Chapter 9

Membrane Technologies for Sustainable and Eco-Friendly Microbial Energy Production

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Contents

Abstract Environmental deterioration and energy crisis caused by ever-increasing exploitation of traditional fossil fuels are urgent problems that need to be addressed. Microbial energy conversion technologies have attracted wide attentions since they can convert chemical energy contained in wastes, like solid wastes and wastewater, into biofuels or bioelectricity, realizing environmental remediation and energy

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production at the same time. But the conventional methods have many limitations, like low mass transfer rate, uneven energy distribution, and strong product or by-product inhibition. The introduction of membranes in the reaction system can effectively relieve these technical bottlenecks by regulating the transfer and distribution properties of mass, heat, and energy, which play important roles on bioenergy productivity and quality.

We review (1) membrane application on liquid biofuels production, mainly on biomass cultivation and harvesting, liquid biofuels generation, and liquid products refining; (2) membrane application on gaseous biofuels production, mainly on photo-dependent biohydrogen production, dark-fermentative biohydrogen production, and gaseous products purification; (3) membrane application on microbial fuel cell; (4) membrane biofouling; and (5) antibiofouling technologies. The membranes mainly act as physical barrier, internal bridge, inhibitors separator, or products extractor in microbial energy production processes, which varies according to the detailed occasions. In overall, the membrane can effectively enhance microbial energy productivity and quality. But biofouling is the vital problem for all cases. Further researches and development on antifouling of membranes are still necessary.

Keywords Microbial biofuels · Membrane · Bioethanol · Biolipids · Microbial fuel cell · Biohydrogen · Bioreactor · Biofouling · Fermentation · Recovery

9.1 Introduction

Currently, traditional fossil fuels like coal, natural gas, and petroleum are still predominant fuel types for human beings. But limited reservoir, depleting supply, and random consumption hinder the dependency on traditional fossil fuels as major energy sources (Chang et al. 2018). In addition, vast utilization of fossil fuels has caused many problems, such as global warming, energy crisis, and environmental destruction (Fu et al. 2018; Guo et al. 2018; Tian et al. 2010). There are pressing needs to develop renewable and environmental-friendly energy sources which are derived from non-fossil sources in ways that can be replenished (Chang et al. 2018). Renewable energy mainly includes solar, wind, hydro, geothermal, and biofuels. Among these different renewable energy types, the biofuels produced via microbial energy conversion are considered as one of the most promising energy types due to its high energy conversion efficiency, mild operating conditions, and environmental remediation ability (Chang et al. 2016a; Li et al. 2017; Liao et al. 2014; Lu et al. 2018).

A variety of materials can be used as feedstocks for biofuels production, and based on that, the biofuels production can be mainly classified into first-, second-, and third-generation biofuels (Nigam and Singh 2011), as shown in Table 9.1. The first-generation biofuels are mainly generated from oil crops or starch-based food

Table 9.1 Various generations of biofuel (Correa et al. 2017; Leong et al. 2018; Nigam and Singh 2011; Kumari and Singh 2018)

Biofuels generations	Feedstocks	Advantages and disadvantages
The first generation	Soybean, sunflower, sugarcane, corn, etc.	Advantages:
		Simple pretreatment process, pure products, and high conversion rate of feedstocks
		Disadvantages:
		Food and freshwater competition with human beings, low economic efficiency
The second generation	Agricultural and forestry residues, like wheat and maize crops, sawdust, and sugarcane bagasse	Advantages:
		Abundant feedstocks, without competition with human beings for arable land, waste utilization
		Disadvantages:
		Sophisticated pretreatment process, low conversion rate, high energy cost, impure products
The third generation	Biofuels or electricity generation with microorganisms, like microalgae and microbes	Advantages:
		High conversion rate, less by-products, high products quality
		Disadvantages:
		High economy investment

