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Environmental, Medical and Textile Applications of Bacterial Nanocellulose: A Patent Review

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Abstract: Microbial cellulose, commonly known as nanocellulose or bacterial cellulose (BC), especially during the last decade, has received increasing attention because of its extraordinary versatility; it can be utilized in various applications. The growing interest is observed in the number of published scientific articles and the development of new production methods resulting in diverse patents. In this current review article, research was carried out regarding BC patents given the economic interest and the advances in specific areas: environmental processes, especially as a filtering membrane, in the medical industry, as an intelligent dressing, and in the textile industry, as a new biotextile. This review aimed to compile biotechnological studies and their applications that address nanocellulose as a promising material in registered patents. A current trend in biotechnology is the development of practical applications in the biopolymer BC area, further contributing to the future expansion of the nanocellulose market and knowledge.

Keywords: Biocellulose, filter, patch, fashion, patents.

1. Introduction

Integrating natural polymers with other materials can be highly functional in various industries, such as textiles, biomedicine, and others. Composites and polymer blends are newly emerging smart hybrid materials, and cellulose is a highly used biopolymer for such combinations [1].

Cellulose is a glucose monomer-based polymer with a major structural mass in vegetables. It is found in several forms of life, such as vascular plants, protozoa, and prokaryotes, in addition to the possibility of being produced by certain fungi and bacteria species. It is considered a bioproduct of great technological importance, as this natural polymer can be used in various applications [2]. With an estimated production of 10^{11} tonnes per year, this biomaterial is considered the most abundant on the planet, and the majority is from vegetable sources [3]. However, the extensive usage of wood has resulted in environmental issues, with the addition of having an expensive manufacturer [4].

Bacterial cellulose (BC) consists of a biopolymer similar to vegetable cellulose (VC), with one of the differences being that several microorganisms produce BC. Either in the build of composites [5] or by themselves, as in the case of bacteria such as *Sarcina* or *Komagataeibacter* [6], this last one being widely studied for its ease of maintenance and its ability to process a wide variety of carbon feeding sources. Brown [7] was the first to report BC, where it identified the growth of the unbranched film with a structure chemically equivalent to vegetable cellulose [8].

Concerning its structure, BC is made up of β-D-glucopyranose units joined by β- (1-4) glycosidic bonds, connected by intermolecular H-bonds [9]. BC has the same molecular formula ($C_6H_{10}O_5$) as VC, but with monomer units being organized so that one molecule is rotated perpendicularly around the other [10,11].

Recent studies point BC as a promising biopolymer due to its wide field of applications, as an alternative to traditional VC production. According to Donini *et al*. [2], BC differs from VC mainly due to the presence of nanometric fibers compared to the micrometric fibers found in the vegetable source. This gives it excellent mechanical properties such as greater purity, crystallinity, water absorption power, enhanced tensile strength, a higher rate of polymerization, and, in addition, better biological adaptability [8].

Due to the 3D nanofibrillar network, BC has a high water absorption capacity (after production, its membrane has ~ 99% water content) and high tensile strength. Such unique properties, in combination with other interesting characteristics, as biocompatibility, biodegradability, and renewable character, result in the renewable character and diverse range of uses, including medicine [12], food [4], surgical masks and protection [13,14], cosmetics [15], electronics [16,17], packaging production plastics [18,19], ultrafiltration filter [20,21] and for separating oily mixtures [5, 22, 23] increasing its wide interest in academic works [24].

Several studies for scientific articles and patents are being carried out on this biopolymer each year; however, the focus chosen for this review, involving the environmental, textile, and medical areas, was motivated by the difficulty of information gathering regarding the BC markets due to the still relatively low number of studies related to these promising applications of this versatile biopolymer.

Therefore, the priority in this research is related to the areas of fundamental importance to prove, effectively, the interest in advancing research in this theme and the applications highlighted in these areas worldwide.

