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Electroconductivity of Bacterial Cellulose and Bacterial Cellulose-Based Nanocomposites:

Varieties, Processing and Applications

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Abstract

Bacterial cellulose (BC) is an eco-friendly material with a unique structure and physiochemical properties that can be used in wider areas. Nanocomposites based on BC show superior mechanical, thermal, and electrical properties compared to BC in its pure form. This review explores the various nanomaterials used in BC composites and their effects on overall performance, focusing on the electrical conductivity of the nanocomposites. The work also covers the fabrication and synthesis procedures used to produce BC nanocomposites with improved properties. The BC-based nanocomposites are used in various industries that include biomedical, chemical, electrical, mechanical, pharmaceutical and so on.. This comprehensive review paper provides valuable insights into BC-based nanocomposites and their promising prospects as advanced materials in various applications.

Keywords: Bacterial cellulose, Conductivity, Supercapacitor, Carbon nanotube, Fuel cell

1. Introduction

Cellulose is one of the most widely found polymers on earth, frequently obtained from plant sources. It is also well known for its incredible strength and versatility. However, did you know that some bacteria can create a kind of cellulose that is even more robust than the cellulose found in plants? Bacterial cellulose has the potential to be a useful biodegradable material. The most remarkable properties of bacterial cellulose are its exceptional strength and mechanical properties. Bacterial cellulose has a higher tensile strength than steel, making it incredibly durable and resilient. It is also highly absorbent and has excellent water-holding capacity, making it ideal for use in various applications such as wound dressings, food packaging, and textiles (Revin et al. 2022). It is a biodegradable material, meaning it can change its chemical and potentially physical form when it comes into contact with living organisms (Song et al. 2009). Biodegradable materials are used in different sectors to lessen the adverse environmental effects of our modern way of life. Green biocomposites or biopolymers are substitutes for petroleum-based polymers widely used in automotive and aviation engineering applications (Rahman et al. 2023). There are significant distinctions between plant and bacterial cellulose. Plant fibers are composed of lignin, hemicelluloses, pectin, and cellulose. On the hand, bacterial

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cellulose is made up of pure cellulose nanofibers from different bacteria. BC exhibits high strength and purity without needing additional refinement processes. Bacterial cellulose (BC) has been thoroughly investigated and characterized as a result of these distinctive properties (Bäckdahl et al. 2006). High crystallinity parallel cellulose chains produced by bacteria like *Gluconacetobacter xylinus* give BC its distinctive ribbon-like structure. On the other hand, plant-derived cellulose is made from the walls of plants and comprises densely packed cellulose microfibrils with crystalline and amorphous regions (Nicolas et al. 2021).

Bacteria like Acetobacter, Azotobacter, Gluconacetobacter, Pseudomonas, Salmonella, and Sarcina ventriculi are the source of BC. These bacteria are synthesized extracellularly to produce BC. The most efficient and widely used bacteria for BC production are *Gluconacetobacter xylinum*, *Gluconacetobacter hansenii*, and *Gluconacetobacter pasteurianus* (Shoda and Sugano 2005). These bacteria make cellulose by polymerizing glucose residues into 1,4-glucan chains, which are then extracellularly secreted. These chains assemble and crystallize into ribbons, forming a pellicle. It is a parallel-aligned, three-dimensional network of cellulose nanofibers and is extremely thin and consistent. This network is named as pellicle.

The intra- and intermolecular hydrogen-bonding networks, hydrophobic and van der Waals interactions are the factors that determine the geometry of the pellicles (Revin et al. 2022). A significant amount of crystallinity is present in BC, a sign of a well-organized arrangement of cellulose chains and a higher mechanical strength. Sharp peaks characteristic of the crystalline regions in BC have been seen through X-ray diffraction (XRD) analysis. BC builds a three-dimensional structure by connecting a network of nanofibers. This network gives BC its distinct gel-like consistency and ability to take on a variety of shapes (Ul-Islam et al. 2015). BC in the form of gel can be utilized in the medical fields and separation processes. It is hydrophilic material and has several hydroxyl groups on its surface. BC has a porous structure with voids and gaps in its construction. Because of these pores, BC has a high water-holding capacity, surface area, and permeability, making it suitable for applications such as filtration and drug delivery (Hutchens et al. 2006). BC water holding capacity ranges from 60 to 700 times of its dry weight, depending on the synthesis conditions (Portela et al. 2019). (Guo and Catchmark 2012) found that BC nanofibers have a diameter of around 20-100 nm in diameter. (Grande et al. 2008) found that BC has an average tensile strength of 241.42 ± 21.86 MPa, a maximum elongation value of 8.21 ± 3.01%, and Young's modulus of 6.86 ± 0.32 GPa.

BC does not possess inherent electrical conductivity. But it exhibits electrical conductivity while incorporating conductive additives or depositing conductive materials onto its surface. For example, BC pellicles produced by *Gluconacetobacter xylinum* are incorporated with multiwalled carbon nanotubes (MWCNTs), and the combination produced Electrically conducting polymeric membranes (Yoon et al. 2006). (Yusman et al. 2023) introduced a new way to increase the conductivity of BC by the pulling technique. In this study, it is seen that a wet film of BC is pulled using a tensile testing machine, and the film's conductivity increased by 153% (σ = 19.2 x 10-3 S/cm).

BC can be synthesized with nanomaterials like metal nanoparticles, metal oxide nanoparticles, mineral nanomaterials, and carbonaceous nanomaterials. Biocomposites can be synthesized from chitosan (Ch), alginate (ALG), hyaluronic acid (HA), starch, gelatin (GT), collagen, keratin, polylactic acid (PLA), etc. (Nunes et al. 2021). The strategy of synthesis of BC composite differs according to application purpose. The most common synthesis methods are in-situ, ex-situ, and synthesis from BC solutions (Shah et al. 2013). Overall, bacterial cellulose is an exciting area of research that has the potential to revolutionize a range of industries, from biotechnology and medicine to food packaging and environmental sustainability. Its unique properties and versatility make it a material worth exploring further.

This review provides a thorough overview of recent developments in the investigation of electrical conductivity in BC and BC-based nanocomposites. The paper discusses various techniques to enhance BC's electrical conductivity, such as doping with conductive materials and adding conductive nanofillers.

Additionally, the characteristics and uses of nanocomposites based on BC are investigated. Furthermore, the review provides insights into the challenges and future directions in developing BC and BC-based nanocomposites with enhanced electrical conductivity. Overall, the potential of BC and BC-based nanocomposites as innovative materials for various electrical and electronic applications is highlighted in this review paper.

2. Bacterial cellulose and its nanocomposites

2.1 Bacterial cellulose

Bacterial cellulose (BC) is manufactured by acetic acid-producing bacteria, for example *Acetobacter xylinus* and *Acetobacter hansenii* using a combination of artificial and natural media (Ha et al. 2008; Hestrin and Schramm et al. 1954; Shah, Ha, and Park et al. 2010; Shezad et al. 2009; Ul-Islam, Khan, and Park et al. 2012). The production steps of bacterial cellulose include the creation of glucose chains inside the bacteria, which are then released through tiny openings on the outer layer of the cell membrane. These chains combine to produce microfibrils, which cluster to form cellulose ribbons (Ross, Mayer, and Benziman et al. 1991).

Subsequently, the ribbons give rise to a mesh-like network structure with numerous vacant spaces among the fibers, resulting in an enlarged surface area and a matrix characterized by high porosity (Dahman et al. 2009; Ifuku et al. 2007; Maria et al. 2010; Meftahi et al. 2010). The distinct characteristics of BC arise from the precisely aligned nanofibers, inherent purity, expanded surface area, and diverse pore structures. BC has remarkable qualities because of its unique properties, which include toughness, improved crystal structure, the capability to hold large quantities of water, slow water evaporation rate, the ability to be chemically modified in many ways, as well as being biodegradable, biocompatible, and moldable into 3D shapes during synthesis. Because of these characteristics, BC is a highly versatile material with numerous possible uses in various industries like food, medicine, and the environment (Jeon et al. 2010; Ul-Islam et al. 2012).

2.1.1 Structure of bacterial cellulose

Cellulose is a molecule composed of glucose rings joined together in a linear chain arranged in a ribbon-like flat arrangement. The primary component of cellulose consists of dual rings of anhydroglucose joined together by a covalent bond between an oxygen atom of the first ring and carbon 4 of the adjacent ring, called the b 1-4 glucosidic bond. The number of glucose rings in a cellulose chain varies between 10,000 to 15,000 depending on the cellulose origin. Hydrogen bonding occurs between the hydroxyl groups and oxygen atoms of adjacent ring molecules, stabilizing the bond and resulting in the cellulose chain's linear structure. BC is a biopolymer composed of a basic fibril structure consisting of β-1→4 glucan chains with the molecular formula $(C_6H_{10}O_5)_n$. Inter and intra-hydrogen bonds bind these glucan chains (Ul-Islam et al. 2012).

During biosynthesis, cellulose chains interact through van der Waals and intermolecular hydrogen bonds between their hydroxyl and oxygen groups, promoting the formation of parallel stacks of cellulose chains. These stacks combine to form elementary fibrils, which aggregate into larger microfibrils that are several microns long and have a 5-50 nm diameter. Cellulose is stable as a polymer and exhibits exceptional axial stiffness in its microfibrils due to the cohesive network of hydrogen bonds between the chains, reinforcing trees, plants, some marine creatures, algae, and bacteria. Cellulose microfibrils have crystalline and amorphous regions, but the distribution of these regions is not well understood. However, the crystalline regions within the microfibrils are extracted to obtain cellulose nanocrystals. Figure 1 shows the carbon sources used for fabricating bacterial cellulose, the biosynthesis of BC fibers by *Gluconacetobacter xylinus*, and the shape of bacterial cellulose microfibrils in an electrolyte-free system.

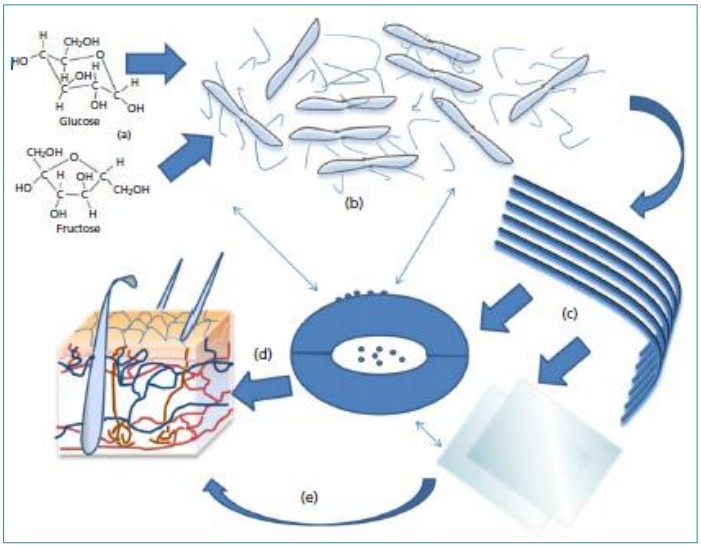

Fig. 1 The different processes involved in bacterial cellulose manufacturing and its potential implementation (Abeer et al. 2014)

The figure also illustrates how bacterial cellulose fibers can be utilized as protective membranes or drug transport systems, with drug molecules entrapped and absorbed in the longitudinal section of a single bacterial cellulose fiber. The route in this figure highlights that bacterial cellulose products have gained significant popularity for their application on the skin or topical treatments (Hoque et al., 2021). However, this can also be used for designing oral and other drug transport systems. The double-headed arrows indicate the interconnectedness of the different processes and applications, all of which focus on the prospective application of bacterial cellulose in facilitating the transportation of drugs (Abeer, Mohd Amin, and Martin et al. 2014).