crops. For example, the oleaginous crops including soybean and sunflower can be used as feedstocks for biolipid extraction through transesterification, and the starch-containing grains like corn, sorghum, and sugarcane are used as substrates for bioethanol and biohydrogen production through fermentation for the first-generation biofuels. The advantages of the first-generation biofuels are relatively simple pretreatment technologies since the starch and fats contained in food crops have simpler structure which are easier to be decomposed than lignocellulose. But the competition of arable land and freshwater for biofuels production with human beings' food demand strongly restricted its application (Correa et al. 2017). The second-generation biofuel fulfills the impractical gap of the first-generation biofuel due to its utilization of nonedible substrates from forestry and agricultural lignocellulose, like wheat and maize crops, sawdust, and sugarcane bagasse (Tian et al. 2009). Through hydrolysis and fermentation of this lignocellulosic biomass, biofuels like bioethanol and biohydrogen are produced in forms which can be utilized as energy sources. However, due to the tightly connected structure of lignin–cellulose association and crystalline structure of cellulose which resist enzymatic hydrolysis, sophisticated processes are necessary to achieve potential biofuels outcome, greatly increasing the energy cost of the second-generation biofuels (Kumari and Singh 2018; Raman et al. 2015). The third-generation biofuels which are derived from microorganisms, like microalgae and microbes, are considered as promising alternative energy sources since they can avoid the major disadvantages of food competition for the first-generation biofuels and non-degradability for the second-generation liquid biofuels (Zhu et al. 2018). Many microorganism species have abilities to accumulate fatty acids in the cells, like microalgae, yeast, and fungi (Leong et al. 2018; Liao et al. 2014; Mathimani and Pugazhendhi 2019). The intracellular fatty acids can be used as substrates for biodiesel production through downstream processing of the microbial biomass.

Biofuels production mainly experiences three steps: feedstocks pretreatment, biofuels generation, and biofuels refining. Until now, the biofuels productivity and quality are still poor attributing to many technical limitations despite the feedstock materials. The limitations are mainly confined to low pretreatment efficiency of the feedstock, poor biomass to biofuels conversion efficiency, and hardness on products separation and purification (Rodionova et al. 2017). Environmental conditions like temperature, humidity, and pH; operating parameters like material proportion, retention time, and inoculum density; and some other intrinsic properties like material composition, yeast activity, and bioreactor structure have important roles on biofuels productivity and quality (Srivastava et al. 2018; Liao et al. 2015; Pei et al. 2017).

During biofuels production processes, transfer characteristic of mass, heat, and energy determines its distribution in the system, which ultimately affects direction and rate of the chemical reactions, like lignocellulose hydrolysis to produce sugars and sugar fermentation to produce bioethanol or biohydrogen. Therefore, regulations on mass, heat, and energy transfer and distribution can greatly improve effectiveness of biomass to biofuels conversion. But conventional methods paid few attentions on transfer regulation attributing to rough system structure, resulting in low biofuels productivity and poor quality. The introduction of membrane modules in microbial energy conversion system can significantly reduce the technological limitations by acting as physical barrier, internal bridge, inhibitors separator, or products extractor. The functions of membrane vary with its utilizing occasions. Major applications of membranes on microbial energy production processes, i.e., liquid biofuels, gaseous biofuels, and microbial fuel cell, are illustrated in Fig. 9.1 and discussed in the following parts in detail.

9.2 Membrane Application on Liquid Biofuels Production

Liquid biofuels, like biolipids and bioethanol, are favored types of biofuels since they can blend with petroleum for combustion, realizing partly replacement of fossil energy by eco-friendly ways without sacrificing power output. In particular, the bioethanol has gained wide attentions since it satisfies the necessities of clean technology, like sustainability, biodegradability, abundant substrate, and reduction in greenhouse gas emissions, and is suitable to be used in most diesel engines with little or no modification (Enagi et al. 2018). In many countries, vehicles using bioethanol and gasoline mixture for transportation have been successfully realized, reducing greenhouse gas emissions to a large extent ranging from 20% to 85% (Wei