2. Material and Methods

The patent review on the applicability of bacterial cellulose related to the specific research areas selected (BC as a filter membrane, an intelligent dressing, and biotextile) was carried out between July 2021 and February 2023 in some of the main worldwide research platforms and patent deposits, including the European Patent Office (EPO), World Intellectual Property Organization (WIPO), Lens and the National Institute of Industrial Property in Brazil (INPI), considering patents filed in any year.

The keywords used for the patent search were bacterial cellulose filter/strainer, strainer with bacterial cellulose membrane, bacterial cellulose/biocellulose skin patch; biocel, bacterial cellulose & fashion, bacterial cellulose & dyeing and, in addition, a filter was used so that the words would be found in the title and/or summary of patents. For the Brazilian properties platform search, the keywords used were the same, but in Portuguese. Any duplicate patents unrelated to searches have been discarded.

3. Results and Discussion

The search was carried out without taking into consideration the patents' date of publication. Based on this, 2,146 patents were found, of which 2,122 were excluded because they were duplicated or not related to BC and its applications. Thus, 24 patents were selected for critical analysis to carry out a technological exploration of BC and its applications. Most patents found were under secondary filing numbers, causing an intense repetition of patents in each database searched. Despite this, patents for products and processes were found that involved the keywords searched for.

Figure 1 shows the searches related to the projects in each research platform, the total number of patents found during the review, and the amount related to the issues discussed throughout the paper.

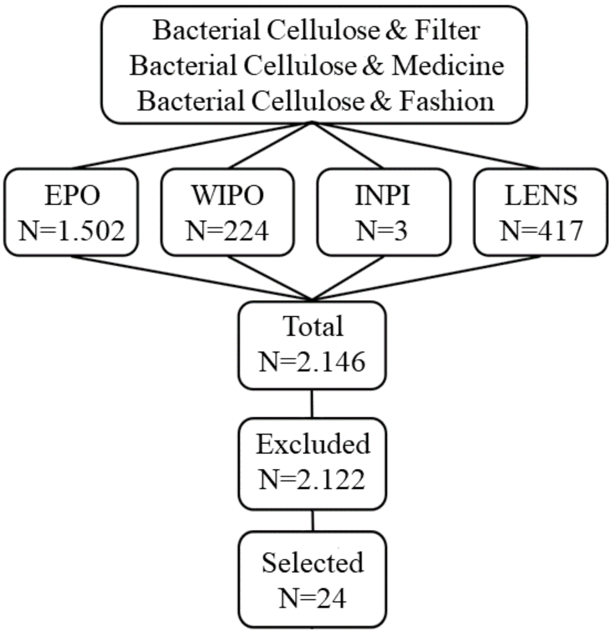

Figure 1. Patent survey flowchart. European Patent Office-EPO; Instituto Nacional da Propriedade Intelectual-INPI (National Institute of Intellectual Property-Brazil); World Intellectual Property Organization-WIPO.

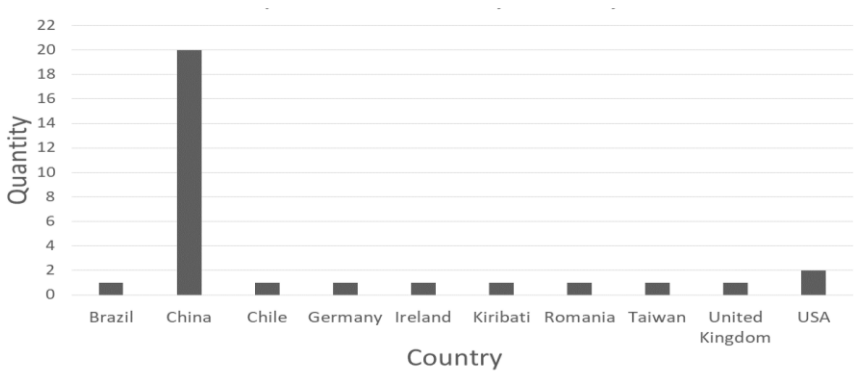

Figure 2. Amount of bacterial cellulose (BC) patents related to selected specific research areas (environmental, medicine, and textile industry) by country.

