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Review

Biocellulose for Treatment of Wastewaters Generated by Energy Consuming Industries: A Review

Alexandre D'Lamare Maia de Medeiros [1,2], Cláudio José Galdino da Silva Junior [1,2], Julia Didier Pedrosa de Amorim [1,2], Helenise Almeida do Nascimento [2,3], Attilio Converti [4,*], Andréa Fernanda de Santana Costa [5] and Leonie Asfora Sarubbo [2,6]

[1] Rede Nordeste de Biotecnologia (RENORBIO), Universidade Federal Rural de Pernambuco, Rua Dom Manuel de Medeiros, s/n—Dois Irmãos, Recife 52171-900, PE, Brazil; alexandre_dlamare@outlook.com (A.D.M.d.M.); claudiocjg@gmail.com (C.J.G.d.S.J.); juliadidier@hotmail.com (J.D.P.d.A.)
[2] Instituto Avançado de Tecnologia e Inovação (IATI), Rua Potyra, n. 31, Prado, Recife 50751-310, PE, Brazil; helenise_almeida@hotmail.com (H.A.d.N.); leonie.sarubbo@iati.org.br (L.A.S.)
[3] Centro de Tecnologia e Geociências, Departamento de Engenharia Química, Universidade Federal de Pernambuco (UFPE), Cidade Universitária, s/n, Recife 50740-540, PE, Brazil
[4] Department of Civil, Chemical and Environmental Engineering, Pole of Chemical Engineering, Genoa University, Via Opera Pia 15, 16145 Genoa, Italy
[5] Centro de Comunicação e Design, Centro Acadêmico da Região Agreste, Universidade Federal de Pernambuco (UFPE), BR 104, Km 59, s/n—Nova Caruaru, Caruaru 50670-90, PE, Brazil; andrea.santana@ufpe.br
[6] Escola Icam Tech, Universidade Católica de Pernambuco (UNICAP), Rua do Príncipe, n. 526, Boa Vista, Recife 50050-900, PE, Brazil
* Correspondence: converti@unige.it; Tel.: +39-10-335-2593

Abstract: Water and energy are two of the most important resources used by humanity. Discharging highly polluting wastewater without prior treatment is known to adversely affect water potability, agriculture, aquatic life and even society. One of the greatest threats to water sources are contaminated effluents, which can be of residential or industrial origin and whose disposal in nature must comply with specific laws aimed at reducing their environmental impact. As the oil industry is closely related to energy consumption, it is among the sectors most responsible for global pollution. The damage caused by this industrial sector is present in all countries, whose legislations require companies to carry out wastewater treatment before disposal or recycling in their production process. Bacterial cellulose membranes have been shown to be efficient as filters for the removal of various contaminants, including biological and chemical agents or heavy metals. Therefore, their use could make an important contribution to bio-based technological development in the circular economy. Moreover, they can be used to produce new materials for industry, taking into consideration current environmental preservation policies aimed at a more efficient use of energy. This review aims to compare and describe the applications of cellulose membranes in the treatment of these effluents.

Keywords: nanocellulose; biotechnology; oleophobic filter; oily effluents; fashion industry effluents

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1. Introduction

Sustainable development is of utmost importance for the planet, so the use of new technologies should be done with awareness and recycling natural resources. That said, biotechnology has been gaining considerable attention in the research community [1]. The development and implementation of industrial biotechnology are linked to the need to reduce the effects of the environmental changes and the depletion of fossil assets. At the same time, they are expected to provide socioeconomic benefits through the generation of new products and competitive processes [2,3], some fields of application of which are energy and wastewater treatment [4].

Water resources are essential for industrial activities, energy production, agriculture, and life on earth [5]. In particular, the access to potable water and efficient treatment methods are essential for the prevention of various types of pollution and waterborne diseases [6]. It is possible to reduce the load of pollutants through the interconnection of different industrial sectors, so that their by-products are treated and reused, and waste production is minimized, with the perspective of becoming raw material in a new production cycle [7].

As for energy production processes, in many cases they are directly linked to the availability of clean water, the obtaining of which requires energy, thus configuring an interdependence process. Furthermore, the use of water for energy production, both for industrial and/or residential applications, results in the pollution of water, whose treatment requires additional energy, causing a vicious circle [8]. Every day, huge amounts of emulsified wastewater are generated worldwide by the petrochemical and other industries at various stages (transportation, maintenance, manufacturing, etc.) and by oil spill accidents [9], which are among the main causes of aquatic environment pollution [10].

Industrial pollutants such as dyes, synthetic chemicals, heavy metals, oils, microplastics and others can have different origins and properties, and many of them accumulate in the environment over time, causing increasing damage [11]. According to Rajasulochana e Preethy [12], the methods of industrial wastewater treatment vary according to several factors, including volume, constitution of the effluent and limits imposed by environmental legislations. Increased research on renewable energy and energy saving technologies has favored the development of new processes and materials as alternatives to treat complex wastewater [10,13].

Many publications describe the application of membrane filtration for the treatment of wastewaters, especially the oily ones. Membrane technologies such as microfiltration, ultrafiltration and nanofiltration are increasingly used for the treatment and purification of wastewater and oily emulsions, as well as for the supply of clean water. However, the most commercially available membranes are made with synthetic polymers of fossil origin [14,15], which require large amounts of solvents and chemicals. In this sense, interest is growing in the production of membranes based on natural polymers, especially those based on cellulose.