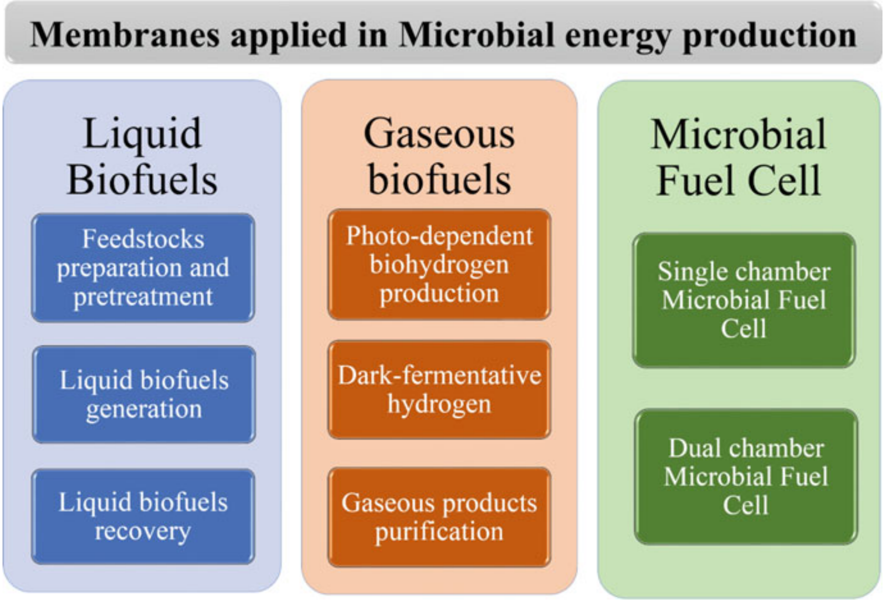


Fig. 9.1 Major application of membranes on microbial energy production processes

et al. 2014). Therefore, developing liquid biofuel technologies are promising approaches for environmental and energy sustainability in the present.

The process of liquid biofuels production mainly includes feedstocks preparation like microalgae cultivation and harvesting, liquid biofuels generation like fermentation and the related processes, and products refining like bioethanol and biodiesel recovery (Carrillo-Nieves et al. 2019). Among these steps, membrane can play an important role on enhancement of liquid biofuels productivity over the traditional technologies. Major applications of membranes in liquid biofuels production process and its advantages are shown in Table. 9.2.

9.2.1 Membranes Used for Microalgae Cultivation and Harvesting

Abundant biodegradable feedstocks are prerequisites for economically feasible liquid biofuels production. Among different materials like corn, sugarcane, lignocellulosic biomass, and microorganisms, microalgae biomass is a promising type attributing to its intrinsic merits (Chang et al. 2018). Microalgae can be cultivated on nonarable lands using CO_2 as carbon source, wastewater as nutrients source, and solar light as energy source to produce intracellular fatty acids and carbohydrates at a photosynthetic efficiency over tenfold than terrestrial plants, realizing energy production, carbon mitigation, and wastewater remediation at the same time

Table 9.2 Major application of membranes in liquid biofuels production process and its advantages

Process	Examples	Advantages
Feedstocks preparation and pretreatment	Microalgae biomass cultivation and harvesting	For carbon supply: higher CO_2 transfer rate with membrane module, like hollow fiber membrane For nutrients supply: effective separation of microalgae with inhibitors in wastewater, like ion-exchange membrane For biomass harvesting: cost-effective microalgae biomass harvesting, like microfiltration or ultrafiltration membrane
Liquid biofuels generation	Fermentation for liquid biofuels generation (bioethanol, biolipids, etc.)	For enzyme recovery: enzyme recovery without damaged enzymatic activity, like microfiltration or ultrafiltration membrane For sugar concentration and inhibitor removal: simultaneously realize sugar concentration and inhibitors removal with low energy cost, like ultrafiltration, nanofiltration, reverse osmosis, and membrane distillation
Liquid biofuels recovery	Liquid products concentrating for downstream processing or utilization	Membrane distillation or pervaporation: low energy cost, pure products, and mild operating conditions, like the porous membrane for distillation and nonporous membrane for pervaporation Hybrid membrane process: realize more functions at the same time, like distillation–pervaporation system

(Georgianna and Mayfield 2012; Guo et al. 2018). It was reported that the lipid content of many microalgae species are over 50 times of the terrestrial oil crops (Chisti 2007). However, there are still many drawbacks that need to be addressed for the traditional approaches of microalgae biomass production, like poor light penetration, low carbon transfer rate, and inappropriate nutrients feeding, and from these aspects, membranes are useful to enhance the performance of the microalgae cultivation system (Chang et al. 2017; Fu et al. 2016).

Carbon is an important element for microalgae biomass, accounting for more than 50% of the microalgal dry cell weight (Chang et al. 2016b). However, the CO_2 transfer rate was usually very low, resulting in low carbon availability in microalgae culture and thus limiting microalgae growth and carbon fixation. To enhance CO_2 transfer efficiency in microalgae cultivation system, hollow fiber membrane (Mortezaeikia et al. 2016), selective CO_2 transfer membrane (Rahaman et al. 2011), and integrated alkali-absorbent membrane system (Ibrahim et al. 2018; Li et al. 2018b, 2018c; Zheng et al. 2016) were successfully adopted in their works. Results demonstrated that the carbon availability in microalgae suspensions was effectively improved and microalgae biomass was enhanced to some extents.