2.1.2 Bacterial cellulose production

Bacterial Cellulose, a highly researched biopolymer, has been known for over a hundred years. Despite its remarkable structural and physiological characteristics, there are concerns about its economic feasibility, particularly regarding production. Although BC can be produced using the chemically synthesized medium, natural media can also be utilized. Cheaper and waste materials have been used as media for BC production to reduce the cost of production (Ifuku et al. 2007). Various media, such as fruit juices, beer waste, agriculture waste, coconut, pineapple water, molasses, and sugar industry waste, have been reported as potential sources for BC production. BC production typically occurs at the interface of air and liquid in the form of a gel, and the thickness of the resulting BC is influenced by the culture time and carbon sources present in the medium. Microscopic studies reveal that BC has a web-like structure formed from microfibrils composed of cellulose chains arranged in a 3D pattern, creating a net-like structure. As new microfibrils are produced, they are added to the first layer, causing the BC to increase in thickness. The morphology of the BC is mainly dependent on the method of production. The produced BC has a highly porous structure and excellent liquid absorption properties (Ifuku et al. 2007).

Numerous sugars, including glucose, fructose, sucrose, and sugar alcohols like xylitol, mannitol, and sorbitol, have been employed as carbon sources for BC production. Glucose, mannitol, and fructose are the most frequently used carbon sources and have demonstrated outstanding outcomes regarding BC yield. Cellulose is produced by plants and various microorganisms like bacteria, fungi, and algae. Many wild and genetically engineered strains of bacteria have been shown to produce cellulose using a range of carbon origins, including various waste-like substrates. Examples of bacterial species known to produce bacterial cellulose include Alcaligenes, Achromobacter, Agrobacterium, Aerobacter, Gluconacetobacter, Azotobacter,

Sarcina, Pseudomonas, Rhizobium, Rhodobacter and Dickeya, encompassing both Gram-positive and Gram-negative strains. Researchers have been working to increase the yield of bacterial cellulose while decreasing the expense of carbon sources used in production. A few successful attempts have included using soy flour extract, sugarcane molasses, konjac powder hydrolysate, processed rice bark, beet molasses, fruit juice, and tender coconut water employing carbon substrates to synthesize bacterial cellulose. These sources have shown promise in lowering the price of BC production through the utilization of affordable waste products, potentially leading to more comprehensive applications of BC in various mass-volume industries. In the future, Some of these resources may be employed for large-scale mass manufacturing of bacterial cellulose (Poddar and Dikshit 2021; Qiu and Netravali et al. 2014).

2.2 BC-based composites

BC composites are composed of a matrix and materials that reinforce it. The porous arrangement of fibers within BC serves as the matrix, accommodating a diverse range of particles derived from various reinforcing materials. The embedded reinforcing materials offer supplementary characteristics to BC, contributing to its inherent biological and physiochemical properties (Shah et al. 2013). A bacterial cellulose composite was fabricated using materials from bulk polymers to nanoparticles (NPs) (AL-Oqla, Hayajneh, and Hoque et al. 2023). Depending on the characteristics of the reinforcing components, BC composite can be categorized into organic and inorganic materials. These two primary classes can be further subdivided into sub-classifications of BC composites containing metals, polymers, metal oxides, NPs, and solid particles of macroscopic size (Shah et al. 2013). Various BC composites and their preparation methods are listed in Table 2.

Table 1 BC composites, prepared using a diverse range of materials, encompassing both organic and inorganic components

Bacterial Cellulose Composites with-	Organic Materials	Polymers	BC-Ch
			BC-PANI
		Nanomaterials	BC-CNT
			BC-GO
	Inorganic materials	Metals	BC-Ag
			BC-Au
		Metal oxides	BC-Ag
			BC-Au
		Solid particles	BC-Si
			BC-MMt

2.2.1 Strategies for synthesizing BC-based composites

Polymer composites are created in various ways, depending on the properties of the polymer and the material with which it will be combined. The choice of synthesis method also varies depending on the desired application of the composite material (Shah et al. 2013). There are several methods available for producing BC/conductive nanocomposites, which include in-situ and ex-situ fermentation techniques (referred to as in-situ and ex-situ modification of BC), electrospinning, particle deposition, and dispersion-casting methods (Poddar and Dikshit et al. 2021).

2.2.1.1 In-situ BC composites synthesis

This process involves an artificial approach where reinforcing components are introduced into the BC culture media at the start of the bacterial cellulose synthesis process (Poddar and Dikshit 2021). In the in-situ fermentation process, the externally added compound interacts with the hydroxyl (-OH) groups of the glucose

units within the bacterial cellulose framework by forming hydrogen bonds. Generally, in the in-situ methods, hydrophilic and water-soluble substances are commonly employed. Nevertheless, certain research studies have also incorporated hydrophobic compounds like Tween 80, carboxymethyl cellulose (CMC), and hydroxypropylmethyl cellulose (HPMC) using this approach (Shah et al. 2013). Figure 2 demonstrates a general in-situ synthesis procedure of BC.

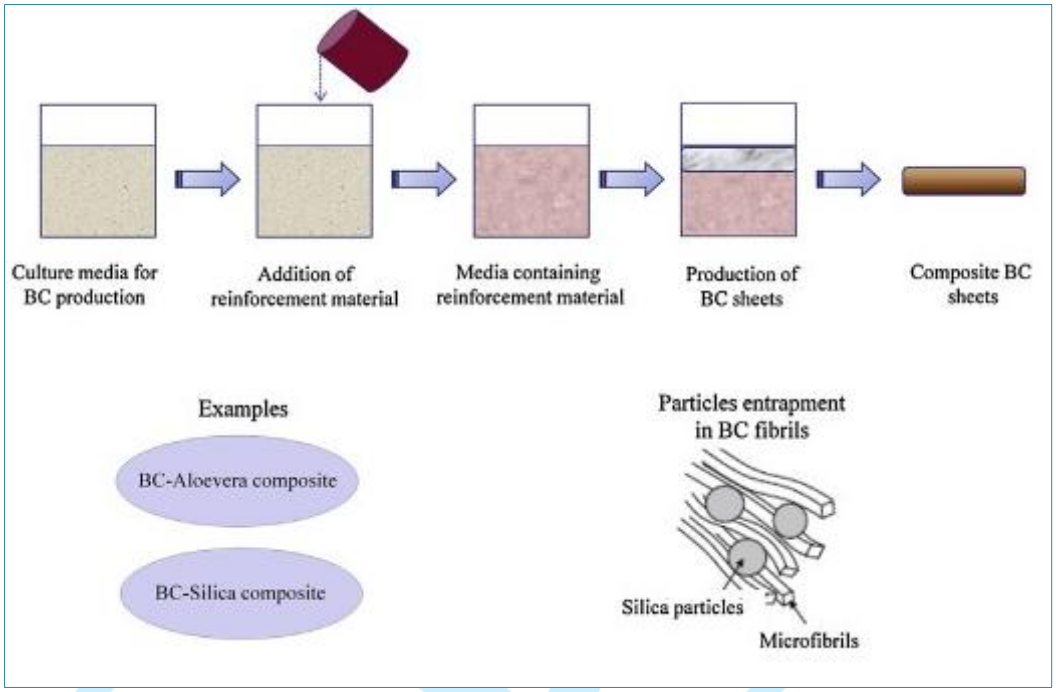

Fig. 2 In-situ synthesis process of BC (Shah et al. 2013)

2.2.1.2 Ex-situ BC composites synthesis

One approach to overcoming the difficulties associated with in-situ BC composite synthesis is incorporating liquids and nanoparticles into a previously prepared structural matrix of Bacterial Cellulose (Shah et al. 2013). In the ex-situ chemical transformation process, a chemical conversion is carried out with reagents in the BC to achieve the desired modification. Nonetheless, a significant drawback of this method is the potential for the bound molecules to be released due to inadequate binding and changes in physiological conditions. The attachment of compounds to bacterial cellulose in this manner can present challenges to the effectiveness of the approach, as it may lead to the dissolution or leakage of the attached compounds. The ex-situ fermentation method consists of a dual-stage process, as depicted in Figure 3. During the first step, conductive polymers, nanofillers, and BC microfibers are dispersed individually in distinct beakers using a compatible solvent under ideal conditions.

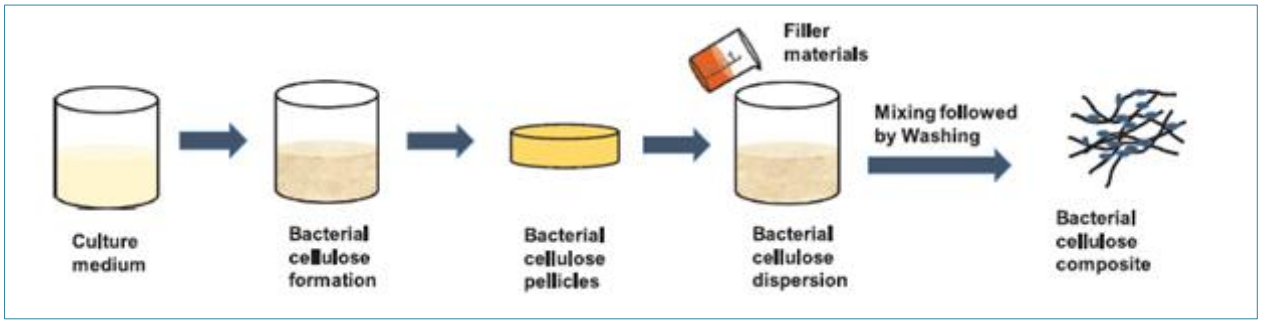

Fig. 3 Ex-situ Synthesis (Poddar and Dikshit et al. 2021)

These filler materials can be incorporated into the BC pellicles by employing mechanical stirring with a homogenizer or intense ultrasonic treatment. Following this stage, the nanocomposites produced through the

ex-situ fermentation method necessitate additional processing, such as rinsing and dehydration, to eliminate any remaining impurities and liquid content from the composites (Poddar and Dikshit et al. 2021). A range of bacterial cellulose and their synthesis methods are presented in Table 1.

Table 2 Various bacterial cellulose and their synthesis methods (Poddar and Dikshit et al. 2021)

Material	Method
PANI	In–situ polymerization
Polypyrrole (PPy)	In–situ polymerization
AuNPs–PEDOT: PSS	Ex–situ polymerization
MWCNTs	Ex–situ polymerization
MWCNT	Ex–situ polymerization
SWCNTs	Ex–situ polymerization
RGO	Ex–situ polymerization
Polypyrrole (PPy)–GO	Ex–situ polymerization
PANI–RGO	In–situ fermentation
Chitosan	Ex-situ Solution penetration
Collagen	In-situ addition
Montmorillonite	Ex-situ Particle penetration
Carbon nanotube	Ex-situ Particle penetration/In-situ addition
Silica	In-situ addition

3. Applications of BC and BC composites

Conductivity comprises one of the essential characteristics of BC, which has been widely studied in recent years. Its conductivity can be attributed to the high crystallinity of the glucose chains in the material. BC is a semi-conductive material, having an electrical conductivity within 10^{-3} S/cm and 10^{-2} S/cm. This conductivity is much higher than other polysaccharides, such as chitosan or alginate. The high conductivity of BC makes it a potential alternative to existing conductive materials for electronic applications. BC has implications in numerous electronic applications. For example, it has been used as a conductive substrate for fabricating electron devices such as biosensors, supercapacitors, thin-film transistors, thin-film capacitors, and light-emitting diodes. BC has also been used in biosensors and biofuel cells. Its conductivity has been demonstrated in glucose, pH, and oxygen detection applications. BC is also suitable for use as a supercapacitor electrode material. So, bacterial cellulose is a complex biomaterial with many potential applications. Additional study is required to comprehend BC's properties better and develop new applications for this exciting material.

3.1 Supercapacitor

In light of the proliferation of portable electronic devices in numerous sectors, there is a growing demand for energy sources that are versatile, affordable, lightweight, and environmentally friendly. We should emphasize the need for novel flexible, and adaptable electrode materials as replacements for the current battery and supercapacitor materials (Xu et al. 2013). Compared to other chemical energy storage devices, supercapacitors have gained much focus due to their high-power density, fast charge/discharge rates, and long cycle life. Nevertheless, the energy density of commercial supercapacitors is minimal. Energy density must be significantly increased while maintaining power density and cycle life to meet the growing energy needs of the next generation of ECs (Long et al. 2014). Long et al. used BC as a template and precursor to produce N-doped activated carbon and carbon/MnO_2 hybrid material as the negative and positive electrode materials for an asymmetric supercapacitor (ASC) device. Figure 4 depicts the process of synthesis. These carbon networks were employed as a foundation for high-capacitance electrode materials. The constructed AC//carbon-MnO_2 asymmetric supercapacitor had a high energy density of 63Wh kg^{-1} in 1.0 m $Na_2SO_{4(aq)}$ solution and a maximal power density of 227 kW kg^{-1}. Moreover, after 5000 cycles, the cycling stability has

retained 92% of its capacitance. The results indicate that this is a promising strategy for synthesizing high-performance energy storage electrode materials using low-cost, eco-friendly biomaterials.

