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Review

Green Nanocomposite Electrodes/Electrolytes for Microbial Fuel Cells—Cutting-Edge Technology

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Abstract: Fuel cell efficiency can be improved by using progressive electrodes and electrolytes. Green nanomaterials and green technologies have been explored for the manufacturing of high-performance electrode and electrolyte materials for fuel cells. Platinum-based electrodes have been replaced with green materials and nanocomposites using green fabrication approaches to attain environmentally friendly fuel cells. In this regard, ecological and sustainable electrode- and electrolyte-based membrane electrode assemblies have also been designed. Moreover, green nanocomposites have been applied to form the fuel cell electrolyte membranes. Among fuel cells, microbial fuel cells have gained research attention for the incorporation of green and sustainable materials. Hence, this review essentially focuses on the potential of green nanocomposites as fuel cell electrode and electrolyte materials and application of green synthesis techniques to attain these materials. The design of and interactions with nanocomposites have led to synergistic effects on the morphology, impedance, resistance, power density, current density, electrochemical features, proton conductivity, and overall efficiency. Moreover, we deliberate the future significance and challenges of the application of green nanocomposites in electrodes and electrolytes to attain efficient fuel cells.

Keywords: green; nanocomposite; electrode; electrolyte; synthesis; microbial fuel cell

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1. Introduction

Environmental pollution has turned research direction towards the need for clean production and storage systems [1]. Fuel cells are considered to be very demanding clean energy systems owing to their environmental friendliness, fuel cell efficiency, energy density, and other properties [2–4]. Various fuel cell kinds have been developed and considered up until now [5]. Among various fuel cell types, the polymer electrolyte membrane fuel cell (PEMFC) has efficient green energy technology [6]. PEMFC has been the most widely adopted fuel cell in transportation. It has the advantages of inexpensiveness, low emissions and environmental pollution, low temperature operation, fast start-up, and high efficiency [7]. PEMFC works on the principle of chemical energy conversion to electrical energy through reactions occurring at electrodes [8]. To increase the oxygen reduction reactions at electrodes and the power output, catalysts have been used [9]. Among the various electrocatalyst materials that have been used are platinum-free electrocatalysts, metal-free electrocatalysts, platinum-based electrocatalysts, alloy-based electrocatalysts, etc. [10]. In this regard, platinum (Pt)-based electrocatalysts have been efficiently used [11]. However, Pt electrocatalysts may have the disadvantages of price and poisoning [12]. Therefore, the

current generation of PEMFC faces drawbacks for large-scale production. The challenges involve the expenses and durability of the catalysts. The platinum electrocatalysts have incurred high costs for fuel cell electrodes.

To resolve these problems, new electrode materials have been continuously researched. Various nanomaterials can replace Pt in fuel cell electrode catalysts [13]. Recently, membrane electrode assembly (MEA) was developed to enhance the fuel cell efficiency and current density and to lower the cost expenses [14]. The performance of MEA-based fuel cells relies on factors such as electrode design, the gas diffusion layer, and cell temperature.

In addition to fuel cell electrodes, polymer proton exchange membranes were formed as imperative components of PEMFC [15]. Instead of traditional Nafion membrane, various non-perfluorinated materials were researched for low-cost production and environmental effects [16]. In this regard, sulfonated polymers and nanocomposites have been developed [17]. Graphene and derived nanomaterials have been found to be promising for fuel cell applications such as polymer proton exchange membranes and electrodes [18,19]. The use of graphene improved the electrode catalytic activity [20,21]. Microbial fuel cell performance and efficiency are usually assessed in terms of electrical parameters such as current density, power density, potential difference, and internal resistance. Moreover, the performance and efficiency of microbial fuel cells depend on biodegradation efficiency and the removal of organics.

Among polymers, green or ecological polymers are preferred [22]. Synthetic green polymers are made from naturally occurring or biologically produced materials, which are easily biodegradable. Green polymers are produced using green or sustainable chemistry. Green chemistry seeks to decrease pollution at its source. Natural polymers are also usually green. On the other hand, several synthetic polymers, such as Nafion, are not synthesized using green chemistry and are not biodegradable. The use of non-green polymers is continuously worsening the environmental pollution problems. For polymer proton exchange membranes, green polymers such as poly(vinyl alcohol), poly(ethylene oxide), poly(vinyl pyrrolidone), etc., are preferred to replace the Nafion membranes [23]. The membrane designs were altered for enhanced proton conduction. The crosslinked polymers with sulfonated graphene or graphene oxide have also been used to form efficient proton exchange membranes using green routes [24].

Hence, this state-of-the-art review focuses on recent attempts to develop green nanomaterials for fuel cell components, especially electrodes and electrolyte membranes. Moreover, the green approaches used for the development of fuel cell materials are also described. This review elucidates the fundamentals, features, and significance of designing green nanomaterials for fuel cell electrodes and electrolytes. Consequently, the material properties and fuel cell performance were analyzed. Specifically, the review outline contains Section 1, i.e., the introduction; Section 2, on green nanocomposites for microbial fuel cell electrodes; Section 3, describing the use of green nanocomposites in solid electrolyte membranes; Section 4, dedicated to the significance and challenges of using green nanocomposites in microbial fuel cells; and Section 5, our conclusions. All the sections thoroughly and comprehensively describe the outlined content. In this leading-edge review, various notable prospects of the green nanocomposite-based electrodes and solid electrolytes for microbial fuel cells are highlighted. The design variations, essential features, and significance of related green nanocomposites are especially emphasized. To the best of our knowledge, such an explicit recent review on green nanocomposites for microbial fuel cells, with a specified outline and a thorough interpretation of the recent literature, has not previously been reported.

2. Nanocomposites in Fuel Cells: Green Nanocomposites for Microbial Fuel Cell Electrodes

Fuel cells offer clean power foundations for various electronics, automobiles, and power production systems, owing to their environment friendliness, enhanced power density, and power conversion efficiency [25,26]. Fuel cell components have commonly

used Pt-based materials [27–29]. However, due to its high cost and toxicity, it has been attempted to replace Pt with low-cost, green materials [30]. Fuel cell electrodes, catalysts, membranes, etc., have been designed using green polymers and nanomaterials and green fabrication processes [31–33]. Using novel nanomaterials has improved the performance of fuel cells component [34]. In this regard, polymeric materials and nanocomposites have been fabricated through facile physical and chemical methods [35]. In situ polymerization, solution mixing, and melt blending have been used to form these materials [36]. In nanocomposites, the type of polymer, nanofiller content, dispersion, and morphology define the performance of fuel cell electrodes [37–39]. Particularly, these materials have been applied to substitute platinum catalysts in fuel cell cathodes [40]. In polymer proton exchange membranes, Nafion has been applied due to its superior selectivity and ion exchange features [41]. Nanomaterials derived from Nafion, polysulfone, perfluorosulfonic acid, etc., have also been investigated [42–44]. Recently, eco-friendly polymers, including poly(ethylene glycol), polyacrylic acid, poly(vinyl alcohol), etc., have been employed as electrolyte membranes [45]. Green polymers and derived nanocomposites have also been used to develop electrode materials and catalysts for PEMFC [46]. The nanocomposite electrodes and electrolytes exhibited high power density, ionic conductivity, fuel cell efficiency, and long working lives. Zhang et al. [47] designed nanocomposite electrodes using Nafion, polyacrylic acid, and Pt/C catalyst. The electrode was applied as a cathode in H_2/air and H_2/O_2 fuel cells. Polymers and Pt/C particles developed a percolation network for ionic or electron conduction through the nanocomposite. Wang et al. [48] reported a Pt/C- and nanocomposite-loaded cathode. The electrode had inter-linked morphology and a high surface area. The cathode had a high power density of 1.090 Wcm^2 for the H_2/O_2 fuel cell. Shabani et al. [49] established a fuel cell electrode of a poly(ether sulfone) and Pt/C catalyst nanocomposite. The electrode had a large specific surface area and fine morphology, proton conductivity, and oxidative stability. Similarly, numerous nanocomposite systems have been designed for fuel cell components.