Among the possible new biotechnological materials, cellulose stands out, and in particular, vegetable cellulose (VC), which is the main biopolymer produced by plants. Although plants are currently the most abundant source of cellulose, several types of bacteria, mainly belonging to the genera *Sarcina*, *Gluconacetobacter* and *Agrobacterium* have also been found to produce it as an alternative source [16,17]. The potential of bacterial cellulose (BC) goes far beyond its existing applications, especially with a view to its large-scale production as a low-cost raw material to provide industrial functionality in various sectors in a sustainable way [18]. BC is highly porous and has a reticular structure with small pore size, which is ideal for fine filtration purposes. However, there is still a limited number of works in the literature on its use as a raw material for filtration membranes to be applied to water treatment [10,19–22].

In this context, the objective of this review is to describe and compare different applications of BC membranes in the treatment of various effluents contaminated mainly by oils and fats. At the end of the review, the reader can realize how much BC membrane-based filtration systems, in addition to being sustainable, can reduce energy consumption and add several advantages to industrial wastewater treatment.

2. Water Resources and Energy Management

Until recently, energy management practices consisted mainly of supplanting wasteful tools. However, one study has shown that in recent decades "alternative" efforts have been made to implement better industrial practices, taking into consideration other important factors such as advanced procedures, increased productivity and better environmental conduct, among others. This has led to better results for companies compared with energy management practices alone [23].

Water and energy are both recognized as indispensable inputs to the economy, and three factors, namely security of supply, sustainability, and economic efficiency, have been considered as crucial for the industry. Some examples of the trade-offs between energy and water security include desalination plants, groundwater pumping for water supply, first generation biofuels, irrigation techniques and effluent disposal [24].

Since energy is one of the main growth factors of a given population, influencing the economic, environmental and social spheres, global energy resources have generated geopolitical tensions since the industrial revolution. However, even with technological advances, global awareness and the start of an energy transition, the main energy resources still come from oil [25,26].

Water and energy consumptions can be closely linked (Figure 1) as they are both essential for industrial production. Water is essential for the production and refinement of various types of motor fuels and for the extraction of coal and oil, but it is also widely used in the cooling process in different sectors and in the generation of hydroelectric power generation, one of the most popular forms of electricity supply in the world [27,28]. Energy is essential for drinking water production, wastewater treatment, water transport and distribution to both industry and population. Therefore, the conscious use of water and energy is a global concern, which has led to the creation of new technologies, making wastewater treatment and use of clean energy essential areas for sustainable development [29,30].

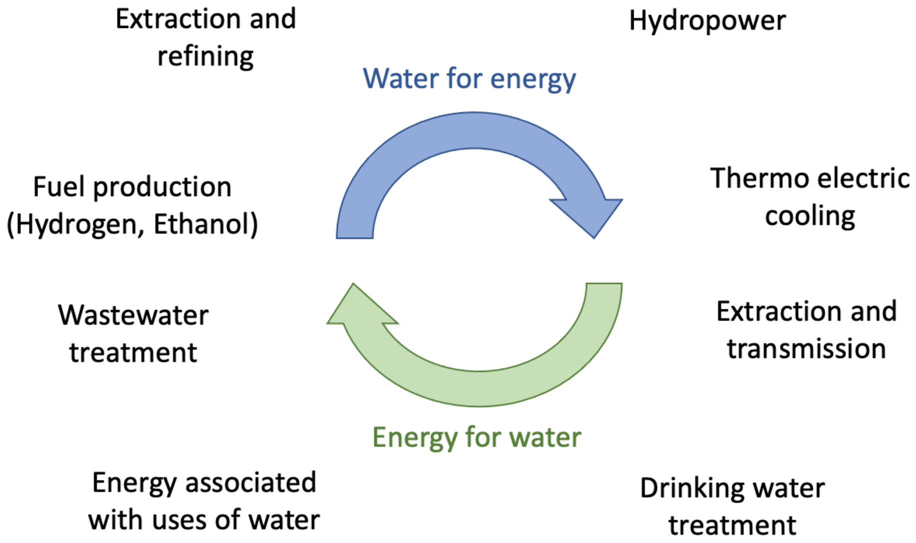

Figure 1. Water and energy consumption cycle.

Throughout history, civilizations have been able to develop various types of water supply systems, water purification devices and wastewater treatment methods. Efforts to obtain abundant quality drinking water have been intensively encouraged over the past two hundred years, leading to an increase in human life expectancy around the world [31].

This context directs research to develop new ways to achieve high-performance wastewater treatment combined with the reduction of energy expenditure, so that the effluents (a) can be properly disposed of in the environment, not causing future damage to fauna and flora, (b) can be reused in industry for example in cooling processes and (c) do not lead to a significant increase in energy purchase costs [32].

3. Water Contamination

The term "pollution" has primarily been linked to the idea of environmental contamination; however, it can also characterize something that poses a threat to all of nature and life on Earth [33]. A better definition would be that pollution is the introduction of contaminants that modify and damage a given ecosystem [34]. Among the main problems generated by pollution, we can mention the destruction of the ozone layer and the consequent greenhouse effect, acid rain, depletion of water resources, eradication of species, genetic mutations, changes in population birth and mortality rates and spread of infectious diseases [35,36].