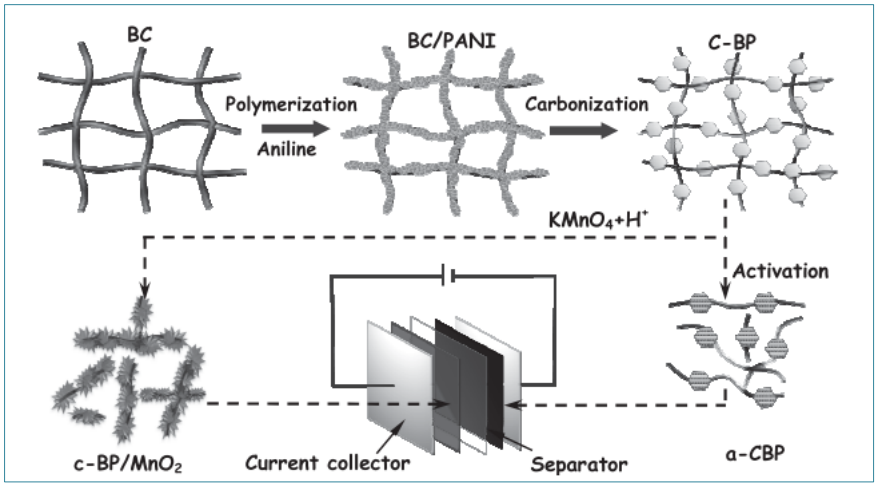

Fig. 4 Diagram depicting bacterial cellulose, known as BC, as a template and precursor for the production of electrode materials for an asymmetric supercapacitor device (Long et al. 2014)

Another investigation(Liu et al. 2015) involved the production of 3D cross-linked PPy/BC/GO (PBG) composites, in which the PPy coating served as conductive networks. This was accomplished by in-situ oxidative polymerization of PPy, which was pre-assembled on a BC/GO layer that was chemically bonded. By varying the weight feeding ratio of pyrrole to BC/GO, various PBG composites with varied mass ratios, designated $PBG_{1:1}$, $PBG_{5:1}$, $PBG_{10:1}$, $PBG_{15:1}$, and $PBG_{20:1}$, were produced. $PBG_{10:1}$ exhibited favorable specific capacitance (Cs), volumetric capacitance (Cv), and cycling retention in both two-electrode and three-electrode capacitor systems, as determined by comparison with prior research.

Polypyrrole plays a significant role in improving the conductivity of supercapacitors. (Peng et al. 2016) used a polypyrrole (PPy) and cobalt sulfide (CoS) coating on bacterial cellulose to improve its electrochemical stability and pseudocapacitance. The supercapacitors based on PPy/CoS/BC have a specific capacitance of 614 Fg^{-1} and an energy density of 54.5 $Whkg^{-1}$ at a power density of 663 Wkg^{-1}. After 300 cycles, 62.4% of the initial specific capacitance is maintained, significantly improving cycling stability. In addition, due to the deposition of the CoS thin layer, the electrical conductivity is slightly reduced, resulting in superior electrochemical performance and increased power density for the BC-based supercapacitor. (Peng et al. 2017) used a coating of copper oxide nanoparticles and polypyrrole to enhance the capacitance. The resulting supercapacitors exhibited a high specific capacitance of 601 F g^{-1}, an energy density of 48.2 Wh kg^{-1}, and a power density of 85.8 W kg^{-1}. Cycling stability was demonstrated by maintaining a specific capacitance of 385 F g^{-1} even after 300 cycles. The copper acetate aqueous solution concentration was adjusted to optimize the electrical conductivity of the electrodes. By oxidatively polymerizing pyrrole with iron (III) chloride as the oxidant and bacterial cellulose as the template, conductive nanocomposite membranes within polypyrrole/bacterial cellulose (PPy/BC) were reported by (Xu et al. 2013). It was discovered that the produced flexible PPy/BC membrane has an electrical conductivity of 3.9 S cm^{-1}, being resistant to bending stress, making it suitable for use in various applications, including energy storage equipment, wearable electronics, and sensors. The membrane effortlessly served as an electrode for a flexible supercapacitor and had a maximal charge capacity of 101.9mA h g^{-1} (459.5F g^{-1}) at a current density of 0.16 A g^{-1}. A conductive nanocomposite has been developed by (Wang et al. 2012), encapsulating bacterial cellulose nanofibers in a polypyrrole layer through the self-assembly of pyrrole and in-situ polymerization. Comparison of the FTIR spectra and X-ray diffraction patterns of BC/PPy nanocomposites to those of unadulterated BC and PPy is shown in Figure 5.

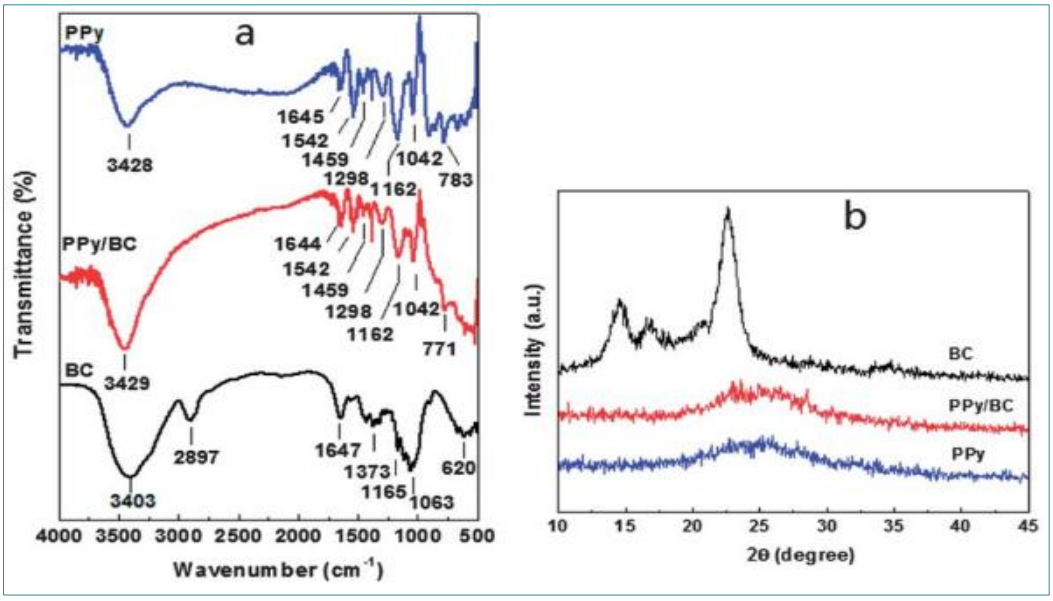

Fig. 5 FTIR spectra and X-ray diffraction pattern of BC/PPy nanocomposites in comparison with those of pure BC and PPy. Pure PPy was synthesized by oxidation polymerization of PPy with 0.5 eq. $FeCl_3$ as oxidant and 1.2 eq. HCl as a dopant in DMF–H_2O (1: 2, v/v) at 0ºC within 6 h (Wang et al. 2012)

To obtain the highest possible electrical conductivity of BC/PPy nanocomposites, specific reaction protocols were applied, including a feeding mass ratio of 1:10 for BC/PPy, a molar ratio of $FeCl_3$/PPy of 0.5:1, a molar ratio of HCl/PPy of 1.2:1, a volume ratio of DMF–H_2O of 1:2, a reaction temp. of 0°C, and a reaction time of 6 hours. Not only did these optimized protocols result in the successful production of BC/PPy nanocomposites, but they also demonstrated their potential applications in supercapacitors. BC/PPy nanocomposites exhibited excellent electrical conductivity of up to 77 S cm^{-1} and mass-specific capacitance of 316 F g^{-1} at 0.2 A g^{-1} current density.

The combination of graphene and polypyrrole is another way that is used in supercapacitor applications. (Liu et al., 2015) described the synthesis and characterization of graphene oxide, bacterial cellulose, and polypyrrole hybrid composite. The composite demonstrated the maximum electrical conductivity (1,320 S m^{-1}) and volumetric capacitance (278 F cm^{-3}) measured for carbon-based electrodes. In addition, it showed excellent retention of gravimetric capacitance (556 F g^{-1}) in asymmetric supercapacitors over 5000 recycling trials and retention of the gravimetric capacitance of 492 F g^{-1} over 2000 recycling trials in symmetric supercapacitors.

Bai et al. (2019) used Graphene/Carbon Nanotube/Bacterial Cellulose (RGO/CNT/BC) substrate for developing and constructing a flexible, highly conductive architecture. Figure 6 depicts the steps of production for PPy/RGO/CNT/BC flexible electrodes as well as SEM images of BC, RGO/BC/CNT, and PPy/RGO/CNT/BC. The PPy was polymerized onto the surface of the RGO/CNT/BC substrate via cyclic voltammetry. PPy/RGO/CNT/BC has excellent areal capacitance (715 mF cm^{-2} at one mA cm^{-2}), rate performance (495 mF cm^{-2} at 30 mA cm^{-2}), cycle stability (86.85% after 5000 charge or discharge cycles), and mechanical strength (57.7 MPa). The PPy/RGO/CNT/BC flexible symmetric supercapacitor has an energy density of 0.0328 mWh cm^{-2} and a power density of 12 mW cm^{-2}. The RGO/CNT/BC structure is promising for wearable electronics. PPy/RGO/CNT/BCx electrodes' current response improves with cycle number from 5 to 20, and PPy/RGO/CNT/BC_{20} performs best in all parameters.

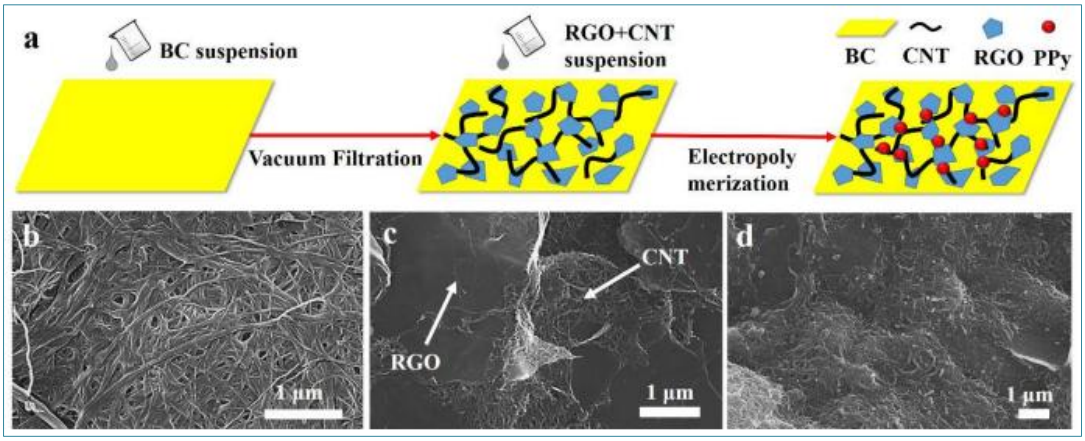

Fig. 6 The illustration of the manufacturing procedures for PPy/RGO/CNT/BC flexible electrodes. The SEM images of b) BC, c) RGO/BC/CNT, d) PPy/RGO/CNT/BC (Bai et al. 2019)