Microbial fuel cells employ microorganism-based catalysts for the conversion of chemical energy to electrical energy [50–52]. The function of microorganisms is to produce electricity using waste materials [53,54]. Therefore, the microbial fuel cells use low-priced and green energy sources. In one study on microbial fuel cell electrodes, the surface was modified with the appropriate catalysts to control the oxygen reduction rate [55]. The modified electrode surface also decreased the charge transfer resistance and enhanced the performance of the fuel cell [56]. In this fuel cell, membrane electrode assembly (MEA) was utilized. The application of nanocomposite material-derived catalysts in MEA may have formed proton and electron conduction pathways [57–59]. Important parameters of the electrode materials for microbial fuel cells which need to be considered include the specific surface area, current density, and power density. In this regard, it is considered crucial to alter the morphology of nanomaterials in order to improve the electrode performance. The type and amount of catalyst in MEA has been found to improve the electrode performance. The catalyst is usually coated on an electrode in MEA [60]. Conventional platinum/carbon (Pt/C) catalysts have also been coated with green materials such as poly(ethylene oxide), poly(vinyl alcohol), polyacrylic acid, etc., for the purpose of enhancing the ecological features of the electrodes [61]. In this regard, various green electrode and catalyst materials have been designed [62]. Ansari et al. [63] used TiO_2 nanocomposites to coat the carbon paper to form green electrode materials for microbial fuel cells. The green method was used with electrochemically active biofilm (EAB) to develop a nanocomposite cathode. The cathode had the capability to efficiently absorb visible light. Figure 1 shows the microbial fuel cell assembly, with the cathode and anode in two 300 mL bottles separated through a proton exchange membrane. The anode chamber contained a carbon paper electrode. This chamber was bubbled with nitrogen to achieve an anaerobic environment. The visible-light-active nanomaterials were formed using EAB at room temperature. The cathodes based on carbon paper and TiO_2-derived nanocomposites had power densities of 2.09 mW/m^2 to 4.34 mW/m^2. It has been observed that the microbial fuel cell with the EAB and TiO_2

nanocomposite-based cathode had a higher power density compared with that of the plain carbon paper electrode. The results were obtained due to the presence of an EAB-based electrode facilitating oxygen reduction reactions. Moreover, the enhanced performance was attributed to increased electrode catalytic activity. The as-synthesized green nanomaterials worked as electrodes in microbial fuel cell devices.

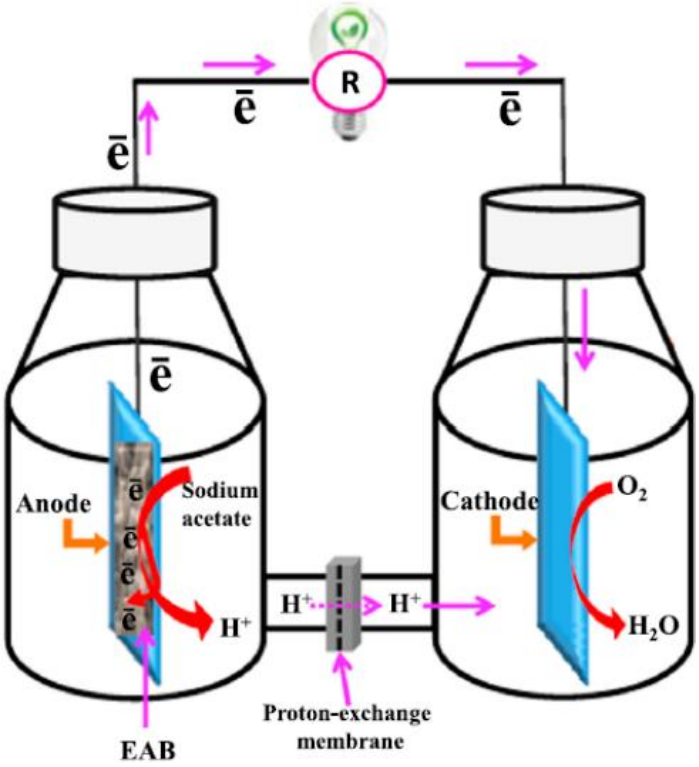

Figure 1. Schematic diagram of the microbial fuel cell [63]. Reproduced with permission from Elsevier.

Bacterial cellulose is a kind of nanocellulose prepared using bacteria. Bacterial cellulose is a low-cost, ecological, biocompatible, water-retaining, and high-strength material for designing fuel cell electrodes [64–66]. Bacterial cellulose has been amalgamated with polyaniline to form electrodes for microbial fuel cells [67]. Due to the fine electrical conductivity and chemical stability, the carbon-derived materials were mostly utilized as fuel cell anodes. The carbon fiber fabric used in the electrodes possessed a large specific surface area, high porosity and power density, and low resistivity to promote the fuel cell performance. Trindade et al. [68] designed a carbon fiber-embedded bacterial cellulose/polyaniline nanocomposite-derived microbial fuel cell anode using carbon paper as a cathode. During electrode formation, the carbon fiber was enfolded on bacterial cellulose to form a green network structure. Figure 2 expresses the bacterial cellulose- and carbon fiber-embedded sample and the scanning electron microscopy images. The bacterial cellulose sample was also dipped in aniline solution to develop a bacterial cellulose–polyaniline-based green nanocomposite electrode. The aniline monomer was adsorbed on the bacterial cellulose nanofiber surface and polymerized in situ in the presence of ammonium persulfate oxidant. The interconnecting bacterial cellulose–polyaniline scaffolds were developed with nanofibers uniformly coated with polyaniline. The conducting polymer coating over the bacterial cellulose nanofibers promoted the electrical conductivity properties of the green electrode. The microbial fuel cell with the carbon fiber/bacterial cellulose/polyaniline nanocomposite had a maximum current density of 0.009 mA/cm^2. Thus, green electrodes have been found to be beneficial for efficient microbial fuel cells.