Over the years, mankind has produced a large amount of waste, resulting in a deterioration of life quality [37]. Figure 2 shows how this damage to the environment has been caused by several factors including sediment, biological and chemical pollution [38].

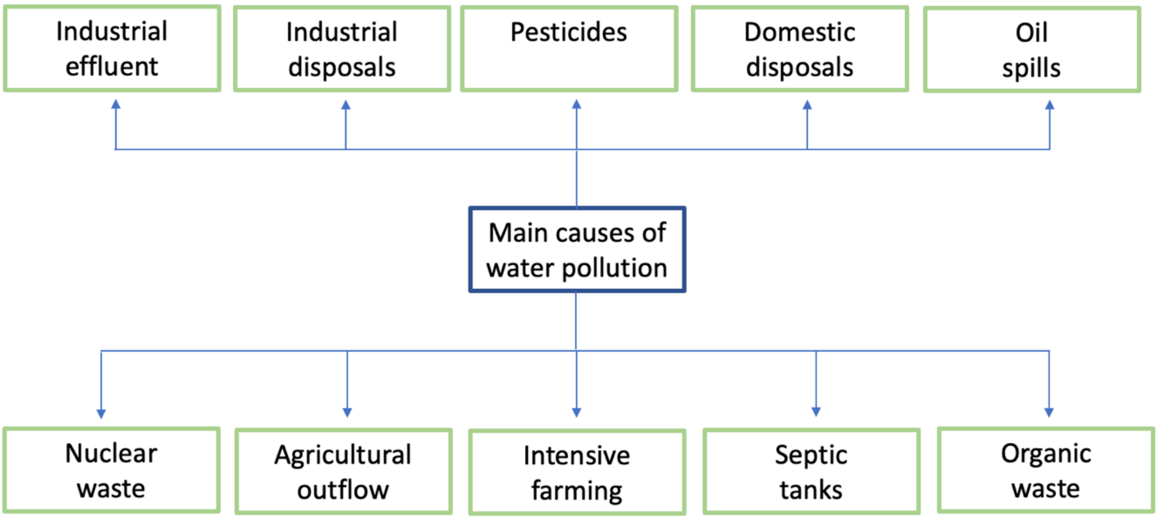

Figure 2. Main causes of water effluent pollution.

Sediment pollution occurs due to the accumulation of chemicals adsorbed in the soil, which concentrate pollutants and affect photosynthesis. It can also be caused by erosion processes, deforestation and mineral extraction, being responsible for several environmental disasters such as the tragedy that occurred in the cities of Mariana and Brumadinho in Brazil in 2015 and 2019 [39,40].

Biological pollution, on the other hand, is due to the release of organic waste from industrial or domestic sewage into watercourses, or even to its infiltration into the soil, with consequent pollution of the groundwater. The nature of this waste is purely organic, being a mixture of lipids, proteins and carbohydrates such as human feces, food leftovers and industrial waste [41]. When such waste begins to decompose, it consumes the oxygen available in water, causing the death of various aquatic organisms, generates an increase in the proliferation of algae (eutrophication) due to the high concentrations of nutrients and might enhance pathogen growth due to the use of contaminated drinking and recreational water [38].

Finally, chemical contamination is mainly produced by the discharge of chemicals into wastewater. Agroindustry is largely responsible for the chemical contamination of underground aquifers, due to the release of agrochemicals from wastewater [15]. Some of the main chemical pollutants discharged into watercourses are heavy metals (present in the residues from various industrial sectors such as textiles), domestic and industrial sewage, pesticides and agricultural fertilizers, plastics and synthetic organic compounds (oil and petroleum derivatives) [38].

Oil, among these pollutants, is one of those that most pollute the environment and are most harmful to nature. Moreover, the residues of its derivatives contain significant quantities of mineral oil, which is highly resistant to biochemical decomposition. These pollutants can be found in their free form or as emulsions made up of complex mixtures of water, oil and additives including emulsifiers, corrosion inhibitors and anti-foaming agents. While free or suspended oils can be easily separated from the aqueous phase of these wastes by simple physical processes, emulsions are chemically stabilized and can only be separated by more complex, and therefore more expensive, separation methods [9,42,43].

Chemical pollution has cumulative effects that cause enormous damage to terrestrial and aquatic life, as well as to the ecosystem and food chain [44]. The treatment and remediation of chemically polluted sites constitute one of the major barriers to be overcome, as in addition to taking time, they have a high cost [15].

4. Filtration Membranes

Over the years, the methodologies used for the treatment of industrial wastewater have been optimized in different ways depending on the circumstances and their variables such as volume, flow rate, specific environmental legislation and, above all, the composition of the effluent to be treated [12]. These treatments have as their main objective the reduction of the concentrations of contaminants present in industrial effluents, so that their discharge into water bodies can take place in a proper way [10].

Filtration is a technique that aims to mechanically selectively separate solid particles or large molecular structures from a liquid suspension with the help of a membrane with specific porosity that acts as a porous bed to perform phase separation [45,46]. They can be produced in a wide variety of configurations and structures depending on the volume and quality of water to be filtered and the separation flow [46].

In industrial filtration processes, filter membranes made up of natural or synthetic fabrics are generally used [47] to retain solid particles suspended in the air, such as microorganisms, specific gases, minerals or other volatile substances, or to treat the effluents from the textile and petrochemical industries [48]. Specific fabrics are also used in hoods, exhausts and outlets of industrial chimneys, thanks to their ability to withstand high temperatures [47,48].