Using composites of polypyrrole (PPy) encased graphene (RGO) on bacterial cellulose, (Ma, Liu, Niu, Zhao, et al. 2016) described a method for making flexible and freestanding electrodes, coating the PPY/RGO composites onto bacterial cellulose results in a flexible PPY/RGO/BC paper that functions as a supercapacitor electrode without needing metallic current collectors, binders, or additives. The electrode achieves impressive results, including a high mass loading of 8.93 mg cm^{-2}, a significant areal capacitance of 2100 $mFcm^{-2}$ at two $mAcm^{-2}$, outstanding rate performance with 1570 $mFcm^{-2}$ retention at 50 $mAcm^{-2}$, and remarkable flexibility capable of bending at arbitrary angles, even up to 180 degrees. It demonstrates a remarkable areal capacitance of 790 $mFcm^{-2}$ and an energy density of 0.11 $mWhcm^{-2}$ when assembled into a symmetric supercapacitor. (Hosseini, Teymouri, et al. 2019) used in-situ fabrication of two series of flexible bio-nanocomposite aerogels. The aerogels are made with silver nanoparticles (AgNPs) and polyaniline (PANI) as conductive agents and are based on BC as a template. The framework of the aerogels shows distinct microstructures: BC/Ag/PANI possesses a core-shell structure, whereas BC/PANI/Ag possesses a branch-grape structure. BC/Ag/PANI has a higher specific surface area (145 $m^2 g^{-1}$) than BC/PANI/Ag (130 $m^2 g^{-1}$), according to BET analysis. Electrical conductivity measurements reveal that BC/Ag/PANI has a higher electrical conductivity (1.25 S cm^{-1}) than BC/PANI/Ag (0.00693 S cm^{-1}). BC/Ag/PANI has a capacitance of 357 F g^{-1} at 0.5 A g^{-1} and 83% cyclic stability over 2000 cycles, whereas BC/PANI/Ag has a capacitance of 232 F g^{-1} at 0.5 A g^{-1} and 74% capacitance retention. In a symmetrical supercapacitor configuration, BC/Ag/PANI demonstrates an energy density of 34 Wh kg^{-1} at a power density of 459 Wh kg^{-1}. Table 3 demonstrates an overview of BC and its nanocomposites as a source of conductive materials.

Table 3 An overview of BC and its nanocomposites as a source of conductive materials

Materials Used	Method	Energy Density	Cycling Capability (specific capacitance retention)	Key Findings	Reference
AC//carbon-MnO_2	BC as template and precursor	63 Wh kg^{-1}	92% after 5000 cycles	Asym, Cs 113 F g^{-1}, P 227 kW kg^{-1}, high-performance energy storage devices based on low-cost, environmentally friendly biomaterials.	(Long et al. 2014)
$PBG_{10:1}$ (two-electrode)	In-situ oxidative polymerization of PPy	77.2 Wh kg^{-1}	93.5% after 5000 cycles	Asym, Cs 486 F g^{-1}	(Liu et al. 2015)

$PBG_{10:1}$ (three electrode)	In-situ oxidative polymerization of PPy	33.8 Wh kg^{-1}	95.2% after 5000 cycles	Asym, Cs 492 F g^{-1}	(Liu et al. 2015)
PPy/BC	-	39.7 Wh kg^{-1}	70.3% after 50 cycles	Sym, Cs 459.5 F g^{-1}, C_d 0.16 Ag^{-1}, flexible, good mechanical adhesion	(Xu et al. 2013)
PPy/BC (DMF–H_2O)[b]	Self-assembly of pyrrole on BC followed by in-situ polymerization	-	88.2% after 1000 cycles	Cs 316 F g^{-1}, C_d 0.2 Ag^{-1}, E_c 77 Scm^{-1}	(Wang et al. 2012)
PPy/CoS/BC-100	Coating of PPy and CoS on BC and PPy on BC	54.5Wh kg^{-1}	62.4% after 300 cycles	Sym, Cs 614 Fg^{-1}, C_d 0.70 Ag^{-1}, E_c 3.7 Scm^{-1}, P 663 Wkg^{-1}	(Peng et al. 2016)
$PPy/RGO/CNT/BC_{20}$	RGO/CNT/BC substrate with PPy coating	0.0328 mWh cm^{-2}	86.85% after 5000 cycles	Sym, C_a 715 mF cm^{-2}, C_d 1 mAcm^{-2}, P 12 mWcm^{-2}, M_s 57.7 MPa	(Bai et al. 2019)
PPY/RGO/BC electrode	Coating of PPY/RGO composites on BC	0.11 mWh cm^{-2}	64.7% after 5000 cycles	Sym, C_a 790 mFcm^{-2}, C_d 1 mAcm^{-2}, P 0.5 mWcm^{-2}, M_a 8.93 mgcm^{-2}	(Ma, Liu, Niu, Zhao, et al. 2016)
PPy/CuO/BC electrode	-	48.2 W h kg^{-1}	-	Cs 487 Fg $^{-1}$(Cv), E_c 7.4 Scm^{-1}, P 85.8 Wkg^{-1}	(Peng et al. 2017)
PPy/BC/GO ($PBG_{10:1}$)	Synthesis of graphene oxide, BC, and PPy hybrid composite	77.2 Wh kg^{-1}	93.5% after 2000 recycling	Cs 492 Fg $^{-1}$, E_c 1320 Sm^{-1}, P 200.1 Wkg^{-1}	(Liu et al. 2015)
BC/Ag/PANI	In-situ fabrication of BC/Ag/PANI	34 Wh kg^{-1}	83 % after 2000 cycles	Cs 357 Fg $^{-1}$, P 459 Wh kg^{-1}, E_c 1.25 Scm^{-1}, S_{sa} 145 m2 g^{-1}	(Hosseini, Teymouri, et al. 2019)

Abbreviations: *Asym- asymmetric; Sym-symmetric; C_d- current density; C_a- areal capacitance; C_s- specific capacitance; Cv- cyclic voltammetry; E_c- electrical conductivity; P- power density; M_s- mechanical strength; M_a- areal mass; S_{sa}- specific surface area*

3.2 Aerogel

Aerogels are promising materials for high-temperature insulation and acoustics because of their high porosity (80%), lightweight (0.004-0.5 g/cm^3), flexibility, large specific surface area, and intricate three-dimensional fibrous structure. But aerogels are too fragile for flexible applications like strain sensors and energy storage devices. Thus, aerogel structural strengthening is challenging. A flexible matrix and high-aspect ratio nanofillers like nanotubes and nanoplatelets can reduce or eliminate these constraints. BC, a renewable and biodegradable polymer, possesses unique properties, including a connected three-dimensional nanofibrous and porous network that gives flexibility and high modulus (Cacicedo et al. 2016; Demilecamps et al. 2015). Salehi et al. 2021 showed a two-step method for synthesizing nano-clay/polyaniline (PANI) and bacterial cellulose (BC) aerogel composites as depicted in Figure 7.

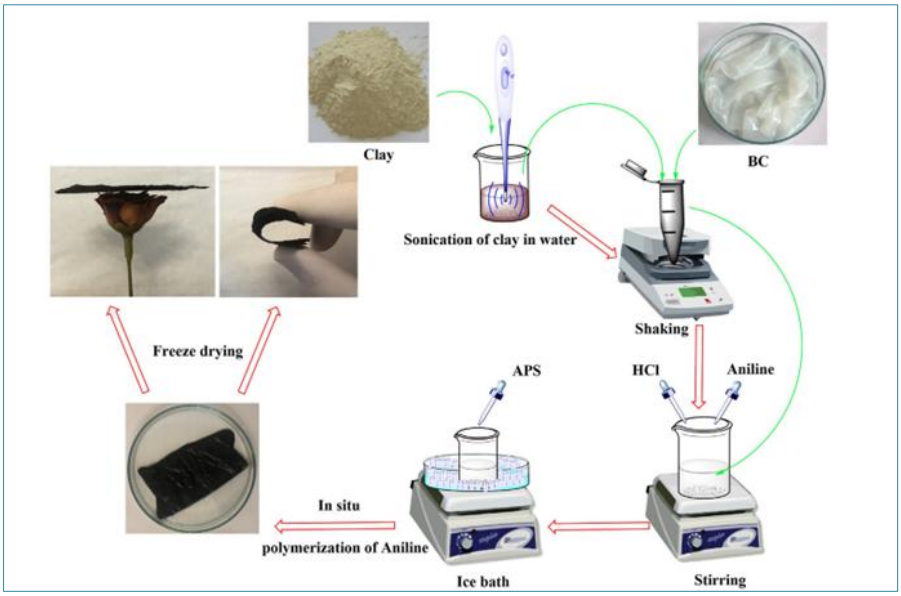

Fig. 7 Different steps of preparation of BC/Clay/PANI nanocomposite aerogels (Salehi et al. 2021)

Spreading clay nanoplatelets within the BC membrane formed an aerogel with a double interconnected network, creating a nano-fibrillated template for aniline polymerization. Some steps are shown in the following figure. The constructed nano-clay/polyaniline (PANI) composite aerogels have better thermal stability, storage modulus, and biocompatibility, making them suitable scaffolds for tissue engineering. The composite aerogel with 5 wt% nano clay exhibited 0.49 S/cm surface electrical conductivity, 16 times higher than the sample without nano-clay. Combining clay and polyaniline enhanced cell adhesion and biocompatibility without mutagenic or carcinogenic consequences. The composite aerogels exhibited improved thermal stability, storage modulus and biocompatibility, displaying them as suitable scaffolds in tissue engineering applications.

In a research, Hosseini et al. 2019 investigated the network formation of two components, i.e., graphene oxide (rGO) and multiwall carbon nanotubes (MWCNTs) in BC matrix-based nanocomposite aerogels. BC/MWCNTs (0.7 wt%) and BC/rGO (0.9 wt%) nanocomposite aerogels exhibited higher percolation values than dynamic mechanical.

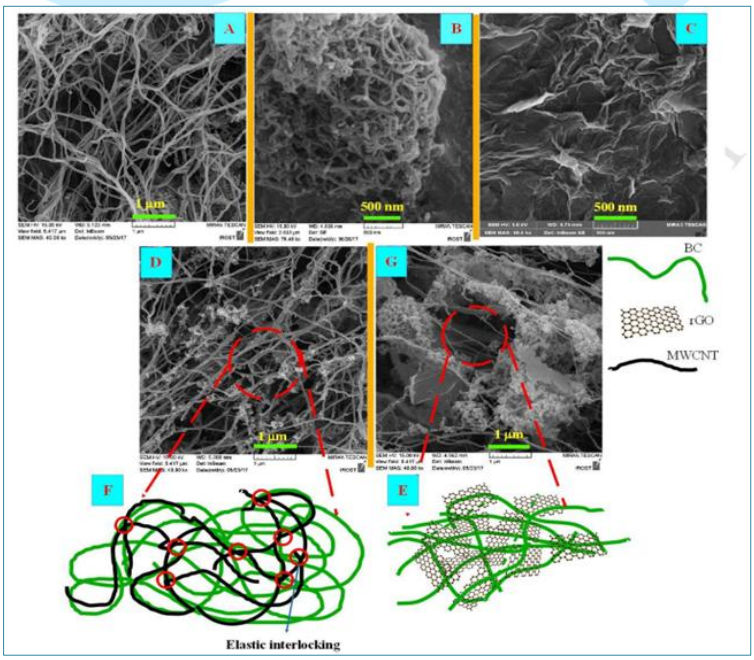

Fig. 8 FESEM micrographs of (A) BC aerogels, (B) as received MWCNT, (C) rGO, (D) BC/MWCNT (0.7 wt%), and (E) BC/rGO (0.9 wt%) nanocomposite aerogels, schematic images about the dispersion of (F) MWCNT and (G) rGO in BC structure (Hosseini, Kokabi, et al. 2019)

Thermal Analysis (DMTA) results: They obtained fractal dimensions of 2.6 (rGO) and 1.86 (MWCNTs). The authors applied DMTA, broadband dielectric spectroscopy, and the box-counting method to evaluate mechanical and dielectric properties, as well as the fractal dimensions of the network. Following experimental data, the study also proposed a modified model that precisely predicted the broadband dielectric properties of BC-based nanocomposite aerogels.