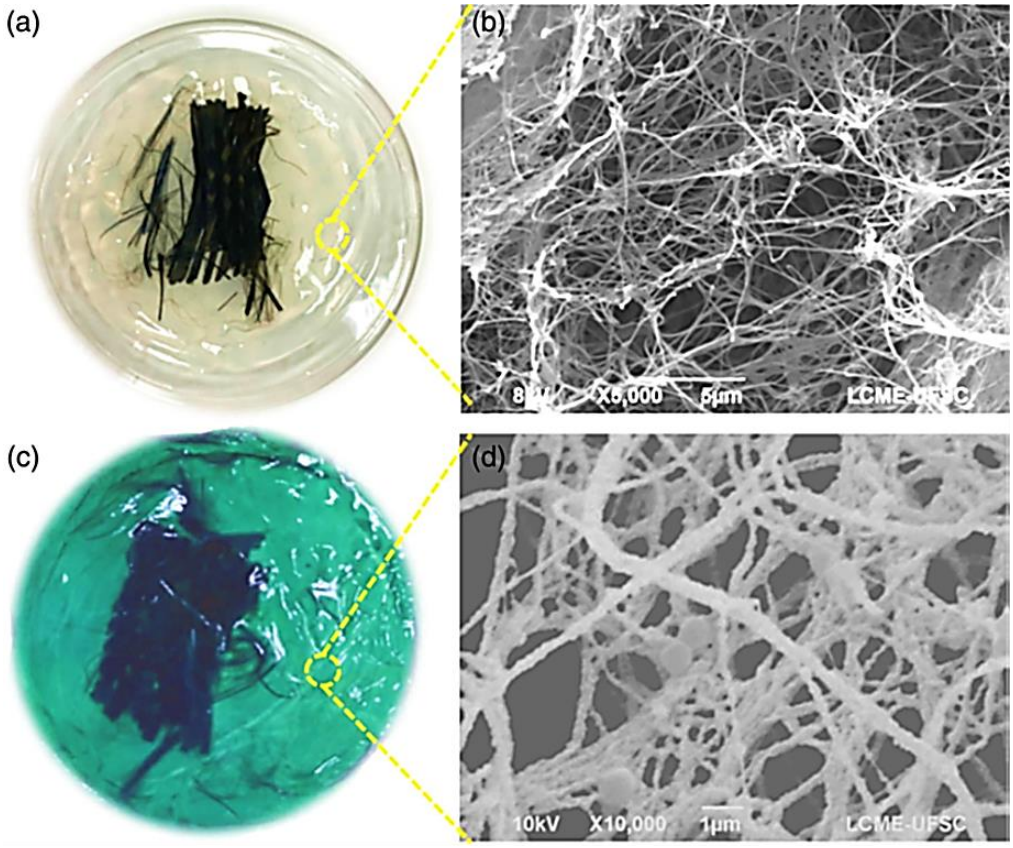

Figure 2. Morphological evaluation of bacterial cellulose sample and bacterial cellulose/polyaniline nanocomposite. (**a**) Carbon fiber embeds into bacterial cellulose hydrogel; (**b**) scanning electron microscopy micrograph of an interconnected network of bacterial cellulose nanofibers; (**c**) carbon fiber embeds into cellulose/polyaniline nanocomposite; and (**d**) scanning electron microscopy image of cellulose/polyaniline nanocomposite [68]. Reproduced with permission from Wiley.

In microbial fuel cells, metal-based anode materials may have corrosion problems, thus decreasing the bacterial growth and long-term firmness of electrodes [69]. Using carbon-based electrodes has the benefits of a high surface area and good conductivity properties [70]. However, the traditional carbon electrode materials possess performance limitations for microbial fuel cells [71]. In the case of carbon nanomaterials such as carbon nanotubes, a large surface area and a high level of electrical conductivity have been obtained; however, they may cause bacterial cellular toxicity and, thus, a decline in fuel cell performance [72]. More recent research has focused on graphene materials in fuel cell electrodes. Modified graphene, e.g., graphene oxide, has a high surface area and good strength properties, biocompatibility, and conductivity features [73–75]. However, graphene and its derivatives have the limitations of high costs and processing resources. In this regard, modified graphene oxide materials have been developed using green routes. Graphene oxide has been produced using agricultural waste, such as lignocellulosic material, which is low-cost and eco-friendly [76–78]. The green-derived graphene oxide possesses biocompatibility, a high surface area, and good conductivity properties for electrode synthesis. Moreover, green preparation techniques have been utilized to develop metal oxides such as zinc oxide and titania nanoparticles [79–81]. These nanomaterials have low costs, rescued toxicity, and superior electron transfer efficiency for utilization in fuel cell electrodes. Yaqoob et al. [82] fabricated the green ZnO and TiO_2 nanoparticles. The metal oxide nanoparticles were linked with green graphene oxide (GO) obtained from lignocellulose to form anodes for microbial fuel cells. Consequently, graphene oxide/zinc oxide (GO/ZnO) nanocomposite and graphene oxide/titania (GO/TiO_2) nanocomposite anodes have been fabricated. Alternatively, a graphite rod was utilized as a cathode electrode. Figure 3 depicts the electron

transport mechanism from bacteria to anode in a microbial fuel cell. The biofilm may transfer electrons from bacteria to anode. The electrochemical impedance spectroscopy (EIS) Nyquist plots of graphene oxide, GO/ZnO, and GO/TiO_2 nanocomposite anodes were studied. Amalgamations of graphene oxide/metal oxide nanocomposite-derived anodes offer high energy efficiency. In the microbial fuel cell, the GO/ZnO and GO/TiO_2 anodes had power densities and current densities of 912 $\times$ 10^{-3} mW/m^2 and 608 $\times$ 10^{-3} mW/m^2, respectively, higher than neat graphene oxide (292 $\times$ 10^{-3} mW/m^2). The current density of the nanocomposite anodes was also found to be considerably higher than that of the graphene oxide electrode. Hence, the green design's electrochemical and power density performance with the nanocomposite anodes was found to be useful for microbial fuel cells.

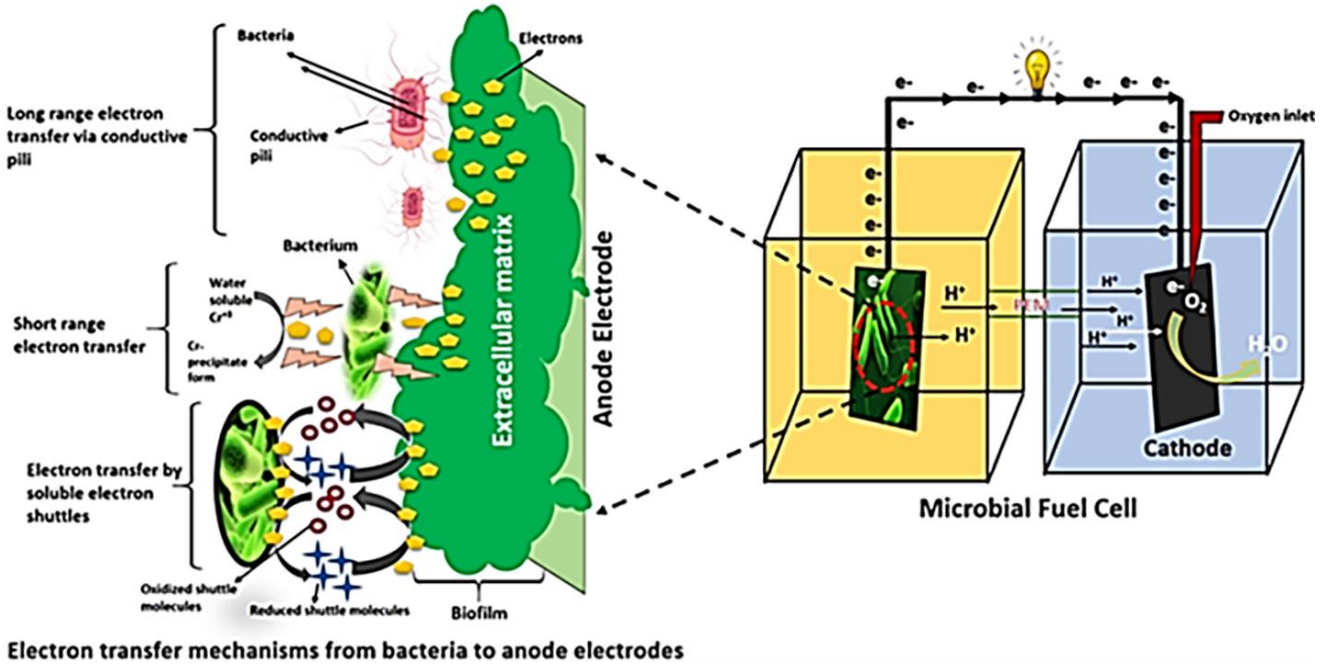

Figure 3. Mechanism of electron transportation from bacteria to anode in the microbial fuel cell [82]. Reproduced with permission from Elsevier.