The economic and practical importance of the choice of synthetics membranes is that they can be engineered to result in membranes with specific characteristics that may optimize the industrial filtration process, in addition to reducing costs for the company, such as those related to filter purchase, maintenance and energy consumption [47].

The efficiency of a filter membrane is directly related to the pore diameter and the specific parameters of the material used [49], since it establishes the exact size of the particle that can pass through the membrane. This defines their specific classification and application, as shown in Table 1.

Table 1. Classification of membranes and their respective applications.

Classification	Application	Pore Size (nm)	Reference
Microfiltration (MF)	Removal of suspended solids, protozoa, and bacteria	100–5000	[50]
Ultrafiltration (UF)	Removal of viruses and colloids	2–100	[51]
Nanofiltration (NF)	Removal of water hardness, heavy metals, and dissolved organic matter	0.5–2	[51]
Reverse osmosis	Desalination, water reuse and ultra-pure water production	0.2–1	[52]

The advantages of separation processes using membranes include (a) low energy demand for operation, (b) easy handling and maintenance, (c) no need for chemicals and (d) higher efficiency than other techniques, which depends solely on the membrane itself [49]. However, the membrane treatment also has its drawbacks. According to Hassan et al. [53], over than 95% of membranes are made up of synthetic polymers of fossil origin, and their production requires the use of solvents and aggressive chemicals. For this reason, this technology cannot be considered totally environmentally friendly, as it the production of the membrane itself has an impact on the environment. Additionally, after use, the membranes are particularly often not reusable and require specific disposal, as they are not easily degradable by the action of chemical agents [10].

For these reasons, over the years there has been a growing interest in the development of new sustainable and biodegradable products made of natural polymers, and in particular nanocellulose such as bacterial cellulose [18,54–56].

5. Bacterial Cellulose Membranes

Cellulose, composed of glucose monomers, is the main structural biopolymer of plants, but it can be also produced by other life forms such as bacteria, fungi and even protozoa. Such a natural bioproduct, whose great technological importance is justified by its wide variety of applications [57], is the most abundant in the world, with an estimated annual production of 10^{11} tons, most of which is of vegetable origin [58].

According to Donini et al. [57], bacterial cellulose (BC) differs from vegetable cellulose (VC) in that it has nanometric rather than micrometric size, better mechanical properties such as higher tensile strength and flexibility, higher purity given that VC is naturally linked to hemicellulose and pectin, higher crystallinity, water retention capacity, biocompatibility, biodegradability and biological adaptability [59,60]. These peculiar properties (Figure 3), attributable to the inter and intramolecular hydrogen bridges that hold the polymer chains together [46], make BC an extremely versatile biopolymer that can be used in various sectors of economic importance.

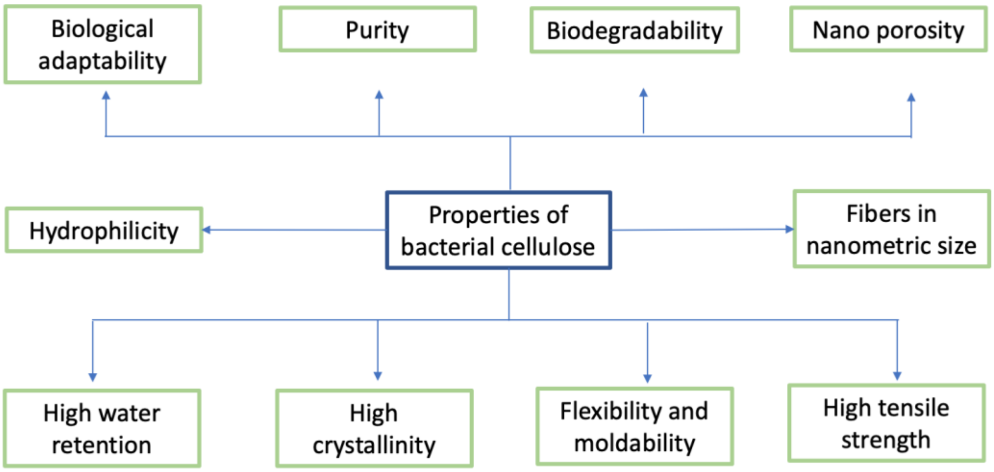

Figure 3. Main properties of bacterial cellulose.

Regarding its structure, BC consists of β-D-glucopyranose units joined by β-(1-4) glycosidic bonds, interconnected by intermolecular hydrogen bonds. Even though BC and VC have the same general formula $[(C_6H_{10}O_5)_n]$, BC's monomeric units are arranged so that one molecule is rotated 180° relative to the other [61,62].

BC is a natural biomaterial (Figure 4) that has aroused much interest in research because, in addition to having the interesting properties mentioned above, it can be subject to different types of modifications depending on the desired application, giving rise to new composites or polymer blends [63–65].

It can be produced by various microorganisms, either in consortia [10] or alone, as is the case of bacteria belonging to the genus *Gluconacetobacter* [66], which have been extensively studied for their ease of maintenance and ability to use a wide variety of carbon sources.

The BC membrane acts as a flotation promoter, which allows bacterial cells to remain in an air/liquid interface to obtain oxygen more easily for growth. It also acts as a physical barrier that protects cells from ultraviolet radiation, increases the ability to colonize substrates, and allows, thanks to its hygroscopicity, to retain moisture and prevent substrate dehydration [57].