Besides carbon source, light and nutrients are also key factors influencing microalgal biomass concentration (Liao et al. 2018; Sun et al. 2016a, 2018). To exploit inorganic salts in wastewater as nutrients for microalgae cultivation, Chang et al. (2016a) designed an annular photobioreactor based on ion-exchange membranes for selectively transferring cations and anions from wastewater chamber to microalgae cultivation chamber but preventing transport of suspended solids in wastewater, ensuring high light penetration and proper nutrients availability in microalgae culture. The biomass concentration was increased to 4.24, 3.15, and 2.04 g/L in the membrane photobioreactor from 2.34, 2.15, and 0 g/L in the membraneless photobioreactor when using simulated agricultural, municipal, and industrial wastewater as nutrients source. Besides, a scalable membrane-based tubular photobioreactor was used in microalgae biomass and biofuels production, which effectively enhanced economic and technical feasibility of microalgae cultivation with membrane photobioreactor (Chang et al. 2019).

In addition to microalgae biomass cultivation, membrane is also used in microalgae harvesting for downstream fermentation or fatty acids extraction. As is known, microalgae suspension contains more than 99% of water in weight ratio. Recovery of biomass from microalgae suspension was estimated to contribute 20%–30% of total energy cost for biomass production (Huang et al. 2019; Wei et al. 2018). In contrast, membrane filtration with microfiltration or ultrafiltration membrane is known as an energy saving method for microalgae biomass harvesting than other methods like centrifugation or drying, since energy cost on transmembrane pressure for membrane filtration is much lower than conventional methods. But the membrane fouling is an inescapable problem for microalgae harvesting with membrane filtration. To cope with the fouling problem of filtering membrane, many approaches were proposed, like nanofiber membrane (Bilad et al. 2018), rotational-dynamic filtration membrane (Hapońska et al. 2018), axial vibration membrane (Zhao et al. 2016), and composite membrane (Khairuddin et al. 2019). However, the antifouling performance of the existing technologies is limited, which is not capable of greatly reducing the energy cost. Further researches on membrane fouling control are still necessary.

9.2.2 Membranes Used for Fermentation

Saccharification and fermentation are important steps for biomass conversion to liquid biofuels, directly determining biofuels productivity and quality. During these processes, membranes play important roles on enzyme recovery from hydrolysis solution, sugar enrichment, and detoxification of the fermentation broth.

Before fermentation, the macromolecular organic matters in the biomass should be firstly hydrolyzed into simple sugars by enzyme for fermentation. In detail, the hexose sugar monomer contained in cellulose and the pentose sugar monomer contained in hemicellulose should be released and hydrolyzed into simple sugars like glucose, and the complex lipids- and proteins-containing organic matters in microalgae biomass should be hydrolyzed into simple structures like long-chain fatty acids, glycerol, and amino acids (Kang et al. 2018). Then, the simple organics can be utilized by microorganisms for fermentation to produce liquid biofuels like bioethanol. Compared with chemical process for hydrolysis of cellulose like dilute acid catalyzed, enzymatic hydrolysis of cellulose has many advantages, including mild operation conditions, low energy cost, and low inhibitors formation (Li et al. 2019). But the cost on enzyme utilization is very high, accounting to almost half of the total cost on hydrolysis process (Wooley et al. 1999).

Recovery and reuse of the hydrolysis enzyme can effectively reduce energy cost on enzymatic hydrolysis process. Membrane-based technology, using various membranes like microfiltration and ultrafiltration membrane as physical barrier, is regarded as a promising approach for enzyme recovery from hydrolysis solution since it can retain the catalytic activity of the enzyme, ensuring high efficiency and low cost of biomass conversion to fermentative sugars (Saha et al. 2017). Membranes used for enzyme recovery are mainly divided into microfiltration and ultrafiltration membranes according to the pore size. Microfiltration membranes are usually made of cellulose acetate, nylon, or polysulfone, which can efficiently remove most of the remaining biomass in hydrolysis solution (Singh and Purkait 2019). And the ultrafiltration membranes which are made of polyethersulfone or polysulfone are frequently used in enzyme separation and extraction from the hydrolysis solution (Enevoldsen et al. 2007).