Mashkour et al. [83] produced graphite paste, graphite paste–titanium dioxide, and hybrid graphene cathodes using green and facile approaches for microbial fuel cells. As compared with the neat graphite paste, graphite paste–titanium dioxide showed higher catalytic activity than the graphite paste and graphite paste–titanium dioxide cathodes. The graphite paste–hybrid graphene electrode revealed further enhanced catalytic activity due to the inclusion of graphene's unique nanostructure. Similarly, the power density of the graphite paste–hybrid graphene electrode was found to be higher (220 mW/m^2) relative to that of graphite paste–titanium dioxide (80 mW/m^2) and neat graphite paste (30 mW/m^2) (Figure 4). The green electrodes formed using the environmentally friendly approach were used to replace Pt in microbial fuel cells.

Moreover, recyclable carbon nanostructure-derived electrodes have been used to form green fuel cells [84]. Gouda et al. [85] utilized waste bottles of polyethylene terephthalate to prepare reduced graphene oxide and reduced graphene oxide/magnetic iron oxide nanocomposite electrodes. The reduced graphene oxide acted as the anode, whereas the graphene/magnetic iron oxide nanocomposite was applied as the cathode. For electrode fabrication, reduced graphene oxide/magnetic iron oxide was mixed with green polymers such as poly(vinyl alcohol), polyethylene oxide, and polyvinyl pyrrolidone. The resulting electrode showed a current density around 0.98 mA cm^{-2}. Moreover, green electrodes have high porosity and electrocatalytic activity, as determined by three-electrode cell

electrochemical measurements and linear scan voltammetry. Table 1 illustrates important specifications of green nanomaterial-based microbial fuel cell electrodes.

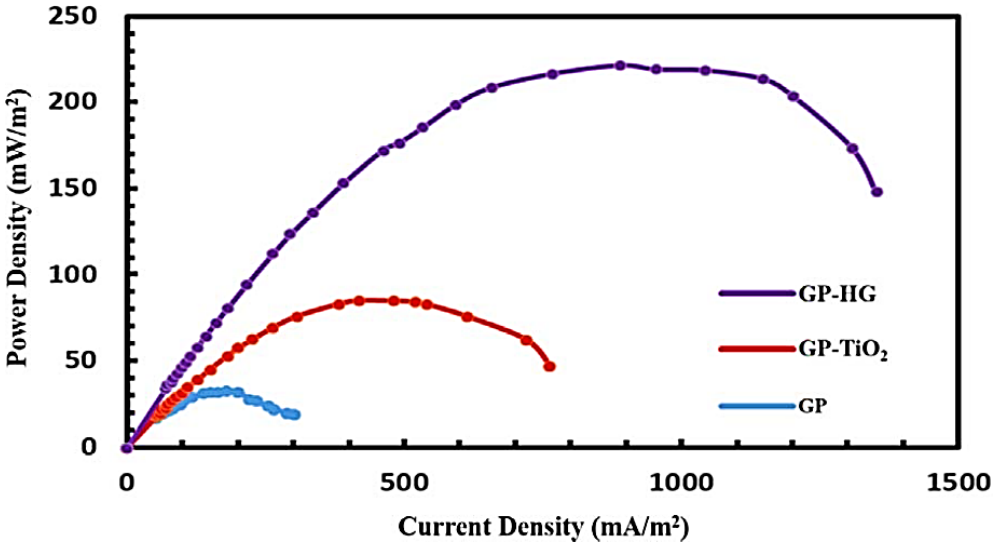

Figure 4. Power output curves (batch mode) of graphite paste (GP), graphite paste–titanium dioxide (GP-TiO_2), or graphite paste–hybrid graphene (GP-HG) cathodes [83]. Reproduced with permission from Elsevier.

Table 1. Specifications of green nanocomposites for microbial fuel cell electrodes.

Green Material	Fabrication	Parameters/Characteristics of Electrodes	Ref
Bacterial cellulose/polyaniline	Amalgamation	Microbial fuel cell electrode	[67]
Carbon fiber-embedded bacterial cellulose/polyaniline nanocomposite	Carbon fiber embedded into bacterial cellulose hydrogel using culturing period	High current density of 0.009 mA/cm^2, relative to neat carbon fiber electrode (0.009 mA/cm^2)	[68]
Lignocellulosic-derived green graphene oxide with TiO_2 and ZnO	Carbonization; solvothermal methods	Power density of 912 × 10^{-3} mW/m^2, relative to neat graphene oxide (292 × 10^{-3} mW/m^2); Surface area 40.1657–63.1991 m^2/g	[82]
Graphite paste–hybrid titanium dioxide	Green and facile approaches such as Hummer's method, paraffin oil, glass tubes	Power density of 220 mW/m^2, relative to neat graphite paste (30 mW/m^2); Surface area 0.125 cm^2	[83]
Polyethylene terephthalate waste bottle-derived reduced graphene oxide and reduced graphene oxide/magnetic iron oxide nanocomposite	Inverse co-precipitation	Power densities of 395 mWcm^{-2}; current density of 0.98 mA cm^{-2}	[85]
Amidoxime-modified bacterial cellulose/carboxylated multi-walled carbon nanotube	Vacuum filtration	Power density of 1.897 W m^{-3}, relative to non-modified electrode (0.813 W m^{-3})	[86]
Bacterial cellulose doped with P and Cu	Food waste fermentation stillage	Maximum output power 572.16 mW·m^{-2}	[87]

3. Green Nanocomposites in Solid Electrolyte Membranes of Fuel Cells

Electrolytes are considered as important components of different categories of fuel cells, including PEMFC, alkaline fuel cell, phosphoric acid fuel cell, molten carbonate fuel

cell, and other devices [88–90]. Modern fuel cell technology has focused on using ecological materials with less fuel ingestion and no toxic emissions [91]. Various non-toxic, biodegradable, and sustainable bio-composite materials have been used in commercial-scale fuel cells [92–94]. Solid electrolyte membranes based on sustainable green materials have broadened the scope of fuel cell devices [95]. For solid electrolytes in fuel cells, important parameters to consider include proton conductivity, ion exchange capacity, proton diffusion coefficient, permeability, power output, power density, etc. Vijayalekshmi et al. [96] designed chitosan-based green ion transportation membranes for PEMFC. Prepared membranes were doped with methanesulfonic acid and sodium salts of dodecylbenzene sulfonic acid to facilitate ion conduction through the inter-linked membrane system. The inclusion of 15 wt.% of dopant to green membrane resulted in a proton conductivity of 2.86–4.67 $\times$ 10^{-4} Scm^{-1} at 100 °C. The membranes have sufficiently high efficiency, low cost, and ecological friendliness to replace the commercial Nafion membranes, and the doped chitosan membranes had advanced mechanical properties compared to the Nafion membranes. Consequently, the doped chitosan membranes had higher tensile strength (38 MPa) compared to standard Nafion membrane (24 MPa). Furthermore, the doped membranes possessed thermal stability up to 260 °C. Accordingly, chitosan doping offered an effective way to enhance the heat resistance, mechanical features, and proton conductivity as needed for developing efficient commercial solid electrolyte membranes for fuel cells. In addition, the inexpensiveness and environmental friendliness of chitosan membranes render them more advantageous than the commercial Nafion in PEMFC. Brodt et al. [97] produced polyacrylic acid nanofibrous membranes for MEA assembly. The uniform morphology of the membranes was suggested to be useful for enhanced MEA-based fuel cell performance [98].