Figure 4. Picture of a bacterial cellulose membrane. Reproduced with permission from [65], Universidade Católica de Pernambuco, 2020.

To form the membrane fiber, several adjacent fibrils join through hydrogen bonds to form layers 40 to 60 nm thick. These intertwined fibers form a gelatinous film called Zooglea (Figure 5) [67,68], which, due to being made up of about 98% water and having a lower density than water, remains on the surface of the culture medium. The thickness of the polymer membrane formed will depend on the fermentation time [18,54].

Figure 5. Picture showing bacterial growth and formation of bacterial cellulose membrane as a flotation promoter. Reproduced with permission from [69], Universidade Federal de Pernambuco, 2020.

With the aim of increasing BC productivity, several researchers have been working on this topic, testing different forms of cultivation in bioreactors and agitated cultures, in contrast to the traditional static culture, as well as different feeding conditions [57,70,71].

As for their biotechnological application as physical barriers [46,72], BC membranes, due to their hydrophilic features, can be used to filter wastewater contaminated by oily substances, in order to increase the efficiency of the entire process and to allow the treated water to have characteristics in compliance with the environmental standards for disposal or with reuse within the company itself [10].

Their characteristics make them suitable for use in the treatment of effluents from various industrial sectors, such as the medical–pharmaceutical [18], food [17], cosmetic [73], electronic [74–76] and packaging [56,77] ones, as ultrafiltration membranes [45,78] or separate apolar effluents [10,79].

6. Influence of Culture Conditions on Bacterial Cellulose Production

All the factors that normally influence fermentations are important for the final BC features. These include the cultivation time, the conditions of culture environment, including the bacterial species, nutrients, pH and dissolved oxygen (DO) level during fermentation [80], and the physical and mechanical parameters such temperature, aeration and agitation [46,81,82].

The biopolymer can be produced by submerged fermentation in liquid media, which can take place statically or under stirring using shakers or bioreactors, and BC will acquire different shapes and conformations depending on the culture conditions [83,84]. When production is carried out under static conditions, the regulation of the dissolved oxygen level and the concentration of the carbon source in the medium are crucial for its formation [85,86], and the thickness of the formed membrane is directly proportional to the cultivation time [87]. However, growth stops when the membrane reaches a size that limits the access of oxygen to the cells in the medium, making them practically inactive [88,89]. Instead, irregular, sphere-like cellulose particles are produced under stirring [90]. In this type of process, the pellets formed have low mechanical strength and a non-flat structure with low contact surface, not suitable for applications where these characteristics are necessary, as is the case of filtration systems. However, this production method aims to greatly increase BC productivity, seeking greater applicability for other industrial purposes [57].

Even in culture media rich in nitrogen and carbon sources, the cultivation period can vary from 48 h to 6 weeks, depending on the strain used [46,91]. Among the possible causes of slow cell growth, the transfer of oxygen and nutrients into the film appears to be the most influential factor [63].

According to Hirai et al. [92], the temperature during BC production not only affects the productivity, but also its structural characteristics such as morphology and crystallinity. This phenomenon was demonstrated for cellulose production by *Acetobacter xylinum* ATCC 23,769 in Hestrin-Schramm medium at two different temperatures, i.e., the typical cellulose II bands were detected in cellulose produced at 4 °C, and those of cellulose I in that produced at 28 °C. Cellulose has the tendency to form as a highly crystalline material, but there are multiple types of structure of this polymer. The most common structures are cellulose I and cellulose II, the former being the native form (found in nature as it is), while the latter being obtained by treating the native one (with high purity). Therefore, cellulose II has greater crystalline regions, meaning that it has a more stable polymer structure when compared with cellulose I [93].

Another important factor is the level of DO in the culture medium, which is essential for cell metabolism [94]. The DO levels must be carefully controlled, because too-high values increase the concentration of gluconic acid, hindering cell production capacity, while too low values limit bacterial growth, equally reducing the BC yield [80].

As for microbial producers, although different bacterial species can synthesize BC, those belonging to the genus *Komagataeibacter*, formerly known as *Gluconacetobacter*, take on a leading role because they ensure high BC yield, essential for industrial production and marketing, and are able to use different carbon and nitrogen sources [71].

Finally, as seen in Table 2, much research has been conducted with the aim of reducing costs by using agroindustrial wastes to prepare alternative low-cost culture media and, at the same time, optimizing cultivation conditions for BC production.

Table 2. Culture conditions found in the literature on bacterial cellulose production using agroindustrial wastes as low-cost media.