The selected deposited patents were from China (20), Brazil (1), Chile (1), Germany (1), Ireland (1), Kiribati (1), Romania (1), Taiwan (1), United Kingdom of Great Britain and Northern Ireland (1) and the United States of America (2), as stated in Figure 2. This demonstrates the ability of researchers in these countries to protect their inventions and reflects the backwardness of countries like Taiwan and Brazil with only one patent.

It is important to observe that patents related to the keywords have their maximum number of publications in 2022, with seven patents. Previously, most of the patents (four) were published in 2019. However, it was noticed that the number of related patents decreased in 2020, and no results related to the researched areas were shown, probably due to the Covid-19 pandemic. In 2021, the number of patents filed increased again, but probably still as a result of Covid-19, it did not have such high numbers (three). The research points to a growing interest in the search for biocellulose related to the researched areas, indicated by the fact that in the first two months of 2023 alone, three patents related to the areas of interest of this work have already been deposited, as illustrated in Figure 3.

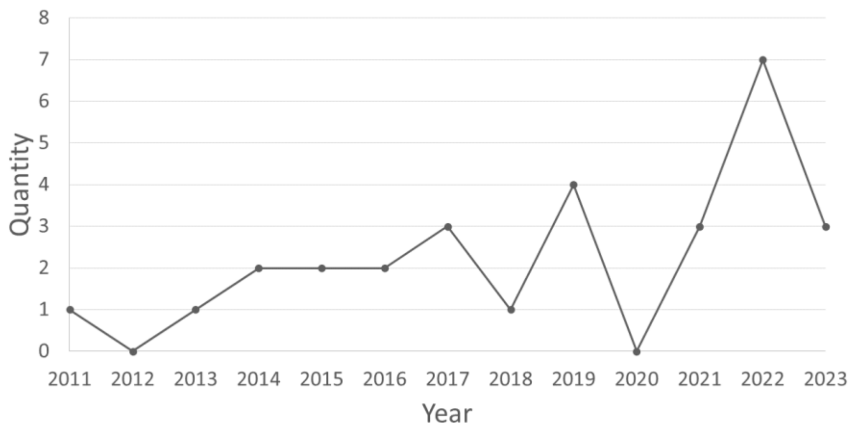

Figure 3. Diagram showing the amount of bacterial cellulose (BC) patents related to selected specific research areas (environmental, medicine, and textile industry) by year.

According to the International Patent Classification (IPC), these filed patents are classified into several classes, as demonstrated in Figure 4. Table 1 describes the name for each of their classification, most of which were found to be classified in the areas A61K, A61L15, A61L27, B01J, C02F, D06M and D06P, which deal with "Products for medical, dental, or personal hygiene purposes", "Chemical aspects of, or use of materials for, wound dressings or bandages in liquid, gel or powder form", "Bacterial cellulose biological adhesive and manufacturing method", "separation processes using semi-permeable membranes", "water treatment", "Treatment of fibers, threads, threads, fabrics, feathers, or fibrous products" and "dyeing for textiles or leather" respectively. This data demonstrates BC's wide range in applicability.