Mohanapriya et al. [99] fabricated green electrolyte membranes using Pectin (PC), poly(vinyl alcohol) (PVA), and sulfonated titanium dioxide (s-TiO_2) for direct methanol fuel cells. Pectin-poly(vinyl alcohol) (PC-PVA) and pectin-poly(vinyl alcohol)/sulfonated titanium dioxide (PC-PVA/s-TiO_2) nanocomposite membranes were developed. The in situ cross-linked PC-PVA and PC-PVA/s-TiO_2 membranes were casted using the solution method. Finely dispersed s-TiO_2 nanoparticles formed an inter-linked network with a low free-void volume. Moreover, the interactions between the PC-PVA matrix and s-TiO_2 nanoparticles enhanced the permeation, selectivity, and transport rate through the membrane. The s-TiO_2 nanoparticles acted as inorganic proton-conducting materials. The inclusion of nanoparticles enhanced the ion conductivity and prevented methanol permeation by the membranes. Figure 5 shows the scheme for the proton transfer over methanol molecules in the PC-PVA/s-TiO_2 nanocomposite membrane.

Figure 6 presents the PC-PVA blend and PC-PVA/s-TiO_2 membranes as well as their methanol permeability. The addition of s-TiO_2 nanoparticles decreased the methanol permeability by 40%. The decline in the methanol permeation was attributed to the presence of proton-conducting channels and varying water retention properties. The electrochemical selectivity of membranes for protons vs. methanol can be defined as the ratio of proton conductivity to methanol permeability, and can be utilized for evaluation of the membrane–electrolyte performance. It can influence both proton conductance and methanol diffusion features. Consequently, the electrochemical selectivity of the PC-PVA blend and PC-PVA/s-TiO_2 membranes was analyzed. Owing to low methanol cross-over and high proton conductivity, high membrane selectivity was attained. Figure 7 shows the variation in cell voltage and power density with the current density. The direct methanol fuel cell showed a high power density (27 mW/cm^2) at 70 °C. Thus, the designed electrolyte membrane can offer low-cost, eco-friendly, and efficient solutions for commercial-level green fuel cell technology.

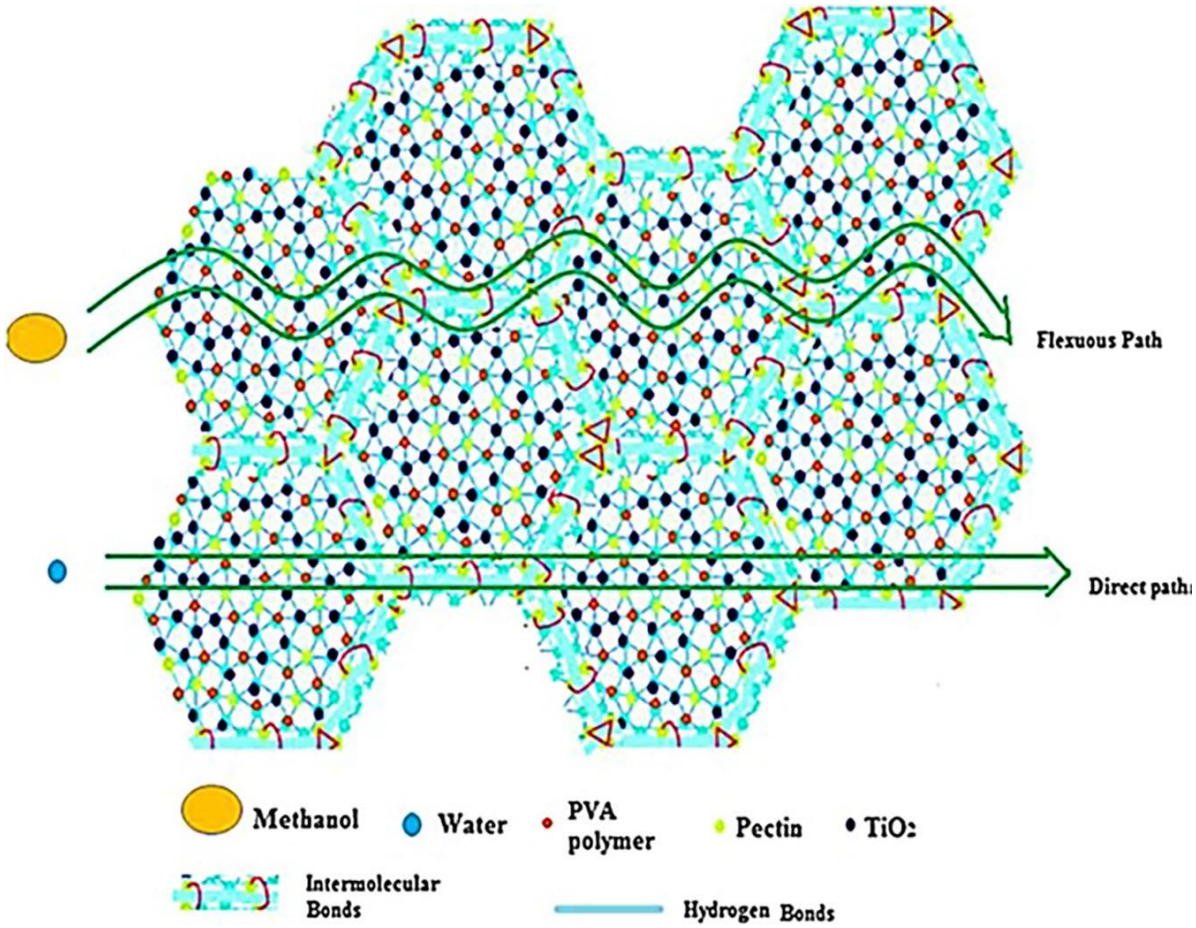

Figure 5. Scheme of facile proton transportation and constrained methanol transportation in PC-PVA/s-TiO_2 hybrid membranes [99]. PC-PVA/s-TiO_2 = pectin-poly(vinyl alcohol)/sulfonated titanium dioxide. PVA = poly(vinyl alcohol). Reproduced with permission from Elsevier.