Culture Medium	Microorganism	Time (Days)	Temperature (°C)	pH	Dry Weight Yield (g/L)	Reference
Lipid fermentation wastewater	*Gluconacetobacter xylinus* CH001	5	28	6.0	0.66	[95]
Hydrolysate of dyed waste cotton fabrics	*Gluconacetobacter xylinus ATCC 23,770*	10	30	5.0	12.80	[96]
Corn Steep Liquor	*Gluconacetobacter hansenii* UCP1619	10	30	6.0	9.63	[54]
Cheese whey	*Komagataeibacter medellinensis* NBRC 3288	10	30	3.5	2.37	[97]
Rotten banana juice	*Komagataeibacter medellinensis* NBRC 3288	10	30	3.5	4.81	[97]
Rotten mango juice	*Komagataeibacter medellinensis* NBRC 3288	10	30	3.5	1.95	[97]
Potato peel wastes	*Gluconacetobacter xylinum* ATCC 10,245	6	35	9.0	4.70	[98]
Tomato juice	*Acetobacter pasteurianus* MTCC 25,117	7	30	4.5	7.80	[99]
Tropical fruit residues	*Gluconacetobacter hansenii UCP 1619*	10	30	6.0	7.60	[89]
Vinasse	*Komagatacibacter xylinus*	10	30	6.0	1.80	[100]
Cashew apple juice and soybean molasses	*Gluconacetobacter xylinus*	7	30	5.5	4.54	[101]
Fruit and vegetable peels (cucumber, melon, kiwifruit, tomato, apple, quince and pomegranate)	*Komagataeibacter hansenii*	21	30	4.5	1.40	[102]
Whey	*Acetobacter pasteurianus*	8	30	4.0	5.60	[103]

It is worth noting that, in addition to reducing process costs, the use of these by-products as culture media allows to increase their value through the development of a sustainable BC production, a possible improvement in yield, and minimization of environmental impact caused by their disposal [101].

7. Bacterial Cellulose in Wastewater Treatment

As BC has hydrophilic and oleophobic properties [49], during the filtration of oily effluents or emulsions only water droplets pass through the nanometric pores of the membrane at a certain pressure applied to the system, which means that the oil remains on its surface (Figure 6).

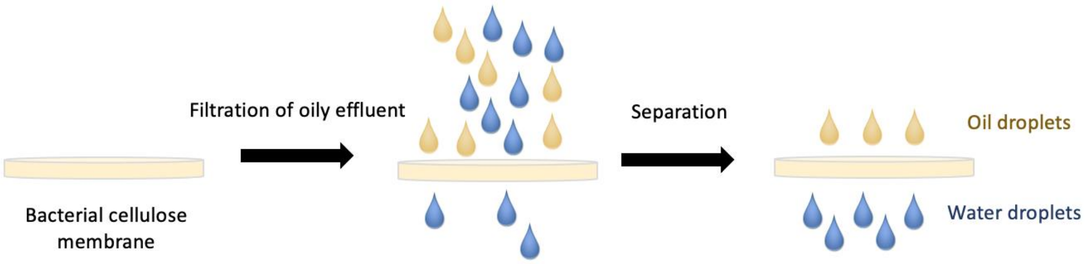

Figure 6. Schematic representation of oil–water separation process using BC as a filter.

For oil/water emulsions, some studies have shown that certain modifying agents are able to make BC membranes hydrophobic, thus enhancing their oil/water selectivity [22]. This is an attempt to obtain a better filtration yield according to the filtrate specifications.

Moreover, thanks to its nanoporous structure and its susceptibility to chemical derivatization, BC is suitable for the removal of heavy metal ions from aqueous solution [104]. In this context, grafting of cellulose-based materials with functional groups, such as amino, carboxyl and thiol groups, has been proposed as a strategy to enhance their adsorption capacity [105]. However, other reports have shown that the capacity of these altered materials to adsorb metal ions, as well as their mechanical properties, still need further improvement through new derivatization strategies [10,104]. For this purpose, studies have been focusing on improving the flexibility and mechanical strength of these materials compared with the simple BC [106], the latter being an extremely important property when they are used to produce reusable membranes. Another strategy to modify the BC structure is to increase the pore size by preparing composite materials, since its dense nanofibrillar structure can impair the performance of the separation process due to liquid infiltration that reduces its efficiency and the possibility of reuse. Furthermore, such a procedure reduces energy consumption because separation takes place using gravity alone, i.e., without the need to apply any additional pressure [79].

Importantly, all these modifications to the polymer can be done alone or in combination, depending on both the desired BC (composite or blend) properties and the specific application [63,64].

The few studies conducted so far using BC as a filter membrane (Table 3) suggest that it has enormous unexplored potential, with particular reference to its low sensitivity to water, so that it does not decompose in contact with substances in liquid state, high degree of porosity, low density and nanofiber structure that allow nanoscale filtration [18,72].

As shown in Table 3, most of the studies have focused their efforts on modifying BC with incorporation of other active materials, with the purpose to enhance its properties [22,46,104,106–112]. The results of a recent study, performed on a BC membrane capable of acting both as a contaminant adsorbent and an antimicrobial agent, have indicated a unique and innovative way to remove viable bacterial cells from water [112]. Such modifications, among which it is worth mentioning amination, addition of aerogels, silica, chitosan and phosphate functional groups [104,106–109], are studied according to the specific characteristics of the desired treatment [46,111].

Another study reported the use of BC associated with polyethyleneimine (PEI-BC) as an adsorbent material for heavy metals, whose maximum adsorption capacity for Cu(II) and Pb(II) was found to be 141 and 148 mg/g, respectively. The polymeric blend PEI-BC also showed good reusability after post-treatment regeneration using Na_2EDTA. After treatment, the Cu(II) readsorption capacity of PEI-BC was lower than the initial one, but kept stable after each cycle of use. Overall, PEI-BC showed good reusability in removing Cu(II) and Pb(II) from aqueous solutions, showing its potential as a bioadsorbent for the removal of heavy metal ions from wastewater [104].

Table 3. Studies relating to the use of bacterial cellulose (BC) as filter membrane to treat effluents.