3.3 Sensors

BC has drawn a great deal of attention in the field of sensors due to its exceptional properties and extensive potential applications. BC, a biopolymer produced by specific microbes, possesses outstanding mechanical strength, a remarkable ability to retain water and unmatched biocompatibility. These extraordinary characteristics make BC an extremely desirable material for developing sensor technology. In addition, BC exhibits intriguing electrical properties, such as inherent conductivity, which scientists have ingeniously exploited for various sensor applications, including strain, pressure, humidity, and temperature sensing. The seamless incorporation of BC-based sensors into wearable electronics, biomedical devices, and environmental monitoring systems has produced encouraging results. Utilizing BC in sensors paves the way for cost-effective, eco-friendly, and biocompatible sensing technologies, opening up an intriguing universe of opportunities (Rabbani, Wadud, and Hoque et al. 2022).

3.3.1 Strain sensor

A study (Hosseini, Kokabi, and Mousavi et al. 2018) showed a flexible and light strain sensor made of conductive BC and multiwall carbon nanotubes (MWCNTs) nanocomposite aerogel. This sensor can detect human motion because of its 21-gauge factor and 390 ms reaction time. In-situ biosynthesis of BC/MWCNT hydrogels and supercritical CO_2 conversion produced the nanocomposite aerogel. A strain detector was made from the nanocomposite aerogel's piezoresistive activity at its percolation threshold under tensile strain. Ten cycles of cyclic tension loading and unloading at 2% and 8% strain amplitudes showed a linear trend until a critical strain, beyond which the sensor started to decrease. Thus, in-situ biosynthesis and supercritical CO_2 drying produced a flexible and lightweight conductive BC/MWCNTs nanocomposite aerogel with increased specific surface area, bulk density, reduced pore size, and volume shrinkage due to changes in its morphology and microstructure (Hosseini et al. 2018).

Another study (Xu et al. 2021) describes the development of a susceptible and stretchable natural rubber strain sensor. The sensor contains a network of polypyrrole-modified bacterial cellulose nanofibers (cPPy/BCNF) created by combining natural rubber and polypyrrole-modified bacterial cellulose nanofibers and curing the mixture. Its sensitivity and gauge factor were evaluated over a wide range of straining degrees (0-388%), and its long-term dependability was determined by 3000 cycles at 60% and 180% strain. Multiple human motions: bending the finger, wrist, elbow, and knee joints demonstrated the sensor's reliable and stable sensing performance.

3.3.2 Bio Sensor

Bacterial cellulose nanofibers are used as biotemplates to make gold nanoparticle-bacteria cellulosic nanofiber (Au-BC) nanocomposites in (Zhang et al. 2010)’s work. These nanocomposites encapsulate horseradish peroxidase (HRP) to detect H2O2 at less than 1 µM. Using a CHI 660C electrochemical workstation with a three-electrode system, the modified electrodes obtained through the described methods serve as the working electrode, a platinum wire as the auxiliary electrode, and an Ag/AgCl electrode as the reference electrode. A magnetic Teflon stirrer aids convection in amperometric experiments. This study fabricates Au-BC nanocomposites employing BC nanofibers as biotemplates, immobilizes HRP for sensitive H2O2 detection, and

characterizes their behavior using electrochemical analysis. (Rebelo et al. 2019) developed electrochemical biosensors by altering a BC nanofibril network with an electrically conductive polyvinyl aniline/polyaniline (PVAN/PANI) bilayer as depicted in Figure 9. In-situ, oxidative chemical polymerization of aniline follows surface-initiated atom transfer radical polymerization of 4-vinyl aniline. The nanocomposites have redox peaks of 0.74 V in the positive scan and -0.70 V in the reverse scan at 100 mV/s. These nanocomposites show the emeraldine form of PANI, which has a charge-transfer resistance of 21 and a capacitance of 39 µF. Voltage-sensitive nanocomposites interact well with brain subventricular zone (SVZ) neural stem cells (NSCs).

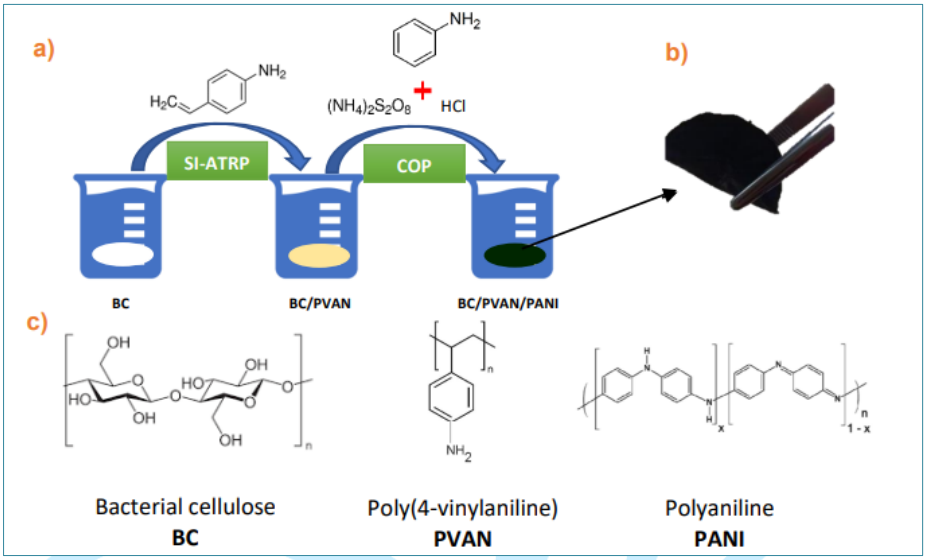

Fig. 9 Nanocomposite BC/PVAN/PANI membranes production. a) SI-ATRP of VAN on BC membranes, followed by COP of aniline in an acidic medium (HCl) with APS as the oxidant. b) Optical image of the BC/PVAN/PANI nanocomposite membrane as prepared. c) Structural composition of BC, PVAN, and PANI (Rebelo et al. 2019)

After seven days, the nanocomposites induce SVZ cells to differentiate into mature neurons with long neurites up to 115±24 µm. Cytotoxic effects are not noticeable. These findings enable efficient nano biosensor devices in regenerative nerve therapy that combine electroactivity and biocompatibility.

A multifunctional sensor that integrates pressure and proximity sensing has been developed in research work by (Guan et al. 2020). The sensor comprises a confined, conductive fiber coated with silver nanowires on bacterial cellulose and PDMS. A flexible and accurate capacitive sensing device is constructed by stacking two fibers. The sensor has a wide detection range for pressure (up to 460 kPa) and high sensitivity (5.49 kPa^{-1}), as well as a wide detection range for proximity (up to 30 cm) and high sensitivity (0.19 cm^{-1}). It is capable of detecting human voices and pulses, as well as pinpointing the location of distant objects. The fiber-based sensor overcomes the limitations of conventional wearable sensors and demonstrates promising application potential in e-skins and wearable electronics. Using biosensors to detect environmental pollutants, such as hydroquinone, Fe3+, Cd(II), and Pb(II), in complex matrices such as wastewater has become popular. They feature portability and high sensitivity for detecting minute pollutant concentrations. In biomedical applications, biosensors have also been used to detect hydrogen peroxide, formaldehyde vapors, and glucose levels. BC has shown promise as a substrate over humidity sensors, with low detection limits and high sensitivity for detecting dopamine and glucose. BC has also been utilized in optical, mass, gas, and strain biosensors, demonstrating its adaptability (Torres et al. 2020).

3.3.3 Other Sensor

Susceptible, wearable, and stable pressure sensors have been developed by (Chen et al. 2022). The sensor is fabricated from a nanocomposite membrane composed of aramid nanofiber-reinforced bacterial cellulose (ANFs@BC) and silver nanowires (Ag NWs). By incorporating aramid nanofibers, the ANFs@BC membrane's

tensile strength increases by 60.6%, from 36.3 to 58.3 MPa. The low electrical resistance of 25.5 is exhibited by the Ag/ANFs@BC membrane containing 0.5 g/m^2 of Ag NWs on its surface. Notably, the sensor retains its sensitivity even after 6000 seconds of constant pressure (5 kPa), demonstrating remarkable long-term stability. The sensor effectively detects a variety of mechanical stimuli, indicating its viability for monitoring human motion in wearable electronics. The polymerization of aniline using ammonium persulfate as an oxidant and BC as a template yielded conductive nanocomposite membranes. Figure 10 illustrates the fabrication process.

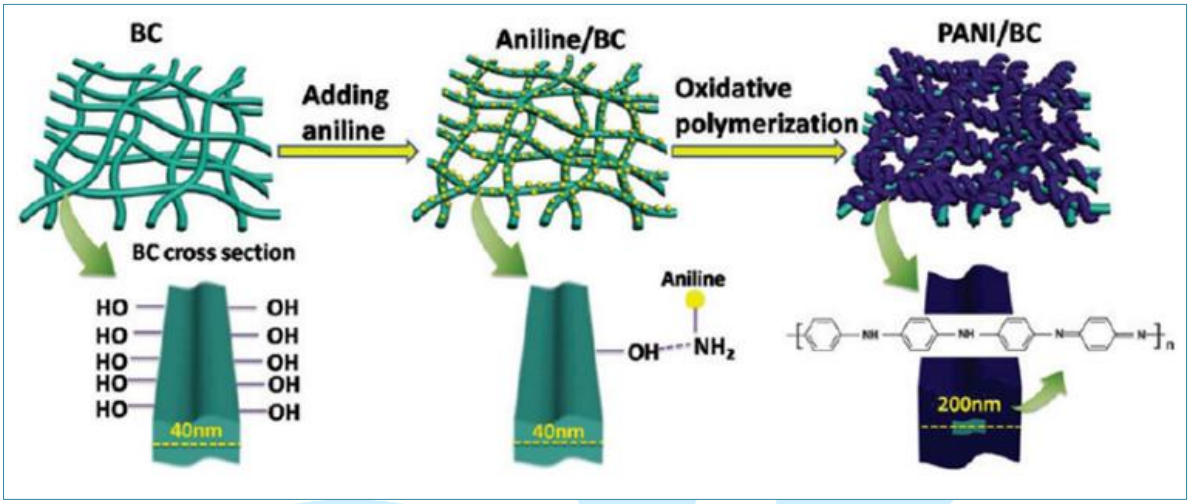

Fig. 10 Schematic illustration of PANI/BC nanocomposites formation (Hu et al. 2011)

PANI-coated BC nanofibrils created a continuous nano-sheath, improving thermal stability. Adjusting reaction time and doping with protonic acids gave the composite membranes 5.0 × 10^{-2} S/cm electrical conductivity. Strain sensitivity made them suitable strain sensors. PANI/BC membranes had 5.6 GPa Young's modulus and 95.7 MPa tensile strength. This discovery suggests a way to make flexible films with high conductivity and mechanical properties for sensors, electrodes, and displays (Hu et al. 2011).

3.4 Graphene-based BC

Due to its superior physicochemical properties, such as its high Young's modulus, outstanding electrical and thermal conductivity, rapid charge carrier mobility, and large specific surface area, graphene is receiving a great deal of attention. It represents the groundbreaking discovery of a 2D crystal composed of a single layer of carbon atoms arranged in a honeycomb lattice(Biswas et al. 2021; Feng et al. 2012; Geim and Novoselov et al. 2007; Li and Shi et al. 2012). Strong sigma bonds and conjugated p-orbitals form electron clouds above and below a planar aromatic macromolecule. Graphene can exist in multiple configurations, including spherical fullerenes (0D), tubular carbon nanotubes (1D CNTs), and layered graphite structures (3D). Graphene and its derivatives, including graphene oxide (GO) and reduced graphene oxide (RGO), have numerous applications in quantum physics, nanoelectronics, catalysis, nanocomposites, and sensor technology(Ge et al. 2012; Qu et al. 2010; Stankovich et al. 2007). Although the initial isolation of graphene in 2004 involved the exfoliation of highly oriented pyrolytic graphite, subsequent processing techniques have been developed to increase productivity and reduce costs, resulting in the creation of various graphene family nanomaterials (GFNs), of which GO and RGO have become the most widely used variants (Novoselov et al. 2004, 2005; Troncoso and Torres et al. 2020). Figure 11 presents the origin of all graphitic forms.