Direct borohydride fuel cells use solid electrolyte membranes for their low cost and low operational temperature [100–102]. This green fuel cell approach utilizes hydrogen peroxide, oxygen, and sodium borohydride [103]. Direct borohydride fuel cells may produce excessive electrons due to oxidation of borohydride BH^{4-} anions and sodium Na^{+} cations during the passage through the membrane [104,105]. The efficient membranes may have fine anion transport and cation exchange features [106]. Initially, Nafion membranes were employed in direct borohydride fuel cells [107]. However, these membranes may have the drawbacks of expensiveness, intricate processing, and perfluorinated structure. Nowadays, it is being attempted to replace electrolytes in direct borohydride fuel cells with economical and green nanomaterial-derived membranes. Gouda et al. [108] produced sulfonated poly(vinyl alcohol) (SPVA) and SO_4 and PO_4 doped titania nanotubes ($SPTiO_2$) for polymer proton exchange membranes. The poly(vinyl alcohol) matrix was converted to SPVA with 4-sulfophthalic acid as the sulfonating agent and crosslinker. The electrolyte membranes were used in direct borohydride fuel cells. Spectroscopic, morphological, and thermal analyses of the membranes have been carried out in addition to specific fuel cell studies. The water uptake and swelling degrees of SPVA- and $SPTiO_2$-derived membranes were observed to be 13% and 7%, respectively. Table 2 demonstrates the oxidative stability, ion exchange capacity, borohydride permeability, and ionic conductivity properties. The ion exchange capacity of the nanocomposite membranes showed an upsurge around 0.10–0.50 $meqg^{-1}$ due to increasing doping agent concentrations of 0–3 wt.%. Furthermore, the ionic conductivity of the nanocomposite membranes was augmented by about 1.2–7.1 $mScm^{-1}$. The noteworthy upsurge in ionic conductivity with increasing doping

agent content occurred because of a decrease in resistivity. The borohydride permeability of the doped nanomaterial was decreased to 0.32×10^{-6} cm^2 s^{-1} from 0.71×10^{-5} cm^2 s^{-1} (undoped membrane). The borohydride permeability was inferior to the that of the commercial Nafion®117 membrane, i.e., 0.40×10^{-6} cm^2 s^{-1}. The obtained outcomes of the analyses of green nanocomposite membranes provide a path towards the development of inexpensive, ecological, and efficient direct borohydride fuel cells.

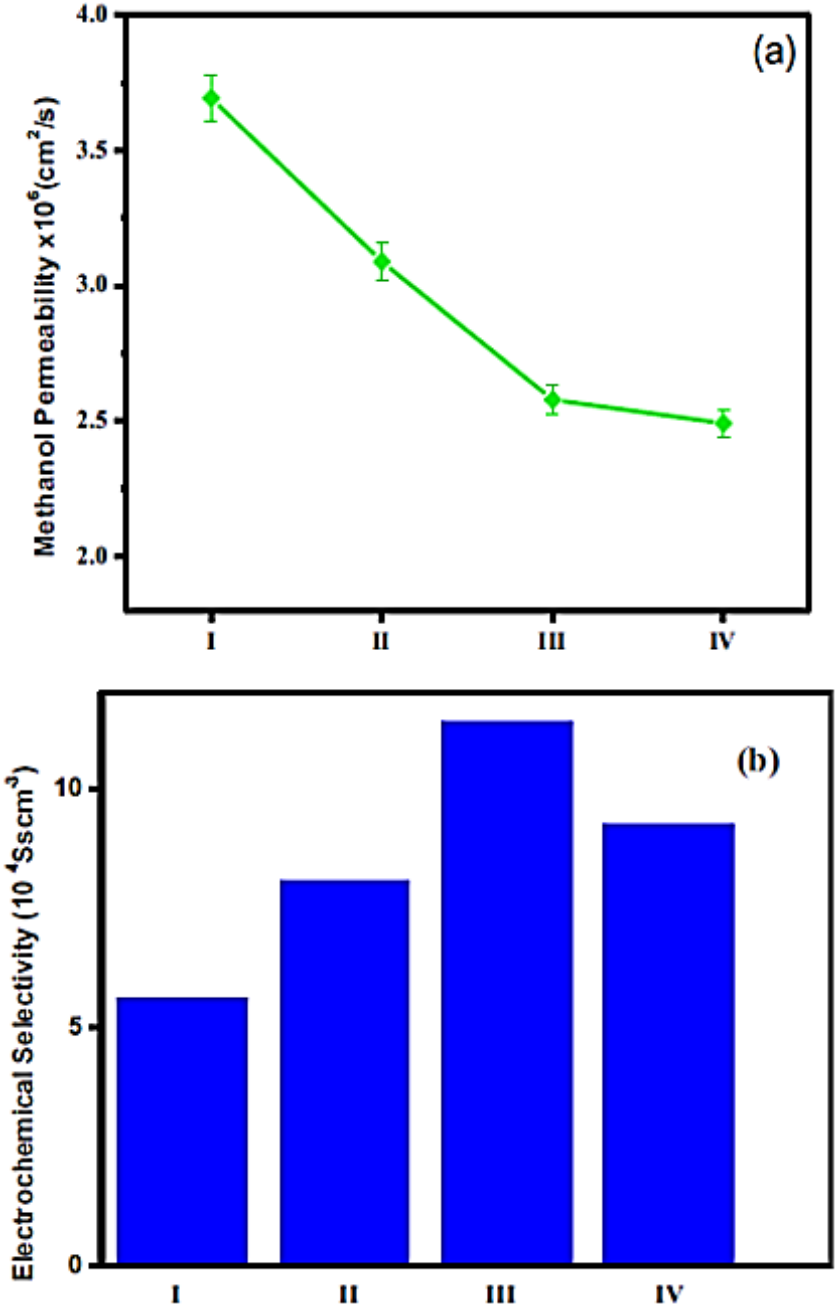

Figure 6. (**a**) Methanol permeability of PC-PVA blend, PC-PVA/s-TiO_2-I, PC-PVA/s-TiO_2-II, and PC-PVA/s-TiO_2-III hybrid nanocomposite membranes measured at 70 °C; and (**b**) electrochemical selectivity of PC-PVA blend, PC-PVA/s-TiO_2-I, PC-PVA/s-TiO_2-II, and PC-PVA/s-TiO_2-III hybrid nanocomposite membranes [99]. PC-PVA = pectin-poly(vinyl alcohol); PC-PVA/s-TiO_2 = pectin-poly(vinyl alcohol)/sulfonated titanium dioxide. Reproduced with permission from Elsevier.

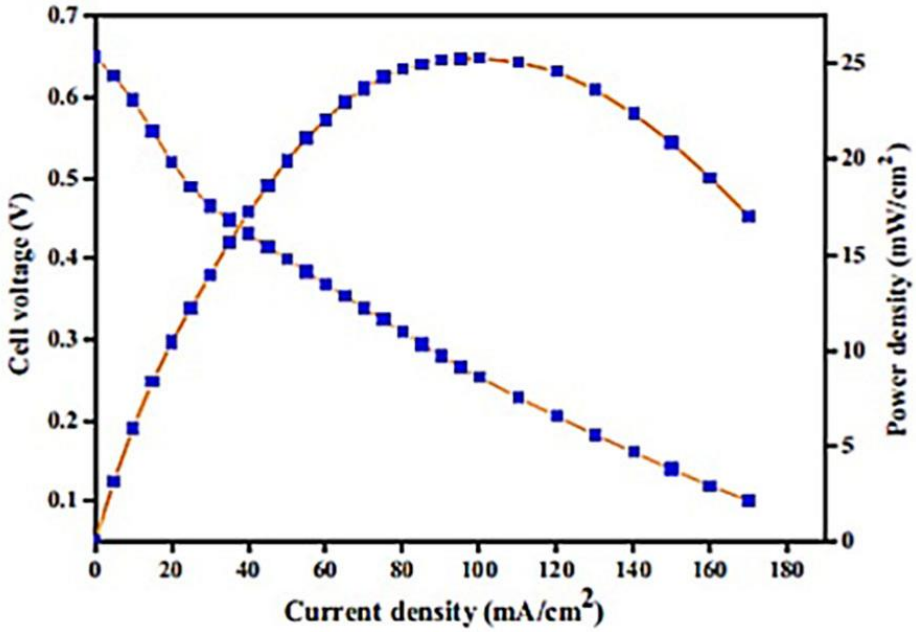

Figure 7. Cell voltage and power density vs. current density for PC-PVA/s-TiO_2-II hybrid membrane [99]. PC-PVA/s-TiO_2 = pectin-poly(vinyl alcohol)/sulfonated titanium dioxide. Reproduced with permission from Elsevier.

