels. The liquids were then used for dyeing.

2.4.2 Fiber preparations

A solution of deionized water with the mordant (potassium alum 99.5 %) at 20 g / L was prepared. Then, the BC membranes, raw cotton, and linen were submerged in the solution and heated for 30 min under agitation.

2.4.3 Dyeing and dye fixation procedure

The obtained volumes from the dye extracts were kept at 90 °C and used for dyeing 0.5 kg of BC fibers (wet mass), raw cotton, and linen. The fibers were submerged in the heated extracts for 1 h, with light agitation. After the dyeing process, the fibers dyed by *Allium cepa L* were washed in running water, and then taken to a fixative bath with 20 g of NaCl / L with water, for 30 min. The fibers dyed by *Eucalyptus globulus L.* were submitted to a post-bite bath composed of deionized water and iron sulfate monohydrate at 10 g / L for the effect of graphite staining. Then they were also washed in running water and submitted to a fixative bath of 20 g of NaCl / L. Afterwards, the fibers were dried at room temperature until a constant weight of each was obtained.

2.4.4 X-ray diffractometry (XRD)

Before and after dyeing, the fibers were scanned to identify the crystallinity index (C.I.%), X-ray diffractometer patterns of the membranes were measured with a Phillips X'pert MPD diffractometer, using CuKα radiation. The percentage of crystallinity index was measured as C.I. (Eq. 2), where I_{max} is the peak height at 2θ = 22.5° and I_{min} the minimum between the peak at θ = 22.5° and the peak at 2θ = 16.3° (Gomes et al., 2013).

$$PWR(\%) = \frac{l_{max} - l_{min}}{l_{max}} \quad (2)$$

3. Results and discussion

3.1 Production of BC Membranes

The average yield of BC hydrated membranes produced during 10 was 512.15 g/L of liquid medium, whereas, in terms of dry membranes, yield of 7.78 g/L was obtained after 14 days. The NaOH purification step favored color uniformity, removal of the metabolites, and possible residues from the culture medium that were adhered to the surface of the membranes.

3.2 Percentage of Water Retention (PWR)

The wet BC membranes were weighed and dried in the oven to completely remove the water to constant weight, the mass values were obtained and then the PWR was calculated as described in Table 1.

Table 1: Wet, dry weights and water retention percentage of BC membranes obtained after 14 days

BC	Mean	PWR
Wet weight (g)	179.25	98.48 (%)
Dry weight (g)	2.72	

The results confirm that the BC membranes present a high-water activity, as described by Costa et al. (2017), reaching over 98%, being this property one of the fundamental characteristics for its efficiency adsorb and fix the dyes.

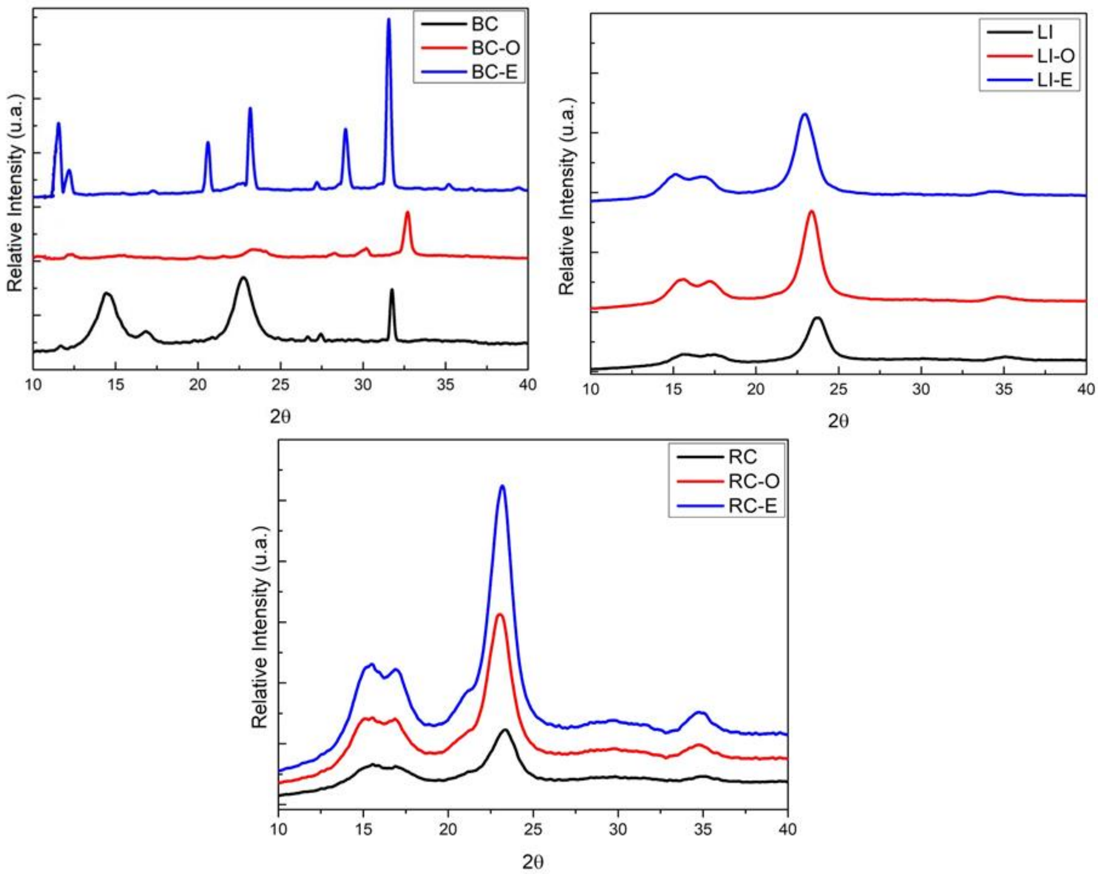
3.3 Dyeing of BC membranes, raw cotton, and linen