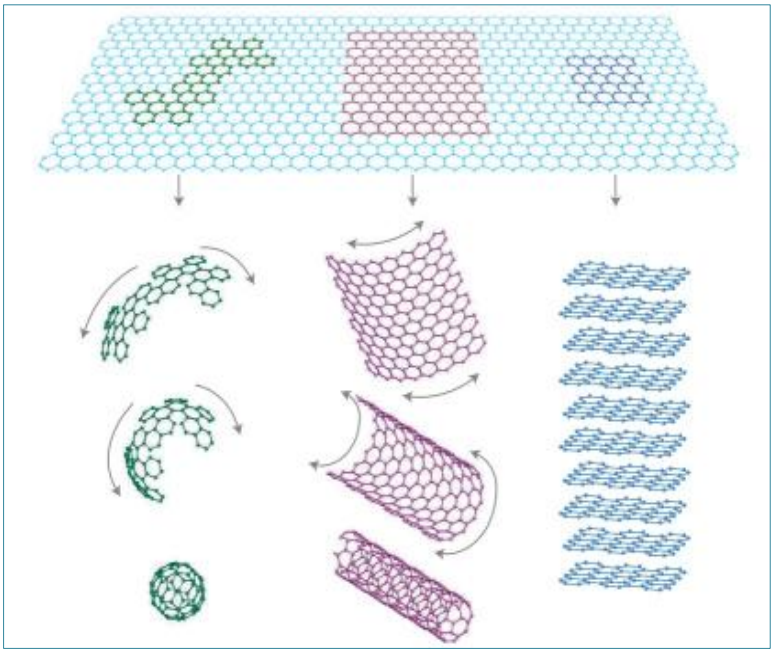

Fig. 11 Origin of graphitic forms. Graphene is a two-dimensional building material for all other carbon materials. It is capable of being coiled into 0D buckyballs, rolled into 1D nanotubes, and stacked into 3D graphite (Geim and Novoselov 2007)

Using polypyrrole (PPy), BC nanofibers, and graphene (RGO),(Ma, Liu, Niu, Wang, et al. 2016) have developed a flexible supercapacitor. The BC nanofiber substrate enabled a high PPY mass loading of 13.5 mg cm^{-2}, which produced remarkable areal capacitance values of 3.66 F cm^{-2} at 1 mA cm^{-2} and 2.59 F cm^{-2} at 50 mA cm^{-2}. With an areal capacitance of 1.67 F cm^{-2}, an areal energy density of 0.23 mWh cm^{-2}, and a maximal power density of 23.5 mW cm^{-2}, the symmetric supercapacitor demonstrated outstanding performance. In addition to its extraordinary stability, the electrode retained 73.5% of its capacitance after 8000 cycles. This low-cost and scalable method has tremendous potential for developing high-performance flexible supercapacitors. In an investigation, (Wan et al. 2018) used a novel technique to create a nanocomposite called bacterial cellulose/graphene/polyaniline (BC/GE/PANI). Bacterial cellulose/graphene (BC/GE) was synthesized using an in-situ membrane-liquid-interface technique, preserving the three-dimensional BC nanofibers while enhancing mechanical properties via uniform graphene dispersion. Polyaniline (PANI) was deposited on BC nanofibers and graphene surfaces to create conductive pathways between graphene nanosheets. This resulted in BC/GE/PANI with an electrical conductivity of 1.7±0.1 S cm^{-1}, exceeding the electrical conductivity of most PANI-based composites. This nanocomposite has considerable application potential in shielding electromagnetic radiation and flexible electrodes.

Another way to combine graphene with BC is to use graphene oxide. Using vacuum-assisted self-assembly, (Feng et al. 2012) combined BC and graphene oxide (GO) to create a flexible and conductive nanocomposite film. The film displayed a homogeneous distribution of GO nanosheets throughout the BC matrix. Compared to BC alone, incorporating 5% GO increased Young's modulus by 10% and tensile strength by 20%. Adding just 1% GO and reducing it in-situ enhanced the electrical conductivity of insulated BC by six orders of magnitude. This study emphasizes the critical role of well-dispersed GO nanosheets in improving the mechanical and electrical properties of the BC/GO composite film, which has potential applications in the development of advanced biochemical and electrochemical devices. By incorporating graphene (GE) into BC, (Luo et al. 2019) have developed a method known as CFLIC for producing highly conductive and flexible paper. Figure 12 illustrates the manufacturing procedure.

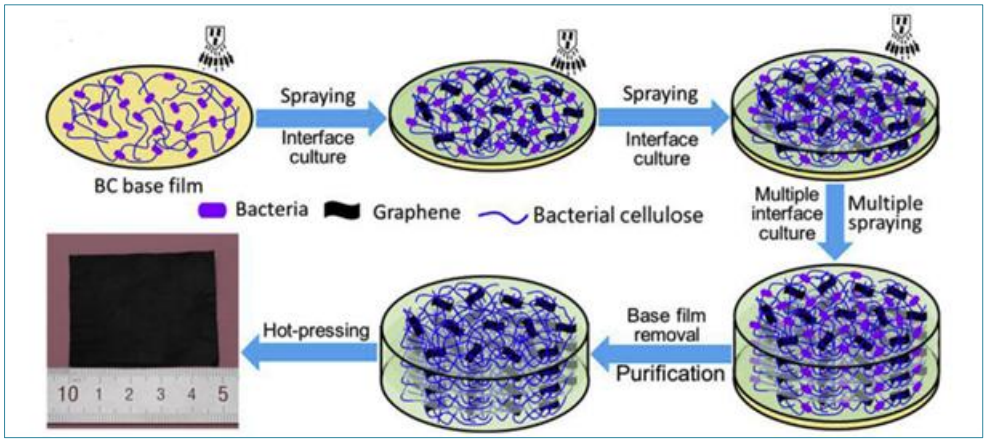

Fig. 12 Processes for manufacturing GE/BC papers (Luo et al. 2019)

Multiple iterations of spraying a GE-dispersed culture medium onto a substrate and promoting interface culture is employed in this method. Compared to BC paper, the resultant GE/BC paper exhibits a remarkable 93% increase in tensile strength and a 10-orders-of-magnitude enhancement in electrical conductivity. In the GE/BC-3 composite, which forms a continuous conductive network, the highest electrical conductivity attained is 101 S m^{-1}. The GE/BC-3 paper's ability to function as an LED's electrical wire demonstrates its high electrical conductivity. These materials exhibit superior mechanical properties, making them promising for applications in electromagnetic interference shielding, batteries, and supercapacitors.

Graphene's potential in biomedical engineering is promising, but its biocompatibility must be due to research indicating that graphene-based nanomaterials may be hazardous to cells and tissues. Sub-100 nm nanoparticles can infiltrate cells, and sub-40 nm nanoparticles can enter the nucleus. It has been observed that graphene oxide (GO) sheets interact with cell membranes, compromising their integrity and disrupting vital processes (Jennifer and Maciej 2013; Troncoso and Torres 2020; Xu et al. 2016). However, BC-graphene nanocomposites demonstrate biocompatibility in several applications as below:

- **Drug delivery system:** Using mouse fibroblast cells (L929), the viability and proliferation of a drug delivery system containing a BC hydrogel laden with GO were evaluated. The results indicated cell viability and proliferation, suggesting that GO-loaded BC hydrogel could be used as a drug delivery system. Similarly, GO-loaded BC hydrogel particles demonstrated promise as a macrophage drug delivery system (Luo et al. 2017; Zhu et al. 2015).
- **Tissue engineering:** MG-63 and NIH 3T3 cells tested BC hydrogel with GO. MG-63 cells increased 110-120% after 24 hours of GO treatment, whereas NIH 3T3 cells increased 80-110%. These findings suggest that graphene oxide could help construct scaffolds or substrates that stimulate cellular growth and tissue regeneration in tissue engineering. Advanced tissue engineering biomaterials can be designed using GO's unique characteristics and BC/hydroxyapatite's porous structure (Ramani and Sastry 2014).
- **Regenerative medicine:** After 24 hours, 95% of PC12 neural cells were viable on a BC/PEDOT film containing GO, indicating biocompatibility. GO film is biocompatible and non-toxic. GO-containing BC/PEDOT films may be used in regenerative medicine, particularly neural tissue regeneration (Chen et al. 2016).
- **Wound dressing:** The biocompatibility of GO-loaded BC hydrogel with human dermal fibroblast cells was demonstrated by 80% cell viability after 24 hours. These results suggest that GO-loaded BC hydrogel could be used as a wound dressing material (Chen et al. 2019).

A comparison of the combination of graphene or graphene oxide with BC is presented in Table 4.

Table 4 Comparison of the combination of graphene or graphene oxide with BC

Materials used	Method	Potential Applications	Key findings	Reference
PPY/BC/RGO	-	Flexible supercapacitors	Areal capacitance 3.66 F cm^{-2} (at 1 mA cm^{-2}), 2.59 F cm^{-2}(at 50 mA cm^{-2}), areal energy density 0.23 mWh cm^{-2}, maximal power density 23.5 mW cm^{-2}, 73.5% retained capacitance after 8000 cycles.	(Ma, Liu, Niu, Wang, et al. 2016)
BC/GE/PANI	In-situ membrane-liquid-interface technique	Shielding electromagnetic radiation, Flexible electrodes	Electrical conductivity of 1.7±0.1 S cm^{-1}, and enhanced mechanical properties via uniform graphene dispersion.	(Wan et al. 2018)
BC/GO	Vacuum-assisted self-assembly	Advanced biochemical and electrochemical devices	Increased Young's modulus by 10%, increased tensile strength by 20%, enhanced electrical conductivity (6 orders of magnitude improvement)	(Feng et al. 2012)
GE/BC	CFLIC method	Electromagnetic interference shielding, batteries, supercapacitors	Electrical conductivity Up to 101 S cm^{-1}	(Luo et al. 2019)

3.5 Carbon nanotube

Due to their extraordinary electrical, physical, chemical, and structural properties, carbon nanotubes (CNTs) have garnered considerable attention in both research and manufacturing. Their remarkable properties have led to numerous applications in various disciplines, such as electronic devices, biosensors, field emission displays, hydrogen storage, and composites. By incorporating small bundles or individual nanotubes of CNTs, polymer composites can achieve superior mechanical strength, electrical conductivity, and thermal conductivity compared to pure polymers (Baughman, Zakhidov, and de Heer 2002; Dong et al. 2005; Jin and Park 2011; Kim et al. 2004; Vigolo et al. 2000).

Embedding multiwalled carbon nanotubes (MWCNTs) into bacterial cellulose membranes creates electrically conductive transparent paper. Before dehydrating, MWCNTs were adsorbed onto the bacterial cellulose hydrogel using an aqueous silk fibroin solution to form transparent nanocomposites. Figure 13 showcases the BC membrane, silk fibroin film, transparent nanocomposite material, CNT-adsorbed BC membrane, electrically conductive transparent paper, and the flexibility of electrically conductive transparent paper. The resulting report exhibited high optical transparency (70.3% light transmittance at 550 nm) and electrical conductivity (5.3×10^{-3} S/cm).