Table 2. Properties of fabricated fuel cell membranes in comparison with Nafion®117 [108]. SPVA = sulfonated poly(vinyl alcohol); SPTiO_2 = SO_4- and PO_4-doped titania nanotubes. Reproduced with permission from MDPI.

Membrane	Oxidative Stability	IEC (meq g^{-1})	Borohydride Permeability (cm^2 s^{-1})	Ionic Conductivity (mS cm^{-1})
SPVA	80	0.10	0.71×10^{-5}	1.25
SPVA-SPTiO-1	90	0.25	0.49×10^{-6}	3.12
SPVA-SPTiO-2	99.5	0.40	0.39×10^{-6}	5.57
SPVA-SPTiO-3	98	0.50	0.32×10^{-6}	7.13
Nafion®117 [109]	92	0.89	0.40×10^{-6}	45.0

Biopolymer composites have been used to form ion exchange fuel cell membranes [110]. Biopolymers obtained from biological sources, in combination with nanomaterials, may enhance the efficiency of PEMFC [111]. Polyhydroxyalkanoates are important microbial polyesters synthesized by species of bacteria [112]. Sirajudeen et al. [113] formed membranes based on polyhydroxyalkanoates with medium chain lengths for microbial fuel cells. A polyhydroxyalkanoate composite was prepared with poly-(R)-3-hydroxybutyrate, and had biocompatibility and biodegradability properties. Relative to Nafion, the potential of the proton exchange membrane was analyzed. The polyhydroxyalkanoate-based electrolyte had higher power density (601 mW/m^2) than that of the Nafion (520 mW/m^2) membrane. Yusuf et al. [114] designed a proton exchange membrane based on medium-chain-length poly-3-hydroxyalkanoate and carboxyl functional multi-walled carbon nanotube-based nanocomposites for the microbial fuel cell. The nanocomposite membranes were formed with 5%, 10%, and 20% *w/w* nanofiller. The poly-3-hydroxyalkanoate/multi-walled carbon nanotube-based nanocomposite membrane revealed a higher power density of 361 mW/m^2 compared to Nafion 117 (372 mW/m^2). The inclusion of a nanofiller in the matrix improved the water uptake and interface surface area. Moreover, the environmentally friendly membranes showed higher coulombic efficiency and proton conductivity, and lesser in-

ternal resistance, than commercial Nafion 117. Thus, green polyhydroxyalkanoate-based membranes had enhanced fuel cells relative to commercial membranes.

Nanoclay mixed green separators have also been reported for microbial fuel cells [115]. Hasani-Sadrabadi et al. [116] prepared poly(ether ether ketone)- and montmorillonite-derived membranes for microbial fuel cells. The inclusion of 3 wt.% montmorillonite nanoclay enhanced the power output by 40% relative to the Nafion 117-based fuel cell. Moreover, a higher open circuit voltage of the nanoclay-based membrane was observed relative to the Nafion 117. Thus, green nanoclay-based membranes acted as talented electrolytes to improve the performance of microbial fuel cells [117]. Table 3 demonstrates the characteristics of green solid electrolyte membranes.

Table 3. Characteristics of green solid electrolyte membranes.

Green Solid Electrolyte Membrane	Fabrication	Features	Ref
Chitosan doped with methanesulfonic acid and sodium salts of dodecylbenzene sulfonic acid	Chitosan acetate solution method	15 wt.% dopant proton conductivity $2.86–4.67 \times 10^{-4}$ Scm^{-1}; Activation energy 5.45 KJ/mol, i.e., comparable to Nafion	[96]
Polyacrylic acid nanofibrous membrane f	Electrospinning	Uniform morphology; proton conductivity higher than Nafion	[97]
Poly(vinyl alcohol)/sulfonated titanium dioxide and pectin-poly(vinyl alcohol)/sulfonated titanium dioxide	Solution casting method	Fine nanofiller dispersion; ion exchange membrane 0.68 meq./g; proton diffusion coefficient 2.7×10^{7} cm^2/s higher than Nafion	[99]
Sulfonated poly(vinyl alcohol) with SO_4 and PO_4 doped titania nanotube; 4-sulfophthalic acid as ionic crosslinker	Solution method	Ion exchange capacity 0.10–0.50 meq./g^{-1} with enhancing doping agent concentrations; ionic conductivity 1.2–7.1 $mScm^{-1}$; decrease in borohydride permeability from 0.71×10^{-5} cm^2 s^{-1} to 0.32×10^{-6} cm^2 s^{-1}; borohydride permeability lower than commercial Nafion®117 (0.40×10^{-6} cm^2 s^{-1})	[108]
Polyhydroxyalkanoates; poly-(R)-3-hydroxybutyrate	Solution route	Biocompatible; biodegradable; power density (601 mW/m^2) higher than Nafion (520 mW/m^2)	[113]
Poly-3-hydroxyalkanoate/carboxyl functional multi-walled carbon nanotube	Solution route	Higher power density of 361 mW/m^2 compared with Nafion 117 (372 mW/m^2)	[114]
Poly(ether ether ketone)/montmorillonite based nanocomposite	Solution route	40% higher power output relative to Nafion 117 based fuel cell	[116]

4. Significance and Challenges of Using Green Nanocomposites in Microbial Fuel Cell

Owing to the current ecological demands for energy devices which produce less pollution, various modified materials and strategies have been adopted for fuel cell applications. Microbial fuel cells were concentrated for the purpose of employing biological resources [118]. The overall fuel cell performance depended upon the electrode (cathode and anode) material used [119]. Various green materials, such as bacterial cellulose, green graphene oxide derived from lignocellulose, and green-derived ZnO and TiO_2 nanoparticles, were employed to form fuel cell electrodes. Moreover, green synthesis routes were applied to develop fuel cell electrodes. However, little research has been conducted regarding microbial fuel cells [120]. Although several green electrode materials have been

devised for microbial fuel cells and MEA to enhance the power performance, future devoted research attempts are still needed [121]. Durability, compatibility, and enhanced power output were especially in demand upon interaction with the microorganisms [122]. The electrode interfaces also need to be modified in order to enhance the connections with the microorganisms. Green electrode materials obtained using green routes have been a research focus to achieve durability, high power output, and rapid start-up time [123].