Title	Description	Reference
Surface modification of bacterial cellulose aerogels' web-like skeleton for oil/water separation	Nanofibers of BC aerogels were modified on their surfaces by trimethylsilylation derivatization followed by freeze-drying. The resulting hydrophobic and oleophilic aerogels were shown to remove a wide range of organic solvents and oils, with potential use in cleaning up oil spills in the marine environment.	[22]
Polyethyleneimine-bacterial cellulose bioadsorbent for effective removal of copper and lead ions from aqueous solution	Reductive amination with polyethyleneimine allowed to transform the BC membrane into a bioadsorbent for the removal of heavy metal ions [Cu (II) and Pb (II)] from wastewater.	[104]
Facile fabrication of flexible bacterial cellulose/silica composite aerogel for oil/water separation	A silica aerogel composite was prepared by BC modification with methylene diphenyl diisocyanate to increase its hydrophobicity and flexibility, thus making it a promising oil sorbent.	[106]
Preparation and characterization of a bi-layered nanofiltration membrane from a chitosan hydrogel and bacterial cellulose nanofiber for dye removal	A membrane was developed by grafting multi-walled carbon nanotubes into BC molecular chains. The BC powder was dissolved in a solution of LiCl and *N*,*N*-dimethylacetamide, and stannous octoate was used as a reaction catalyst. The membrane exhibited greater tensile strength, Young's modulus and pressure resistance, which practically tripled its flow rate and allowed for a yield of dye removal above 90%.	[107]
Design of reusable novel membranes based on bacterial cellulose and chitosan for the filtration of copper in wastewaters	Chitosan-modified BC membranes were developed by ex situ (BC immersed in solutions with different chitosan concentrations) or in situ (addition of chitosan solutions to BC production medium) techniques for Cu (II) ions adsorption. The membrane produced by the ex situ technique showed greater efficiency in removing ions.	[108]
Removal of U(VI) from aqueous solution using phosphate functionalized bacterial cellulose as efficient adsorbent	BC membranes were modified by grafting phosphate functional groups soaking them in dimethylacetamide and urea. Membrane characterization confirmed the successful incorporation of phosphate groups. Due to the presence of polar hydroxyl groups and electrostatic attraction, the membranes at pH between 4 and 8 were able to adsorb 9 mg/g of U (IV) ions.	[109]
Bacterial cellulose membranes for environmental water remediation and industrial wastewater treatment	BC was produced and cleaned with NaOH to be used as a filter membrane for the treatment of microbiologically contaminated effluents (*Escherichia coli*) and dyes from the textile industry. BC membranes showed better results than the commercial ones, removing 100% of cells present in the effluent and being able to be reused for 10 cycles without loss of efficiency.	[110]
Impact of incubation conditions and post-treatment on the properties of bacterial cellulose membranes for pressure-driven filtration	Studies on the permeation properties of BC derivatized with poly-oxyethylene were carried out to determine the filtration efficiency of both dry and wet membranes at different pressures and water flow rates.	[46]
Film-like bacterial cellulose/cyclodextrin oligomer composites with controllable structure for the removal of various persistent organic pollutants from water	A film-like water purifier, prepared by loading cyclodextrin oligomer onto ultrafine BC, was described. The system showed high and stable adsorption capacity toward various target pollutants such as phenol, bisphenol A, glyphosate and 2,4-dichlorophenol.	[111]
Bacterial cellulose-polyaniline porous mat for removal of methyl orange and bacterial pathogens from potable water	BC membranes were modified with polyaniline by in situ oxidative polymerization and posterior lyophilization. BC was applied to remove methyl orange dye and bacterial cells present in drinking water. Membranes showed an absorption capacity of approximately 300 mg/g and antimicrobial activity, reducing the microbial load present in the effluent by up to four times.	[112]

The use of microbial cellulose membranes in association with chitosan for copper removal has also been reported. Membranes prepared using concentrations of chitosan and cellulose of 50 and 250 mg/L, respectively, were able to remove 50% of Cu. As for membrane reusability, a decrease in the removal efficiency of less than 10% was observed after two treatment cycles. These results suggest the possibility of using BC-based polymer blend membranes to remove copper from wastewater [108].

A nanofiber polymer blend membrane with chitosan hydrogel and BC was also reported for dye removal from wastewaters [107]. The rejection rate was higher than

90% for dyes with molecular weight above 600 g/mol and pressure below 0.5 MPa, and the membrane showed good antifouling properties for both oil and proteins during the filtration process. These highly promising results indicate that nanofiltration membranes are effective in removing dyes from wastewater, with high rejection rate and flux at high pressure [107].

Another interesting application is the use of BC filters for the treatment of pigmented textile effluents and microbial cell removal. BC membranes were effective in the removal of *Escherichia coli*, dye effluents and solids for up to ten cycles, which suggests that they could be used in various types of wastewater treatments, in addition to playing a role in the development of new biotechnological tools such as enhanced filtration methods with more cost-effective materials [110].

Moreover, one of the main advantages of BC membranes, compared with the traditional ones made of VC, is the possibility of washing them after filtration. Taking this into account, saturated BC membranes, whose filtration capacity is reduced, can be removed from the filtration system, washed and reused a few times, without losing their efficiency [10].

There are no patents yet on the use of BC membranes to filter oily effluents. However, there are some scientific publications on this subject that are worth mentioning, as shown in Table 4.