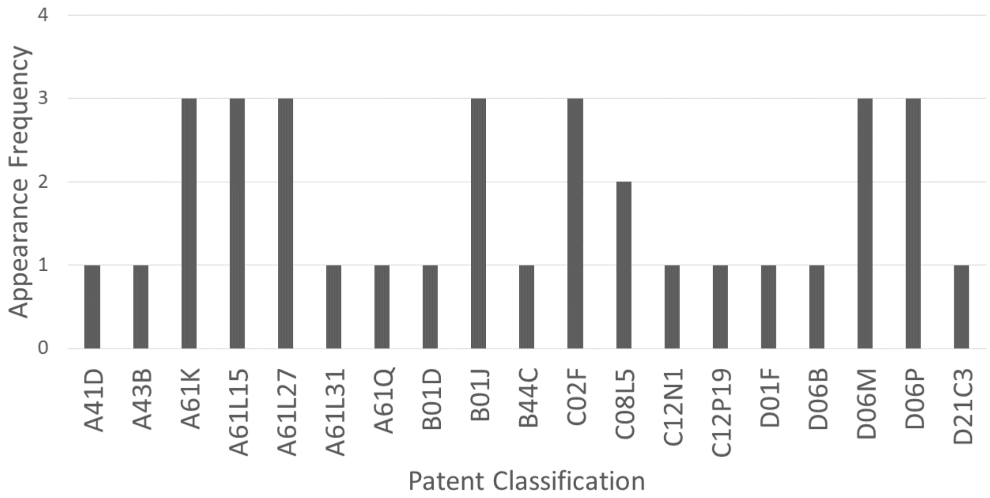

Figure 4. Frequency of bacterial cellulose (BC) patents related to selected specific research areas (environmental, medicine, and textile industry) filed by classification (International Patent Classification - IPC).

Table 1. International Patent Classification (IPC's) names.

IPC's codes	Classification names
A41D	Protective clothing
A43B	Parts of footwear
A61K	Products for medical, dental, or personal hygiene purposes
A61L15	Chemical aspects of, or use of materials for, wound dressings or bandages in liquid, gel or powder form
A61L27	Bacterial cellulose biological adhesive and manufacturing method
A61L31	Integrated bacterial cellulose repair adhesive and preparation method
A61Q	Specific use of cosmetics or personal care products
B01D	Separation processes using semi-permeable membranes
B01J	Chemical or physical processes
B44C	Production of decorative effects
C02F	Water treatment
C08L5	Film composed of medical pectin / bacterial cellulose and preparation method
C12N1	Preparations containing material from protozoa, bacteria, or viruses
C12P19	Preparation of compounds containing saccharide radicals
D01F	Chemical features in the manufacture of man-made filaments, threads, fibers, bristles or ribbons
D06B	Treating textile materials using liquids, gases or vapours
D06M	Treatment of fibers, threads, threads, fabrics, feathers, or fibrous products
D06P	Dyeing for textiles or leather
D21C3	Pulping cellulose-containing materials

After the search results, 24 patents were selected because of their relevance to the selected research areas. Seven patents for BC as a filter membrane for water treatment, six for the medical area, and eleven for the membrane applied in the textile industry. Each application and patent is presented and discussed separately, as described below.

3.1. Bacterial cellulose filter.

The great exploitation of oil and its derivatives generates large amounts of effluents contaminated by oils and greases. As a result, this effluent treatment has become an urgent and indispensable need in various industrial sectors, including the oil industry and all existing environmental and health agencies [23, 25, 26]. However, one of the greatest challenges for environmental agencies is the disposal within acceptable environmental parameters.

Among the possibilities of supplying new biotechnological materials, bacterial cellulose's obtention has potential beyond its existing applications. Production on a large-scale is possible using low-cost raw materials and industrial byproducts [12,15,27].

The biopolymer can be used to separate and treat oily and petroleum-derived effluents (annually, the most polluting industry). A great BC advantage as a filter is its reusability after its application by the process of washing, thus making the useful life of the membrane similar to conventional filters already on the market. In addition, as BC is biodegradable, when used for oily treatments, it can be discarded in the environment after a simple cleaning process, therefore causing no damage to nature [23].

It is necessary to discuss aspects related to the properties of the BC membrane as a filter for separating oil mixtures [5]. Therefore, a search for registration of patents linked to the membrane as a filter was carried out to validate the possible configurations already used with BC in the world market.

Patent CN103301815B demonstrates a method of preparing a BC water purification filter. The cellulosic membrane is used as a biomaterial for ultrafiltration using a silica suspension incorporated into its nanofibrils to refine the filtration process. The BC used in the process works as a separation membrane material for drainage and can even retain microorganisms in the water. The process is referred to as a simple, easy, and inexpensive water purification method [20].

The invention CN204217973U demonstrates the usage of the dry BC membrane as an air filter material in protective masks. The entire mask is produced in this configuration using only BC as the matrix material. The nanometric structure of the biopolymer allows the mask to work as a separating agent, intercepting harmful levels of pathogens, such as smog present in the air, and providing a comfortable use, reusable and biodegradable [13].