Figure 8 shows the needs of energy devices and the advantages of fuel cells. Microbial fuel cells must be enhanced to compensate for their inexpensiveness, sustainability, and eco-friendliness [124–126]. Moreover, the use of non-toxic materials for electrodes and catalysts, low pollution emissions, room temperature functioning, and optimum operational conditions must be also be topics of focus [127]. The durability and design flexibility must also be considered for efficient microbial fuel cells [128]. The production of green energy by microbial fuel cells depends upon various factors, such as the conveyance of electrons and protons through the green design and fabrication of electrodes and membranes; the biocatalytic reaction of microorganisms that produces protons and electrons; and the reduction in oxygen through electrons for safe energy [129]. Carbon cloth, carbon fiber, carbon paper, and graphite rod-based electrode materials need to be further modified to enhance the processability, surface area, and electron conductivity. In this regard, granular carbon and granular graphite materials were applied to develop fuel cell electrodes [130–132]. To enhance the surface area, graphite or carbon structures must be cut down into fine small segments. In this way, brush-like anode materials have been produced (Figure 9). Such advancements have proved effective in enhancing the surface area of anode materials to develop the fiber brush anode. Moreover, eco-friendly materials have been developed using green routes to form proton transportation membranes for PEMFC. Hence, effective fuel cell performance of electrodes and proton exchange membranes depends on green material design, green synthesis route, and safe interactions with microorganisms [133,134].

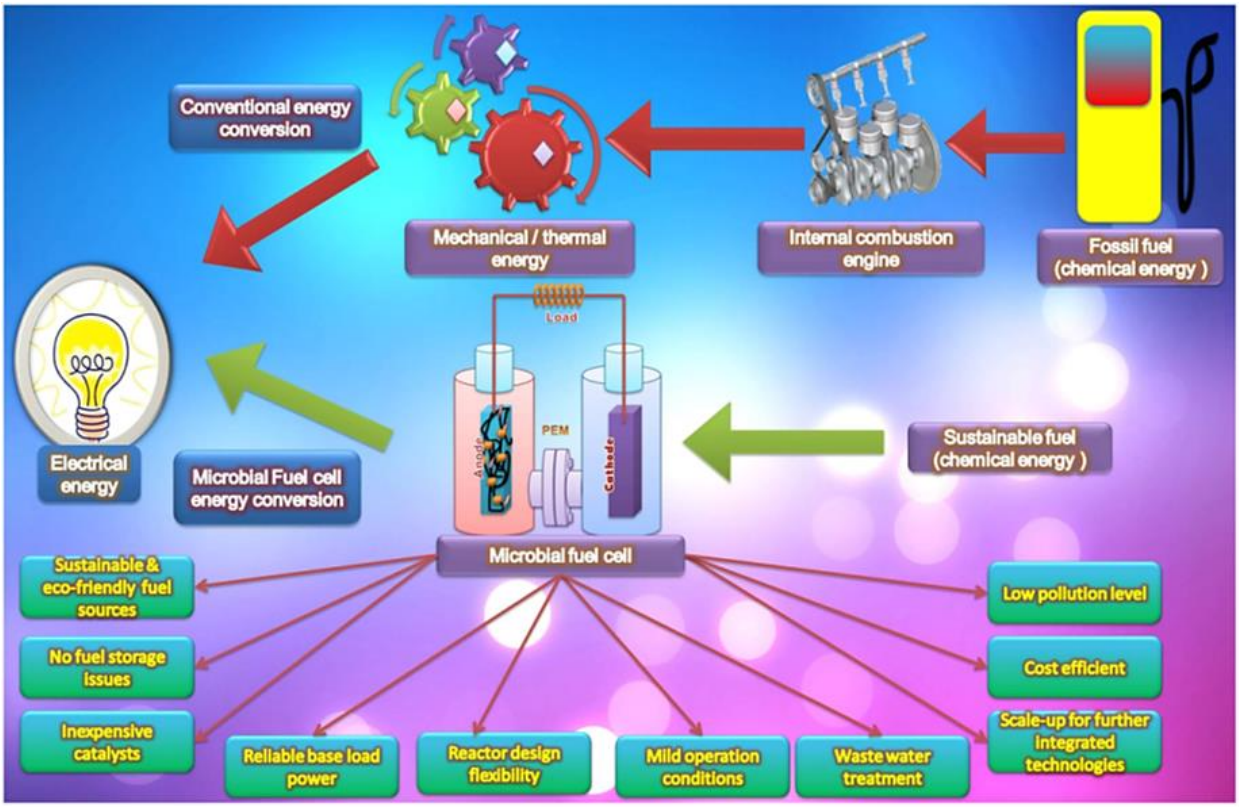

Figure 8. Energy diagram of energy devices and benefits of microbial fuel cells [135]. Reproduced with permission from Elsevier.

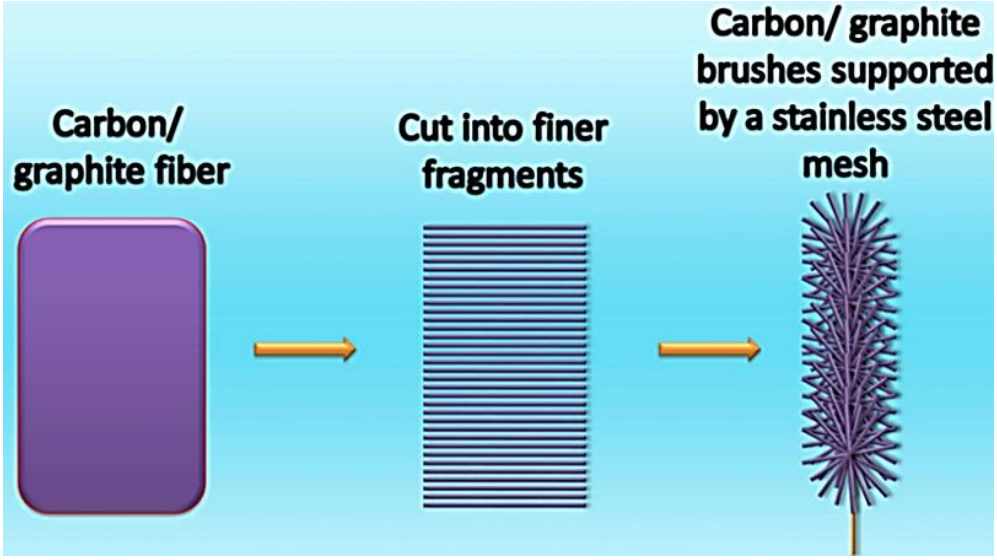

Figure 9. Mechanism of increasing graphite electrode surface area [135]. Reproduced with permission from Elsevier.

5. Conclusions

In short, this review article provides a transitory benchmark for the use of green and sustainable nanomaterials and green approaches for the electrodes and electrolytes of fuel cells. In particular, using green or green-derived nanocomposites in the electrodes and electrolytes of microbial fuel cells has been observed to be a rapidly rising research area. The structural, morphological, and physical properties of green nanocomposites have been determined for fuel cell application. Consequently, their microstructure, electrical conductivity, electrochemical properties, mechanical stability, thermal constancy, power density, current density, resistance, and fuel cell efficiency have been researched. The obtained results from reports in the literature have intensified the study of nanocomposites in the electrodes of microbial fuel cells. Using innovative nanomaterials revealed innovative perspectives for the use of high-performance advanced fuel cells. The forthcoming progress in green or green-synthesized nanocomposites for electrodes and electrolytes depends on new and environmentally friendly design possibilities and green fabrication courses to attain the optimum processing parameters of the ecological fuel cells.

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