Table 4. Articles related to the use of bacterial cellulose (BC) membranes as porous bed for separation of water–oil mixtures.

Title	Description	Reference
Use of bacterial cellulose and crosslinked cellulose nanofibers membranes for removal of oil from oil-in-water emulsions	Wet BC and crosslinked cellulose nanofibers were used for the removal of oil from stabilized and non-stabilized oil-in-water emulsions with droplet size of less than 1 µm. The efficiency of oil removal from stabilized and non-stabilized emulsions was higher than 92%.	[53]
Functional bacterial cellulose membranes with 3D porous architectures: Conventional drying, tunable wettability and water/oil separation	A BC membrane was functionalized by the hydrolysis of alkoxysilanes. This procedure was able to preserve the 3D nanofibrillar architecture of the membrane even after the drying process and increased its surface wettability and ability to separate oily emulsions.	[113]
Use of a bacterial cellulose filter for the removal of oil from wastewater	BC membranes produced in an alternative medium based on corn steep liquor were cleaned with NaOH without further treatment. When used as filters, they made it possible to retain almost 100% of the oil present in the emulsion.	[10]
Facile and green route to fabricate bacterial cellulose membrane with superwettability for oil–water separation	A simple method was described to weave BC fibers and BC nanofiber clusters in aqueous dispersion on a stainless-steel mesh, which resulted in an increase in the roughness and consequently in the wettability of the biopolymer. The oil–water separation process showed 99% efficiency.	[114]
Sustainable, superhydrophobic membranes based on bacterial cellulose for gravity-driven oil/water separation	Needle-leaf bleached kraft pulp was added to BC to increase the biopolymer pore size, thus forming a superhydrophobic/super-oleophilic membrane. The membrane showed not only an oil–water separation yield by gravity >95%, but also an excellent recyclability, as it was washed and reused without significant structural changes after 10 separations.	[79]

The study developed by Galdino et al. [10] has shown that the separation of oil molecules present in oily emulsions can take place with high efficiency even without membrane modification. However, further studies are needed to improve factors such as time of filtration and maximum pressure.

While several studies compared the yield of removal achieved during filtration [107,110,113,114], none of them developed an effective method for maintaining the retention of contaminants in a qualitative manner (oily or not). This aspect is important to develop a production or modification method that aims to increase the flow rate and consequently the filtration speed [10].

Although BC form quickly and its regeneration is not season-dependent, production on an industrial scale is still relatively expensive [79]; therefore, BC association to form composites [63] or polymer blends [56] is being studied to make it more feasible.

As shown in Table 5, only three patents have been filed on the use of BC as a filter material [45,78,115]. These inventions propose the use in ultrafiltration and dialysis of BC membranes treated with silica or gelatin solutions to fill the pore voids.

Table 5. Patents filed on the use of bacterial cellulose membranes (BC) as porous bed for the separation of water–oil mixtures.

Patent	Description	Reference
CN103301815B	A method for preparing a BC filter to purify water was described. The membrane nanofibrils incorporated a silica solution to assist in the refinement of the filtration process. BC membranes also acted as a material for drainage and removal of bacterial cells present in water.	[78]
BR 1020180097369A2	BC membranes functionalized with silica solution, using sodium tetraborate to bind the silica to cellulose nanofibrils, were used in chromatographic analyses. BC did not clog (saturated) even after 10 filtrations, and no biological matter was detected in the filtered material.	[45]
CN110354693A	BC membrane was modified by incorporating gelatin microspheres to improve the filtration of reactive and acid dyes used in textile processes.	[115]

The study by Galdino et al. [10] showed that BC membranes can remove almost 100% of the oil present in synthetic effluents and can be washed and reused for the same filtration process more than 20 times without losing their structural characteristics and filtration capacity. Recent studies have also shown that the filtering properties of BC membranes are directly linked to the structural network of their fibrils, which can be modified by variations in culture conditions and ex situ treatments. The permeation properties of BC vary in fact according to the flow rate and pressure used in the filtering system, as well as the size of the particles to be retained, which determine the time that the membrane will take to completely saturate [46].

The great potential of BC membranes as a filter material for oily effluent separation has already been demonstrated in some scientific works, as described earlier, and the key to optimizing their use in the industrial field lies in research relating to the control of their saturation and to the increase in pressure and flow rate they are able to withstand.

8. Conclusions and Perspectives

Bacterial cellulose (BC) is considered an eco-friendly and extremely versatile biopolymer, and this is why studies have increased over the years that envisage its use in the form of filter membranes for wastewater treatment. The present review showed the great potential of BC membranes, produced in standard or alternative culture media by single bacterial species or microbial consortia. Thanks to their peculiar characteristics, mainly their nanofibrillar structure, they have proven effective as filters to retain small particles and have been successful in the treatment of industrial effluents, in particular those of the petroleum industry. Characteristics of BC such as high water retention and tensile strength make it an excellent sustainable, biocompatible and biodegradable porous filter bed.

Until now, no reports are available on a direct relationship between BC membrane filtration and related energy expenditure. Further studies are needed to make these membranes resistant to higher flow rates and pressures, so that they can replace today's methods on an industrial and ecofriendly scale. In fact, it is expected that this technology will allow a reduction in energy and maintenance costs, a possible improvement in the treatment and a lower emission of pollutants.

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