Unlike the previously referenced patent, in the CN107486033A patent, BC is used not for the mask as a whole but as a nanofibrillar film for air filtration in masks. The authors demonstrate the assembly of the final mask product in addition to the BC's filter obtention itself. After the manufacture, the final product's surface is modified by making it hydrophobic (in contrast to BC's naturally hydrophilic property) to filter solid particulate matter in the air more effectively [14].

The invention BR 1020180097369A2 describes the obtained BC's composition and its application as a filter material for hemodialysis, chromatography, water purification, and effluents. This patent involves modifying the polymer membranes with silica solution and binding the silica with sodium tetraborate ($Na_2H_{20}B_4O_{17}$). This procedure is conducted so that the membrane can be used in chromatographic analysis. In addition, the BC did not show saturation (clogging) by the tested biomass after 10 filtrations, and it did not even present any type of biological matter in the filtered material [21].

Patent WO2023004521A1 [28] demonstrates a nanocomposite filtering material of hydroxyapatite ($Ca_{10}(PO_4)_6(OH)_2$) and BC produced by an *in situ* synthesis. Their intention is to develop a material that can remove heavy metals from liquid effluents, particularly potable or household water. The use of the new biomaterial in absorption processes was able to remove the total filtering of heavy metals in the water to be treated. The methodology is referred to as having a low cost and high biosorption efficiency that reduces the use of chemical products and the generation of biological sludge.

The invention CN107164425A [29] demonstrates a flocculation treatment method for dyeing wastewater by using a composite based on bacterial cellulose and activated carbon. The method involves the production of BC in an agitated manner in order to have its fibers more defibrillated. This method is described as simple to prepare and perform, giving rise to a new green and biodegradable material applicable to wastewater treatment from dyeing industries and other similar effluents.

Lastly, the CN110354693A invention demonstrates a method of preparing a BC membrane for filtration applications. This invention's distinction is modifying the membrane by combining gelatin microspheres to aid in the filtration process of acid or reactive dyes [30].

Bacterial cellulose has been researched with satisfactory results over the years as a filtering membrane for different types of separation from different physical states of matter. However, the treatment of effluents contaminated by oily emulsions still lacks research on the treatment of large quantities of this material since these effluents are mass-produced daily by practically all industrial sectors [22, 25, 31, 32].

3.2. Bacterial cellulose & medicine/patch.

The development of science and technology contributes to the constant improvement and/or development of health products. Advanced medical biomaterials are of great interest due to their expansion in fields such as wound healing, controlled release of medications, bandages, pressure clothing, and implantable medical devices, among others [6].

Among the materials widely used in this segment, natural polymers stand out, such as BC, which has a great diversity of medical applications due to its biocompatibility with dermal tissues, high purity, ultrafine network structure, and high porosity (with a consequently a large surface area) [24]

In both academic and industry research, BC has sparked much interest as a biodegradable biopolymer [33]. Its hydrophilic nanofibers result in a high water retention capacity and adherence to assets and drugs. As a result, research on the registration of membrane-related patents being explored as a biomaterial in the medical industry was carried out to validate the possible configurations already used by BC on the world market.

The widespread use of BC in medical sciences to augment or substitute the normal function of damaged tissue or organs has resulted in many biomedical materials, some of which have already been commercialized. According to the application, this biomaterial could be temporary or permanent in the patient's body [33]. The research focus was on BC used as a transdermal dressing to demonstrate the growing potential of applying this biopolymer in this area.

BC has undergone extensive research and development as a biomaterial for diverse biomedical applications. However, as it lacks antibacterial, antioxidant, and conducting properties, studies have been modifying such characteristics by using biosynthetic (in-situ) and ex-situ methods to improve its properties for medical applications [12].

The CN103861146A invention demonstrates the method of manufacturing a biotechnological bacterial cellulose adhesive, with its improved characteristics for such application, and, as a result, excellent mechanical properties were obtained, in addition to better biological fitness, better anti-adherence, and antimicrobial properties in soft and wet tissue environments [34].