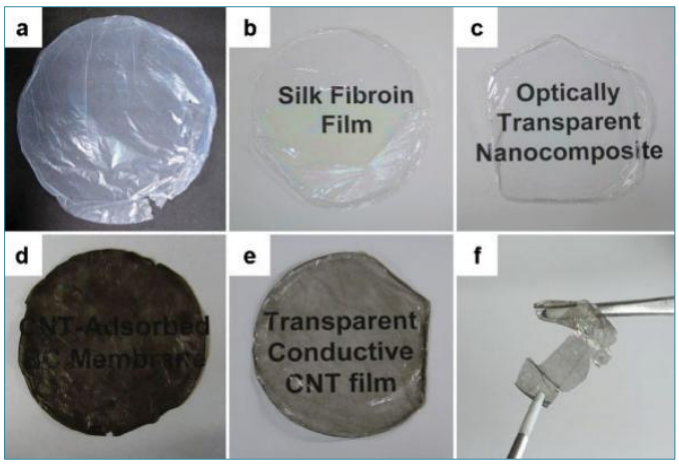

Fig. 13 Images of (a) BC membrane, (b) silk fibroin film, (c) transparent nanocomposite material (d) CNT-adsorbed BC membrane, (e) electrically conductive transparent paper, and (f) the flexibility of the electrically conductive transparent paper (Jung et al. 2008)

It was flexible and retained its initial features. Flexible and transparent electronics may use bacterial cellulose and MWCNTs (Jung et al. 2008). Combining multi-walled carbon nanotubes (MWCNTs) with bacterial cellulose is a well-established method for enhancing electrical conductivity. (Yoon et al. 2006) used this combination which *Gluconacetobacter xylinum* generated, to increase the electrical conductivity of produced membranes. The MWCNTs were uniformly dispersed in a surfactant solution and then immersed in cellulose pellicles for varying amounts of time. Electron microscopy confirmed the dense incorporation of MWCNTs, yielding a conductivity of 1.4×10^{-1} S/cm (approximately 9.6 wt% of MWCNTs). The diameter of the cellulose pellicles' ultrafine network structure was about 30 nm. Due to the interaction between cellulose and oxygen-containing species on the nanotubes, the adhesion of MWCNTs to cellulose surfaces was ascribed. This procedure resulted in the uniform dispersion of MWCNTs, which enhanced the polymeric membranes' electrical conductivity. Wet-spinning was used to create regenerated BC fibers and BC/MWCNT composite fibers in the (Chen et al. 2009)s' study. Ethanol coagulated BC and BC/MWCNTs dissolved in dimethylacetamide/lithium chloride. Incorporating MWCNTs improved the mechanical properties, resulting in a remarkable 430 percent increase in modulus over BC fibers. BC/MWCNTs composite fibers had 1.7×10^{-10} S/cm electrical conductivity (at 1.0 wt% ratio). BC/MWCNTs composite fibers have high electrical conductivity and mechanical performance (modulus: 38.9 GPa). These fibers can be employed as precursors for cellulose-based carbon fibers, improving materials exploration in medicine, mechanics, and electronics.

Low density, high conductivity, and a large surface area are unique properties of carbon-based aerogels. Aerogels comprised of nitrogen-doped graphene exhibit exceptional absorption capacity and capacitance. Flexible graphene foam conductors maintain outstanding electronic conductivity even under extreme strain. Traditional methods are costly and detrimental to the environment. A novel approach proposed by (Wu et al. 2013) employing BC enables the production of carbon nanofiber (CNF) aerogels with a density of 4-6 mg/cm^3 that are both lightweight and fire-resistant. These CNF aerogels absorb organic solvents and oils up to 310 times their weight. Additionally, they exhibit pressure-sensitive behavior, with electrical resistance decreasing linearly with increasing compressive strain. This eco-friendly synthesis method enables the development of absorbents, sensors, battery materials, and catalyst supports. CNF aerogels derived from BC offer versatility and high performance. Dispersed CNT solutions were used to improve nanostructured cellulose's electrical characteristics in one of (Toomadj et al. 2011)'s research work. The conductivity of flexible cellulose films varied with CNT concentration and immersion time. Increasing the time, volume, and quantity of immersion made the film more conductive. For instance, the conductivity of BC pellicles modified with DWCNT increased from 0.034 Scm-1 to 0.15 Scm-1. The specific surface area of BC pellicles modified with MWCNT increased from 0.12 Scm^{-1} to 1.6 Scm^{-1}. These films exhibited a high sensitivity to strain, with more significant resistance variations for increasing CNT concentrations. The dispersion of MWCNTs on the BC surface was superior to that of DWCNTs. These results demonstrate the potential for modified cellulose films to serve as susceptible strain sensors.

Farjana et al. (2013) investigated the strain sensitivity of electrically conductive, flexible cellulose. BC is modified with double and multi-walled carbon nanotubes (DWCNTs and MWCNTs, respectively). Figure 14 shows the preparation processes. Depending on the modifying agent along with the dispersion procedure, the electrical conductivity ranges from 0.034 Scm^{-1} to 0.39 Scm^{-1} for DWCNT-modified bacterial cellulose (BNC) and from 0.12 Scm^{-1} to 1.6 Scm^{-1} for MWCNT-modified BNC. Tensile force is applied to evaluate the strain-induced electromechanical response, and the MWCNT-treated sample demonstrates a maximal gauge factor of 252, indicating high sensitivity. These results establish a relationship between strain sensitivity and conductivity, where samples with more excellent conductivity exhibit greater sensitivity. (Kim et al. 2013) showed the development of conductive composite films using CNTs and BC to facilitate direct electron transfer (DET) of glucose oxidase (GOx).

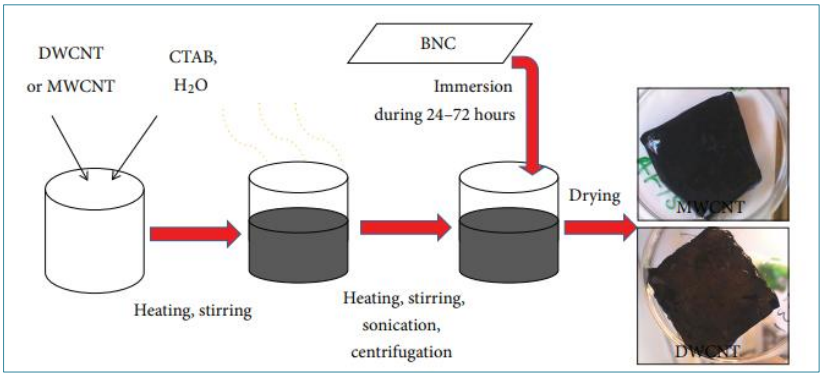

Fig. 14 Procedures associated with preparing the sample (Farjana et al. 2013)

CNTs (diameter range: 6-9 nm) were filtered through a BC hydrogel under vacuum, followed by the successful immobilization of GOx from Aspergillus niger. The electrodes exhibited distinct cyclic voltammograms with well-defined peaks, and the formal redox potential peak was reported at -496 mV (vs. Ag/AgCl), which corresponds closely to FAD. This indicates that DET between GOx and the BC-CNT electrode was effective. Immobilized GOx retained its glucose oxidation catalytic activity. In general, BC-CNT-GOx electrodes present promising prospects for biomedical devices. Table 5 represents the comparison of different BC and CNT-based composites.

Table 5 Comparison of different BC and CNT-based composites

Materials Used	Method	Conductivity	Key Findings	Reference
BC/MWCNT	Immersion of MWCNTs in cellulose pellicles	1.4×10^{-1} S·cm^{-1} (9.6 wt% MWCNTs)	It can be used as membranes	(Yoon et al. 2006)
BC/MWCNT	Wet-spinning of BC/MWCNT composite fibers	1.7×10^{-10} S·cm^{-1} (at 1.0 wt% ratio)	Modulus: 38.9 GPa (430% increase over BC fibers), can be used for cellulose-based carbon fibers	(Chen et al. 2009)
BC/MWCNT	Adsorption of MWCNTs onto BC hydrogel to create transparent nanocomposites	5.3×10^{-3} S·cm^{-1}	Flexible and transparent electronics, Light transmittance: 70.3% at 550 nm	(Jung et al. 2008)
BNC/MWCNT	Modification of BC with DWCNTs and MWCNTs	DWCNT-modified BNC: 0.034-0.39 S·cm^{-1}; MWCNT-modified BNC: 0.12-1.6 S·cm^{-1}	MWCNT-modified BNC: Maximal gauge factor of 252, Strain sensors	(Farjana et al. 2013)
BC-CNT-GOx	Immobilization of GOx on BC-CNT electrodes// Filtration of CNTs through BC hydrogel and immobilization of GOx	-	Formal redox potential peak: -496 mV (vs. Ag/AgCl), Biomedical devices	(Kim et al. 2013)
BC/CNF	BC-based synthesis of CNF aerogels	-	Lightweight, fire-resistant, absorbents, sensors, battery materials, catalyst supports. The density of CNF aerogels: is 4-6 mg/cm^3; Absorption capacity: Up to 310 times their weight; electrical resistance decreases linearly with increasing compressive strain	(Wu et al. 2013)

3.6 Fuel cell

Although plenty of studies have been conducted in the field of fuel cells, only a few studies have focused on using alternative bio-based materials to replace the present perfluorinated sulfonic acid membranes (PFSA) (Scofield, Liu, and Wong et al. 2015). BC is a highly purified, crystalline form of cellulose with a nano-fibrillar three-dimensional structure(Chowdhury, Hoque, and Rahman 2022). It combines eco-friendliness, exceptional thermo-mechanical stability, and minimal hydrogen permeability(Klemm et al. 2011). Evans and colleagues recognized BC's potential as a substitute for PFSAs such as Nafion in 2003(Evans et al. 2003), and there has been growing interest in employing it either as a mechanical reinforcement or in conjunction with a highly proton-conductive phase. BC-constructed fuel cells can draw energy from organic waste as well as water purification, presenting a promising solution for generating sustainable energy (Gadim et al. 2017). Figure 15 illustrates the fabrication process of nanocomposite membrane.

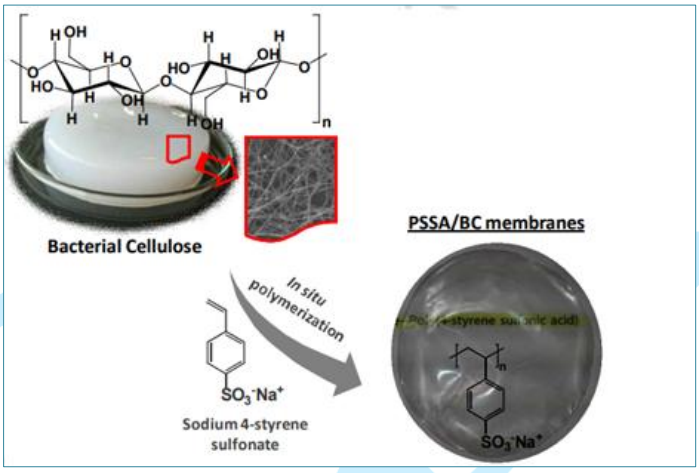

Fig 15 Fabrication process of nanocomposite membrane (Gadim et al., 2017)

Gadim et al. (2017) evaluated the effect of aligned BC nanofibrils on the conductivity of composite proton-conducting liquids, including poly(4-styrene sulfonic acid) (PSSA). At 40% relative humidity (RH), the in-plane conductivity was over ten times greater than the through-plane conductivity, indicating a disruption in proton transport at the PSSA/BC interface. Under nearly saturated conditions (98% RH), this difference decreased to less than 20%, highlighting the function of water in facilitating proton transport across interfaces. Conductivity anisotropy had a negative impact on fuel cell efficacy, which was mitigated by operating in humid conditions. Vilela et al. (2020) proposed a novel method for producing conductive natural-based dividers for polymer electrolyte fuel cells (PEFCs). BC and fucoidan were combined to produce proton exchange membranes (PEMs) made entirely from biomaterials. These membranes exhibit superior thermal-oxidative stability (180-200°C) and dynamic mechanical performance (storage modulus 460 MPa). The protonic conductivity of BC/Fuc membranes increases with increasing relative humidity (RH). At 94 °C and 98% RH, the maximal protonic conductivity of these materials is 1.6 mS cm^{-1}. BC and fucoidan-based membranes have the potential to serve as sustainable alternatives to conventional PEMs for PEFCs, which bodes well for the development of green energy technologies.

Polyaniline (PANI) is one of the most commonly used polymers that is used with BC. BC and PANI have enhanced microbial fuel cell (MFC) anodes. In-situ chemical oxidative polymerization generated and spread PANI onto BC fibers as a continuous phase in (Mashkour, Rahimnejad, and Mashkour et al. 2016)'s research work. BC/PANI nano biocomposites with hydrogel and conductivity were excellent for active microbial biofilm development on the anode surface. The BC/PANI anode outperformed a graphite plate in MFCs, obtaining a maximum power density of 117.76 mW/m^2 at a current density of 617 mA/m^2. BC's hydrogel properties allowed biofilm nutrient transfer, boosting bacterial colonization and reducing spoiling. PANI increased BC's

conductivity, creating a conductive porous composite that lets bacteria transport electrons to the external circuit. A carbon fiber-embedded bacterial cellulose/polyaniline (CF/BC/PANI) nanocomposite electrode has been developed for MFC applications to produce renewable energy from biomass waste. The CF/BC/PANI electrode was made by integrating carbon fiber into bacterial cellulose fibers and synthesizing polyaniline on BC nanofibers. Various characterization techniques like SEM, Fourier-transform infrared spectroscopy, X-ray diffraction, etc. analyses were used. CF/BC/PANI electrode MFC studies showed a current density of 0.009 mA/cm^2. The CF/BC/PANI electrode's excellent milieu for microbial biofilm development highlights its MFC potential. The study emphasizes the need to develop renewable source materials and optimize MFC electrodes to harness energy from organic waste and wastewater treatment, giving a promising approach to sustainable energy generation (Trindade et al. 2020).