The fermentative sugar concentration in hydrolysate is usually low mainly due to low hydrolysis efficiency, limiting bioethanol production. In addition, many inhibitors for bioethanol fermentation are produced along with the hydrolysis process, which also plays negative effects on bioethanol output (Nguyen et al. 2018). Therefore, sugar enrichment and inhibitors removal of the hydrolysate are important steps to improve bioethanol productivity and reduce cost on downstream processing. Some conventional methods for sugar concentration and inhibitors removal include physical adsorption, thermal evaporation, solvent extraction, and ion exchange (Sambusiti et al. 2016; Tanaka et al. 2019; Zhang et al. 2018a). But these methods are energy intensive and cannot simultaneously realize sugar concentration and inhibitors removal. The application of membrane process can greatly reduce the energy cost and deal with the technological problems, like incompatible operation of sugar concentration and inhibitors removal. Nowadays, the commonly used membrane technologies for sugar concentration and inhibitors removal are ultrafiltration, nanofiltration, reverse osmosis, and membrane distillation. The characteristics of different membrane technologies have been reviewed by previous authors (Wei et al. 2014; Zabed et al. 2017). Although membrane technologies have many advantages for fermentation process, membrane fouling is still a troublesome problem which limits economic feasibility. Works to conquer the problem of membrane fouling is vital to reduce cost of hydrolysate pretreatment.

9.2.3 Membranes Used for Liquid Biofuels Recovery

The final liquid biofuels concentration is influenced by many factors, such as feedstock compositions, fermentative sugar concentration in hydrolysate, activity of the fermentative yeast, and operating parameters like pH and temperature. Taking bioethanol as an example, the final bioethanol concentration in a fermenter is usually low when using lignocellulose as feedstocks than that with food as feedstocks (Ferreira et al. 2018). In general, the bioethanol concentration is lower than 5% (in w/w) when using cellulose as feedstocks, meaning that the produced bioethanol must be firstly concentrated to a higher concentration for downstream processing. Besides, the products are usually inhibitive to yeast cells for continuous production. Therefore, separation and recovery of the bioethanol from a fermenter are significant for economical production of bioethanol at continuous mode. Among different biofuels recovery processes, membrane-assisted bioethanol recovery has particularly advantages of low energy requirement, pure products, and mild operating conditions over the traditional processes like distillation (Balat et al. 2008). The known membrane-based bioethanol recovery technologies include ultrafiltration, reverse osmosis, membrane distillation, pervaporation, and hybrid process; among them membrane distillation and evaporation are the two well-established methods nowadays (Bayrakci Ozdingis and Kocar 2018).

The working mechanism of membrane distillation is based on the differential vapor pressure at microporous hydrophobic membrane surface, which acts as the driving force for biofuels separation. For example, the ethanol partial pressure is higher than water; thus, ethanol vapor can transfer across the membrane in priority, and based on that, the separation of bioethanol from broth can be realized (Tomaszewska and Białończyk 2013). The commonly used membrane types for membrane distillation are prepared from low surface energy hydrophobic polymer like polypropylene, polytetrafluorethylene, and polyvinylidene fluoride (Saha et al. 2017). And a nonporous membrane is usually used in the pervaporation process to recover biofuels from solution by partial vaporization based on the solution–diffusion model (Trinh et al. 2019). During pervaporation, permeation of a component from solution to membrane and evaporation of the specific component from the membrane to vapor stream successively happen. In this way, the biofuels in solution can be selectively separated and recovered. Pervaporation membrane can be roughly classified into two types, i.e., hydrophilic membrane and hydrophobic membrane. The hydrophilic membrane is mainly used to remove water from the mixed solution, while the hydrophobic membrane is mainly used to extract biofuels from the liquid stream (Huang et al. 2008). Therefore, the hydrophobic membrane is more energy efficient for biofuels recovery when biofuels concentration in liquid is low, especially in the case for bioethanol recovery from digestate in which bioethanol concentration is usually less than 10% w/w.

In recent years, the hybrid processes have attracted wide attentions since it can fulfill the requirements for high-efficiency continuous biofuels production. The hybrid process integrates various units together for some specific functions. For example, the hybrid fermentation–pervaporation process can remove the produced bioethanol in situ to offset product inhibition and avoid yeast cells washout by holding back the yeast biomass with the membrane module (Santos et al. 2018). A hybrid system integrating membrane fermentation and cogeneration was proposed by Lopez-Castrillon et al. (2018), which effectively improved energy output efficiency of the fermentation system with possibility of additional electricity generation (275 kWh/t of cane). A hybrid extractive distillation column with high selectivity pervaporation was implemented in alcohol dehydration process, which demonstrated that the hybrid system could save up to 25%–40% of the total annual cost and energy (Novita et al. 2018).