Patent CN104403136 provides a method of preparing a polymeric composite film formed by pectin and BC. BC forms the composite BC pectin in an *in-situ* form, that is, during the formation of the cellulose membrane. The pectin / BC composite film, provided by the invention for medical use, showed transparency greater than 29%, less porosity and permeability to water vapor, transparency greater than 29%, less porosity permeability to water vapor, and excellent sealing properties. It has a simple production process, resulting in a safe and non-poisonous dressing with a great prospect of application in the field of medicine [35].

The CN106074458 invention refers to a method of preparing a fibrous material through the processing of BC with polyacrylonitrile (PAN), resulting in a polymeric composite with the ability to carry drugs with anti-inflammatory capacity for transdermal administration. Such composite, in conjunction with drugs, is used to achieve an appropriate and controlled drug release rate [36].

The CN109966566A patent provides the methodology of preparation of an integrated transdermal wound repair adhesive based on BC. In the form of adhesive, the dressing has a double-layer structure, where one layer is the biopolymer with its nanoporous structure, and the second layer is composed of BC modified to achieve a microporous structure. The inventors reinforce that the integrated BC repair dressing can provide a research base for hernia repair dressings, cartilage scaffolding, and other biological materials [37].

Invention CN113499363 [38] entitled "Biodegradable sports contusion health-care patch and preparation method thereof" discloses a methodology with nano bacterial cellulose powder being used as a gel matrix, which later is cast into the shape of the patch. Saponins and anti-inflammatory drugs are added to the BC gel in order to achieve a controlled release of the medicine component. A long-acting analgesic effect is obtained.

The invention RO137149A2 [39] refers to a hydroactive bandage made with BC for wet management of the dermis. The final dressing contains BC film, such as matrix, propolis extract, natural tocopherol, natural retinol, citric acid, clove, and eucalyptus essential oils. The patent describes their product with a calming, regenerating, anti-inflammatory, scarring, antibacterial, and antifungal effect. This patent is idealized for the local treatment of skin lesions.

Based on its hydrophilicity, biocompatibility, mechanical properties, moisture retention, and porosity, bacterial cellulose has made significant progress in biomedical applications in various medical fields. Nonetheless, there are still a lot of production options on degradable or non-degradable tailor-made biomaterials from BC that have synergistic and superior properties for regenerative medicine, neuroendovascular applications, drug delivery, hemoglobin purification, cancer detection, dentistry, and a range of medical devices [33].

3.3. Bacterial cellulose & fashion/dyeing.

The use of biomaterials in fashion and textile products is becoming more popular, and it opens new possibilities for sustainable designs that promote societal well-being and creativity. Its production methods are the most recent ecological concerns regarding the use of clothing [40]. Biotechnology should be considered by fashion designers to be employed as part of their creative process on textile items. With that being said, bacterial cellulose is a material that has great potential for its use in such industrial segments [41].

BC, in its natural state, is biodegradable and inert, meaning that it does not contaminate nature, being a well-known sustainable biomaterial [5]. In addition, because of its properties, such as high porosity and three-dimensional nanofibrillar structure, it can be dyed and function as an attractive textile matrix for the industry with socio-environmental awareness. BC is also a very suitable material for modifications both *in situ* and *ex-situ*, as it can be manufactured/grown in a variety of shapes, processed to achieve enhanced properties, and functionalized to obtain new applications [42].

Therefore, it is important to discuss aspects related to BC's properties, production methods, polymeric modifications, dyeing, and its application perspectives, focusing on the textile industry [43]. In this sense, the registration of patents linked to dyeing and the textile area of BC are explored to demonstrate the growing potential of applying this polymer in the market.

Patent EP2331699 [40] demonstrates a production method for cellulose synthesized by bacteria. The finality of the invention is to design a universally usable method suitable for large-scale production, allowing homogeneous materials containing cellulose to be produced efficiently at any defined thickness and length unrelated to the used reactor's geometry [44].