UV-induced polymerization was used to investigate the sorption and transport properties of proton-conducting membranes made of BC biopolymer grafted with 2-acrylamido-2-methyl-1-propanesulfonic acid (AMPS) by (Lin et al. 2013). Pulsed field-gradient nuclear magnetic resonance (PFG-NMR) technology was employed to determine the self-diffusion coefficients of water and methanol. Self-diffusion coefficients of the AMPS-g-BC membrane were 1.48×10^{-5} cm^2/s for water and 5.30×10^{-6} cm^2/s for methanol. The permeability of methanol was measured to be 5.64×10^{-7} cm^2/s, which is approximately 42% of the value for Nafion 115. With a water-to-methanol absorption ratio (i.e., $\lambda_{CH3OH}/\lambda_{H2O}$) of 0.07, the sorption characteristics indicated a higher affinity for water than for methanol. These results suggest that the AMPS-g-BC membrane functions effectively as a barrier against methanol and shows promise as a solid electrolyte for direct methanol fuel cells. On the other hand, N-butyl guanidinium tetrafluoroborate (BG-BF_4), a thermally stable protic ionic liquid, has an ionic conductivity of $8x10^{-3}$ S/cm at ambient temperature and 0.18 S/cm at 180°C. BG-BF_4-impregnated BC generates a composite membrane with 80 wt% of the ionic liquid, 35 MPa tensile strength, and $4.5x10^{-4}$ S/cm ionic conductivity at 180°C. Maximizing BC saturation with BG-BF_4 to 95 wt% yields an outstanding composite membrane with a lower tensile strength of 6 MPa but excellent ionic conductivity of $5.2x10^{-2}$ S/cm at 180°C. Incorporating PANI further enhances the ionic conductivity, resulting in $4x10^{-3}$ S/cm at 180°C for the BC film containing 80 wt% of the ionic liquid. BC/BG-BF_4 and BC/PANI/BG-BF_4 composites have 1000 MPa and 835 MPa storage modulus at 180°C, respectively. BC/BG-BF_4 may be a proton-conducting membrane for fuel cells working at high temperatures without water. The BC/PANI nanocomposite impregnated with the ionic liquid offers an economical alternative with a proton conductivity of $4x10^{-3}$ S/cm at 180°C and activation energy values (Ea) of 18 to 45 kJ/mol, suggesting a Grotthuss mechanism for proton transfer (Rogalsky et al., 2018). Mixing phosphorus and copper into bacterial cellulose (Li et al. 2019) developed a highly effective catalyst for MFCs. The techniques of freeze-drying and high-temperature pyrolysis were utilized. The resulting catalyst had a specific surface area of 580.09 m^2/g. Cu and P insertion increased the catalyst's catalytic activity by adding active sites to its pores. The coating of the catalyst onto the MFC air cathode resulted in a highest output power of 1177.31 mW/m^2 and a current density of 6.73 A/m^2. This alternative enhances bioelectrical generation and wastewater treatment while reducing costs. The MFCs' long-term stability was adequate.

BC pellicles were infused with carboxylic multi-walled carbon nanotubes (c-MWCNTs) to produce composite membranes with electrical conductivity in (Lv et al. 2016)'s research work. Figure 16 illustrates the EBFC with 3D hybrid BC/cMWCNT electrodes. Biocathode and bioanode were prepared using a straightforward adsorption method. With an open circuit voltage of 0.76 V and a maximum power density of 55 uW/cm^3, the resulting enzyme biological fuel cell (EBFC) demonstrated exceptional performance. Various characterization techniques are used to determine

The performance of composite membranes: The cell was stable for thirty days. Over 30 days, the biofuel cell with glucose oxidase (GOD) at the anode and laccase (Lac) at the cathode showed a gradual decline in open circuit voltage while remaining stable. Based on its electrochemical behavior, the Lac/BC/c-MWCNTs

electrode exhibited distinct redox peaks corresponding to laccase redox transition. At a cell voltage of 0.37 V, the biofuel cell, which was powered by glucose/O_2 and modified with glucose oxidase (GOD) and laccase (Lac), attained a maximum current density of 0.4 mA/cm^3 and a maximum power density of 55 W/cm^3. These results demonstrate the potential of BC/c-MWCNTs as a biofuel cell carrier.

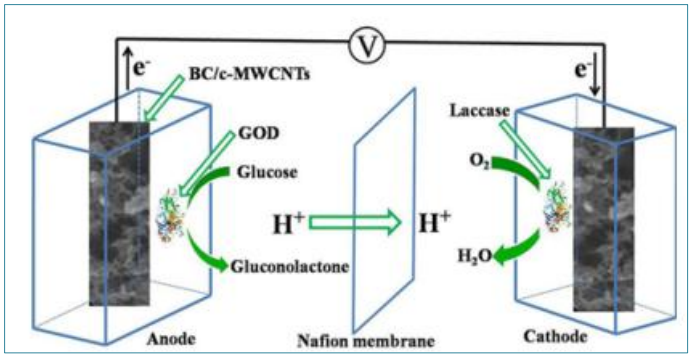

Fig. 16 Demonstration of the EBFC with 3D BC/cMWCNTs hybrid electrodes (Lv et al. 2016)

BC-CNT-PANI is a proposed new bioanode for microbial fuel cells (MFCs) by (Mashkour et al. 2020). BC was coated with CNT to create BC-CNT, and then PANI was electro-polymerized onto BC-CNT to create BC-CNT-PANI. The impedance analysis revealed that R_{ct} (charge transfer resistance) increased from 14.5 to 72 for BC-CNT, whereas R_{ct} decreased by 50% for BC-CNT-PANI. After biofilm formation, BC-CNT-PANI demonstrated twice the capacitance of BC-CNT. BC-CNT-PANI enhanced the power density of SCMFCs (super capacitive MFCs) by 20% compared to BC-CNT. This study introduces a flexible hydrogel anode with increased redox activity, capacitance, and power density for SCMFCs. In the SCMFC system, the dual-anode configuration of BC-CNT-PANI and BC-CNT obtained the highest capacitance and power density. Table 6 outlines various BC in fuel cell applications.

Table 6 BC in fuel cell applications

Materials Used	Results	Key Findings	Reference
PSSA/BC	In-plane conductivity >10x greater than through-plane conductivity at 40% RH, reduced to <20% at 98% RH	Disruption in proton transport at PSSA/BC interface, humidity facilitates proton transport, anisotropy negatively impacts fuel cell efficacy, PSSA/BC-based membrane electrode assemblies achieved high power density in fuel cells.	(Gadim et al. 2017)
BC/Fuc	maximal conductivity of 1.6 mS cm^{-1} at 94 °C and 98% RH, thermal-oxidative stability in the 180-200 °C temperature range, and strong dynamic mechanical performance (storage modulus ≥460 MPa).	Protonic conductivity of BC/Fuc membranes increases with increasing RH, with potential as sustainable alternatives for PEFCs.	(Vilela et al. 2020)
BC/PANI	maximum power density 117.76 mW/m^2	BC/PANI anode outperforms graphite plate in MFCs, hydrogel properties boost bacterial colonization and electron transport.	(Mashkour et al. 2016)
CF/BC/PANI electrode	maximum current density 0.009 mA/cm^2	CF/BC/PANI electrode shows potential for microbial biofilm development and renewable energy generation from biomass waste.	(Trindade et al. 2020)

AMPS-g-BC membrane	water self-diffusion coefficient: 1.48×10-5 cm2/s, methanol self-diffusion coefficient: 5.30×10-6 cm2/s	AMPS-g-BC membrane functions as a barrier against methanol, a potential solid electrolyte for direct methanol fuel cells	(Lin et al. 2013)
BC/BG-BF_4 composite	ionic conductivity $5.2x10^{-2}$ S/cm at 180°C	BC/BG-BF_4 and BC/PANI/BG-BF_4 composites exhibit excellent ionic conductivity and storage modulus for high-temperature fuel cell applications.	(Rogalsky et al. 2018)
Cu and P-inserted BC catalyst	highest output power 1177.31 mW/m^2, current density 6.73 A/m^2, specific surface area 580.09 m^2	Cu and P-inserted BC catalyst enhances bioelectrical generation and wastewater treatment in MFCs	(Li et al. 2019)
EBFC with c-MWCNT-infused BC membrane	open circuit voltage 0.76 V, maximum power density 55 uW/cm^3	BC/c-MWCNT composite membrane demonstrates exceptional performance in enzyme biological fuel cells	(Lv et al. 2016)
BC-CNT-PANI bioanode	50% decrease in charge transfer resistance, a 20% increase in power density compared to BC-CNT	BC-CNT-PANI enhances redox activity, capacitance, and power density in SCMFCs	(Mashkour et al. 2020)

4. Challenges and future aspects

The elevated expenses associated with carbon-based nanofillers, which are responsible for achieving high electrical conductivity, pose a significant obstacle for widespread commercial applications requiring large-scale production of bacterial cellulose composites. Enhancing electrical conductivity while utilizing lower fillers, incorporating cost-effective conductive nanofillers, and reducing fabrication costs emerge as crucial impending hurdles that must be addressed to successfully commercialize bacterial cellulose composites (Poddar and Dikshit et al. 2021). In recent times, there has been a significant surge in research interest surrounding bacterial cellulose (BC). The diverse factors driving BC research growth are the remarkable applications, various market demands, synthesis methods, limitations, economic viability, and strategies for creating BC composites. Incorporating different conductive nanofillers (such as graphene sheets, metal oxides, and carbon nanotubes (Chinnappan et al. 2022) into the matrix of bacterial cellulose has demonstrated a remarkable enhancement in the electrical conductivity of the resulting nanocomposites. Due to their inherent biodegradability, optoelectronic devices based on bacterial cellulose are increasingly infiltrating and supplanting conventional petroleum-based plastic optoelectronic devices in various applications, including supercapacitors, flexible electronics, touch screens, biosensors, and electromagnetic shielding. The emphasis is shifting toward the practical utilization of BC and BC composites, particularly in developing conductive materials, artificial organs, and display devices utilizing BC as a substrate material (Ul-Islam et al. 2015).

5. Conclusions

Cellulose is the most prevalent polymer and widely available organic material on our planet. The crystalline arrangement of cellulose plays a vital role in determining the durability and robustness of materials (Jeon et al. 2014). Bacterial cellulose (BC), due to its inherent composition and remarkable physicochemical characteristics, such as superior mechanical potency, crystalline structure, porous fibrous architecture, and liquid uptake abilities, has surfaced as a sophisticated biomaterial (Ul-Islam et al. 2015). Within composites, the foundation is a matrix that serves as a supporting framework, while reinforcement components bestow additional attributes to the matrix (Shah et al. 2013). The synthesis of a wide assortment of composites is made possible by utilizing an extensive array of synthetic methods and employing various matrices and reinforcing materials. Polymers, including biopolymers, have been infused with many reinforcement materials to create multifunctional composites (Maneerung, Tokura, and Rujiravanit 2008; Ul-Islam et al. 2011).

Scientists have extensively explored diverse processes for synthesizing bacterial cellulose and bacterial cellulose-based composites. Integrating bacterial cellulose with metals, metal oxides, graphene, nanomaterials, and solid particles, among others, significantly modifies physical and chemical properties. These modified properties have found applications in various sectors, showcasing the potential for wide-ranging utilization of these composites. The electrical conductivity of bacterial cellulose and its composites is a highly significant property that exhibits a diverse range of applications across various sectors.

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