Figure 1 shows the matrixes analyzed before (a, b, and c) and after the dyeing process and by washing the fibers with neutral soap and drying in the shade (a', a”, b', b”, c', and c”). When looking at Figures 1a (LI) and 1b (RC), it can be seen that before dyeing processes, their matrixes color had a light beige hue, close to white, however, after processing, it was observed that *Allium cepa L*. colored the fibers of the materials with an earthy hue, close to brown (Figures 1a 'and 1b') and *Eucalyptus globulus L.* made the fibers obtain a graphite gray color (Figures 1a”, and 1b'').
Meanwhile, when analyzing the wet BC membrane, after washing, purification and neutralization, we can see that it obtained a uniform, beige appearance and smooth texture, and highly flexibility (Figure 1C). When the fiber is dipped in a dye bath, the dye is adsorbed to the surface and disseminates into the fiber. This phenomenon was observed in the BC membranes dyed with *Allium cepa L.* which obtained the mustard yellow color (Figure 1C ') and *Eucalyptus globulus L.* (Figure 1C ”) which obtained the graphite gray color.

Figure 1: Linen, raw cotton and Bacterial Cellulose (BC) membranes dyeing phases: A - Linen, A’ - Linen dyed with Allium cepa L., A” - Linen dyed with Eucalyptus globulus L., B - Raw Cotton, B’ - Raw Cotton dyed with Allium cepa L., B”- Raw Cotton dyed with Eucalyptus globulus L., C - BC , C’- BC dyed with Allium cepa L., C”- BC dyed with Eucalyptus globulus L.

All experiments showed good visual quality and uniformity in pigmentation, in addition to corresponding to a varied color chart that can be used in fashion products. Indicating the success of fixing the dyes in the fibers after the process.

3.4 X-ray diffractometry (XRD)
The CI was calculated based on the intensity of the peaks, as shown in Figure 2. Natural cellulose fibers consist of dense, organized crystalline structures and looser amorphous structures, which contain less ordered and organized chains. The higher the crystalline degree, the more difficult it is for dyes to penetrate in its matrix, meaning that the amorphous part facilitates the entry of the dyes. In some cases, where the fiber structure has a high CI, the aid of the mordants in dyeing, facilitates the penetration and fixation of the dyes (Boutrup & Ellis, 2019).



.

Figure 2: X-ray diffraction diagrams of natural Bacterial Cellulose (BC), natural Linen (LI) and Raw Cotton (RC) and after dyeing process with Allium cepa L. (-O), and Eucalyptus globulus L. (-E).

The dyeing process decreased the CI of the matrices, as shown in Table 2. The pure matrices obtained CI higher than after the dyeing process. According to Costa et al. (2019), this change in crystallinity can directly affect the mechanical properties of the membrane, reducing its mechanical tension and generating a more elastic material in comparison to pure BC. In this case, results show that dyed BC demonstrated a larger amorphous region, resulting in a more flexible polymer. This malleable property enables a better application of the BC as textile because it can better fit a variety of formats.

Table 2: Crystallinity index C.I. (%) of natural Bacterial Cellulose (BC), Linen (LI), and Raw Cotton (RC) and dyeing process with Allium cepa L. (-O) and Eucalyptus globulus L. (-E).