The patent CN103481720A [41] provides a method of manufacturing a decorative design using biocellulose. The described methodology is as follows: (a) partially dried BC membranes are soaked in dyeing solutions with the desired colors; (b) the dyed BCs are dried and (c) molded according to the standard of the wanted base painting, resulting in the obtention of a decorative bacterial cellulose painting. Due to BC's relatively dense matrix structure, the decorative paints are fixed effectively after the dyeing process, allowing the final product to have a longer life [45].

The US2017314193A1 invention provides a technique for the creation of fabric and garments. More particularly, it refers to a process for producing a fabric with a multi-shaded appearance. The procedure includes a step of incorporating a layer of the BC. After dyeing, the biopolymer layer is removed from the fabric to get the desired coloring effect [46].

The patent TW201716659A describes a method for dyeing bacterial cellulose. The process consists of an ultrasonic vibration step to disperse the dye in the dyeing solution. After dispersion, the BC membrane is immersed in the solution with the dye and dye solution, where the ultrasonic vibration is used to assist in fixing the color into its pores. In the end, the cellulose is washed to remove unfixed dyes under running water, and the cellulosic fabric is ready to be used [47].

The invention KR102039415B1 describes a method for making a colored BC sheet for mask packages. The method used by the invention results in producing a colored cellulose sheet using natural materials [48].

Patent WO2021146484 describes a biomaterial produced by employing microbial cellulose, which naturally has a nanometric character, to be used as an industrially attractive biotextile that has several desirable and/or improved properties compared to commercial ones [49].

The invention CN109736097A discloses a preparation method for leather made out of bacterial cellulose. The development was prepared by taking some steps and performing a conventional after-finishing, thereby obtaining superfine fiber synthetic leather [50].

The CN114574991 invention describes a preparation method for a durable multifunctional fibrous fabric made of polymers, polyacrylonitrile, and bacterial cellulose biomass. The product went through a dyeing process by adopting a natural fat-soluble dye in a stock solution. Uniform coloring was obtained [51].

The invention CN114108336 is a preparation method of a colored bacterial cellulose mask By means of the method, the bacterial cellulose mask can be endowed with diversified colors, can have the function of natural pigment, and the application field is wider [52].

Patent WO2023007194A2 is related to a process for preparing biobased bacterial cellulose materials. The process involves layers of bacterial cellulose, fungi, or algae and their combinations to form a versatile material. This forms the biobased bacterial cellulose material with different property characteristics and a wide range of applications [53].

The invention CN114197226A relates to a preparation method of colored bacterial cellulose. The preparation method comprises some steps and can endow the bacterial cellulose with diversified colors and can also endow bacterial cellulose with the functions of natural pigments [54].

Some of the characteristics expected by the textile business include a great array of colors, clothing utility, sustainability, and, most importantly, quality [55]. Biotechnological studies are not new, but their application is rapidly reshaping society with a limitless number of beneficial possibilities. Furthermore, science's current scope provides a new way of producing materials and their applications in our society [56].

4. Conclusion

The data survey carried out in this research demonstrates the promising future of BC in the environmental, medical, and textile areas. However, the number of records related to the covered applications is still small, indicating that the research developed in these areas is in its initial stages. Filing patent applications can be extremely challenging; however, registering this material guarantees scientific and commercial security for the innovations described in the document. Consequently, the capital flow destined for scientific research can return to investors and be destined to society, thus reaching its main purpose.

The use of biotechnological materials such as BC in industrial processes can positively influence the economy and consumers. The current inclination to research biomaterials for products is increasing and offers perceptions of science and technology based on sustainable design, aiming at social welfare and innovation. Renewability and sustainability are taken into consideration in every phase of scientific research.

Therefore, this study demonstrates that BC byproducts are expected to replace conventional supplies at an industrial scale soon. This review demonstrates that BC can be used in an extensive range of applications due to its versatility, non-toxicity, and incredible diversity of other interesting properties. However, its production can still cost more than conventional methods, even with its sustainable characteristics, so more research is still needed to enable wide production.

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Conflicts of Interest

The authors declare no conflict of interest.

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