9.3 Membrane Application on Gaseous Biofuels Production

Gaseous biofuels, like biohydrogen and methane, are also important renewable energy types which have been widely and practically used. For example, the biogas digester is commonly constructed in medium or small size dispersedly for household cases attributing to simple digester configuration and low investment (Chen et al. 2017). The bioreactors with sophisticated structure, like membrane-based bioreactors, are not suitable to be used in rural places attributing to their high cost but are frequently used in hydrogen production. Hydrogen is a clean energy than traditional fossil fuels, which generates only water as a by-product with zero greenhouse gas emissions during combustion while embracing larger energy content per unit mass (142 kJ/g) over other fuel types (Di Paola et al. 2015; Zhong et al. 2017). Compared with hydrogen production via thermochemical method like steam reforming and electrochemical method like electrolysis, biological hydrogen production has attracted particular interests due to its mild operating conditions, low energy consumption, and abundant feedstocks (Aslam et al. 2018a). However, biohydrogen productivity in large-scale application is still very low, hindering the commercialization of biohydrogen.

Many process parameters and environmental factors have significant influences on biohydrogen productivity, such as pH, temperature, substrate and nutrients availability, by-product and product concentration, microbial competition, and other hazardous materials (Liao et al. 2013; Prabakar et al. 2018). Researches are necessary to solve the remaining bottlenecks to practical applications of biohydrogen energy. Among many emerging approaches for high-efficiency biohydrogen production, membrane-integrated biohydrogen production system is for sure a promising technology allowing for dealing with various kinetic inhibitions in biohydrogen production, like biomass washout and substrate or product inhibition, as shown in Table. 9.3 (Aslam et al. 2018a).

Biological hydrogen production is a technology that produces hydrogen gas with microorganisms. It can be roughly classified into photo-dependent biohydrogen production via photolysis of water by algae and cyanobacteria or photo-fermentation by decomposing organic matters with photosynthetic bacteria and dark fermentation

Table 9.3 Major application of membranes in gaseous biofuels production process

Process	Target of membranes	Characteristics
Photo-dependent biohydrogen	Algae, cyanobacteria, or photo-fermentation with photosynthetic bacteria	Membrane application mainly focused on downstream products refining
Dark-fermentative biohydrogen	Anaerobic conditions that avoid oxygen inhibition and light inhibition	Submerged membrane bioreactor: low energy cost but high membrane area Side-stream membrane bioreactor: small membrane area but high transmembrane pressure, high energy cost
Products purification	Remove impurities for quality upgrading of gaseous biofuels	Gas transfer mechanisms of the membrane: (1)viscous flow, (2) surface diffusion, (3) Knudsen diffusion, (4) capillary condensation, (6) molecular sieving, (7) solution diffusion, (8) facilitated transport, etc. (Bakonyi et al. 2018; Li et al. 2015a; Lundin et al. 2017) Key criteria for the membrane: (1) permeability and (2) selectivity

for hydrogen production with facultative or obligate anaerobic bacteria (Trchounian et al. 2017).

9.3.1 Membranes Used for Photo-dependent Biohydrogen Production

During photolysis, which is the first case of the photo-dependent biohydrogen production, some oxygenic photosynthetic microorganisms like algae or cyanobacteria strains absorb solar energy and convert it into chemical energy by splitting water to proton (H^+) and molecular oxygen (O_2) with intracellular pigments (Yilanci et al. 2009). Then the generated H^+ acts as electron acceptor for H_2 production in the downstream combination with excessive electrons assisted by intracellular enzyme of algal or cyanobacterial cells (He et al. 2017). Besides H_2 generation, the technology also realizes high-efficiency carbon mitigation since the growth and metabolism of algae or cyanobacteria can absorb ambient CO_2 as carbon source at solar energy conversion efficiency of tenfold than terrestrial plants (Khetkorn et al. 2017). Thus, biohydrogen production via photolysis is regarded as the cleanest way of hydrogen production, but its application is severely inhibited by low hydrogen productivity, oxygen inhibition, and strict light requirement (Argun and Kargi 2011). Many works were reported on enhancement of photolysis biohydrogen production. Ban et al. (2018) found that Ca^+ was capable of decreasing the rate of chlorophyll reduction, maintaining the protein content at high level, and scavenging most of reactive oxygen species, which improve direct and indirect photolysis H_2 production, with the maximum value of 306 ml/L H_2 under Ca^+

adding amount of 5 mM. Rashid et al. (2013) applied mechanical agitation of culture medium in the photobioreactor to enhance oxygen escape from suspensions to reduce inhibiting effect of oxygen on biohydrogen production in microalgae system.