	BC	BC-O	BC-E	LI	LI-O	LI-E	RC	RC-O	RC-E
C.I. (%)	66,90	61,22	56,06	63,64	56,77	55,98	68,83	64,40	63,44

4. Conclusions

The use of biotechnology and materials such as BC in industrial processes can have a positive and sustainable influence on companies and consumers. The current trend of using biomaterials in fashion and clothing products is growing and offers perspectives on sustainable design that assimilates science and technology, aiming at social well-being and innovation. Although synthetic dyes are used more than natural ones, the latter are more environmentally friendly, less toxic, and less allergenic. The dyes used in this work can be successfully applied as pigments for BC membranes, controlling the conditions and supporting the cast used in the dyeing and fixation process. The obtained BC membrane had a crystallinity of 66,90 % and high-water retention of 98.48 %, being these properties, combined with the use of mordants, fundamental characteristics for its efficiency adsorb and fix the dyes. The combination of natural dyes and biodegradable fibers, such as BC membranes, can provide new biotechnological products that meet the needs of the world market, which seeks safe and environmentally friendly options for the chemicals used today. Studies of physicochemical characterization of the obtained fabrics and the improvement of the dyeing method will allow the obtention of a commercial product for the textile industry, as well as other sectors of the industry.

Acknowledgments

This study was funded by the following Brazilian funding agencies: Foundation for the Support of Science and Technology of the State of Pernambuco (FACEPE), the National Council for Scientific and Technological Development (CNPq), and the Coordination for the Improvement of Higher Education Personnel (CAPES).

References

Albuquerque R.M.B., Meira H.M., Silva I.D.L., Silva C.J.G., Almeida F.C.G., Amorim J.D., Vinhas G.M., Costa A.F.S., Sarubbo L.A., 2020, Production of a bacterial cellulose/poly(3-hydroxybutyrate) blend activated with clove essential oil for food packaging. Polymers and Polymer al Composites, 1-12.

Amorim J.D.P., Galdino C.J.S., Costa A.F.S., Vinhas G.M., Sarubbo L.A., 2020, BioMask, a Polymer Blend for Treatment and Healing of Skin Prone to Acne, Chemical Engineering Transactions, 79, 205-210, doi: 10.3303/CET2079035

Brenot A., Chuffart C., Coste-Manière I., Deroche M., Godat E., Lemoine L., Ramchandani M., Sette E., Tornaire C., 2019, Water footprint in fashion and luxury industry, Water in Textiles and Fashion, 95-113.

Boutrup J., Ellis C., 2019, The Art and Science of Natural Dyes: principles, experiments, and results, Vol 1, Schiffer Publishing, Pennsylvania, EUA, p.176.

Carvalho C. & Santos G., 2015, Global Communities, Biotechnology and Sustainable Design – Natural / Bio Dyes in Textiles, Procedia Manufacturing, 3, 6557-6564.

Costa A.F.S., Almeida F.C.G., Vinhas G.M., Sarubbo L.A., 2017, Production of bacterial cellulose by Gluconacetobacter hansenii using corn steep liquor as nutrient sources, Frontiers in Microbiology, 8, 1-12.

Costa A.F.S., Rocha M.A.V., Sarubbo L.A., 2017, Bacterial cellulose: an ecofriendly biotextile, International Journal of Textile and Fashion Technology, 7, 11–26.

Costa A.F.S., Amorim J.D.P., Almeida F.C.G., Lima I.D., Paiva S.C., Rocha M.A.V., Vinhas G.M., Sarubbo L.A., 2019, Dyeing of bacterial cellulose films using plant-based natural dyes, International Journal Of Biological Macromolecules, 121, 580-587.

Galdino C.J.S., Maia A.D.M., Meira H.M., Souza T.C., Amorim J.D.P., Almeida F.C.G., Costa A.F.S., Sarubbo L.A., 2020, Use of a bacterial cellulose filter for the removal of oil from wastewater, Process Biochemistry, 91, 288-296.

Galdino C.J.S, Meira H.M., Souza T.C., Amorim J.D.P., Almeida C.G., Costa A.F.S., Sarubbo L.A., 2019, Evaluation of the Potential of Bacterial Cellulose in the Treatment of Oily Waters, Chemical Engineering Transactions, 74, 313-318, doi: 10.3303/CET1974053

Gomes F.P., Silva N.H.C., Trovatti E., Serafim L.S., Duarte M.F., Silvestre A.J.D., Pascoal Neto C., Freire C.S.R., 2013, Production of bacterial cellulose by Gluconacetobacter sacchari using dry olive mill residue, Biomass and Bioenergy, 55, 205 – 211.

Rizwan M., Mujtaba G., Memon S.A., Lee K., Rashid N., 2018, Exploring the potential of microalgae for new biotechnology applications and beyond: A review, Renewable and Sustainable Energy Reviews, 92, 394-404.

Sharma K., 2019, Developments in the Process of Textile Dyeing: a review, The Bombay Technologist, 66, 1, 25-30.

Villarreal-Soto S.A., Beaufort S., Bouajila J., Souchard J.P., Taillandier P., 2018, Understanding Kombucha Tea Fermentation: a review, Journal of Food Science, 83, 3, 580-588.