Unlike photolysis with algae or cyanobacteria, photo-fermentation with photosynthetic bacteria like non-sulfur purple photosynthetic bacterium, which is regarded as the second case of photo-dependent biohydrogen production, is unable to derive electrons from water. Photo-fermentation bacteria usually use simple sugars and volatile fatty acids as feedstocks (Zhang et al. 2018b). And many problems like high energy demand, low light conversion efficiency, and uneven light distribution in bioreactors still need to be addressed for photo-fermentation. To enhance the light conversion efficiency and improve the uneven light distribution in reactors, two kinds of optical fibers with high surface luminous intensity have been developed by using the polymer optical fiber and hollow quartz optical fiber (Xin et al. 2017; Zhong et al. 2016, 2019), respectively, and the prepared fibers have been applied in the photoreactors (Zhong et al. 2019). Tian et al. (2010) adopted a cell immobilization technique to a biofilm-based photobioreactor to enhance light conversion efficiency and biohydrogen production rate with photosynthetic bacteria *Rhodopseudomonas palustris CQK 01*. By cultivating photosynthetic bacteria on the surface of packed glass beads in the work by Tian et al. (2010), the maximum biohydrogen production rate was improved to 38.9 mL/L/h and the light conversion efficiency was enhanced to 56%. Fu et al. (2017) adopted light guide plate in photo-fermentation system to realize uniform light distribution in the system and enhance biohydrogen production. In the system, light was supplied from one side of the light guide plate and then emitted from the surface of the plate, in which way the light was elaborately dispersed in the culture. As a result, the hydrogen production rate was improved to 11.6 mmol/h/m^2.

Unfortunately, applications of membrane technology on photo-dependent biohydrogen production system are relatively scarce up to date, which are mainly focused on downstream purification of hydrogen products (Lin et al. 2018). Since some membranes have the ability to selectively separate gas and liquid components as well as regulate mass and heat transfer, membrane integrated photobioreactors for biohydrogen production are expected to enhance photo-biohydrogen production.

9.3.2 Membranes Used for Dark-Fermentative Biohydrogen Production

Compared with biohydrogen production via photolysis or photo-fermentation, dark-fermentative biohydrogen production occupies more predominant status nowadays. Dark fermentation presents many advantages over photo-fermentation. Since light is unnecessary for dark fermentation process, reactors design is more flexible for dark fermentation, and the volume utilization of the bioreactors can be fully exploited (Łukajtis et al. 2018). In addition, oxygen inhibition is no longer a problem in anaerobic conditions; dark-fermentative biohydrogen production shows more reliable and faster hydrogen production rate.

For conventional dark fermentation process, continuous stirred-tank reactor (CSTR) is widely used due to its simple construction, effective mixing, and ease of operation. But low biomass density in fermentative broth of the CSTR caused by high biomass washout rate and by-product and product inhibitions are crucial shortcomings for feedstocks conversion and hydrogen production (Kariyama et al. 2018). The membrane modules in anaerobic membrane bioreactor (AnMBR) typically assist the biochemical conversion processes of feedstocks to hydrogen by ensuring high solid retention time (SRT) and selectively removal of inhibiting products (Shin and Bae 2018). In detail, membranes can separate liquid stream from biomass and thus retain biomass in the bioreactor, in which way long SRT required for efficient wastewater treatment and short hydraulic retention time (HRT) for cost-effectiveness are satisfied at the same time (Aslam et al. 2018b). In addition, membranes in the bioreactors can retain the metabolites in the system for further conversion to produce biohydrogen, enhancing the substrate conversion efficiency (Park et al. 2017). For example, Nielsen et al. (2001) used a heated palladium–silver membrane reactor to separate hydrogen from the gas stream, in order to eliminate the inhibiting effects of products (H_2) on H_2 generation. Teplyakov et al. (2002) integrated active polyvinyl-trimethyl-silane membrane system with dark-fermentative bioreactor for hydrogen removal to reduce partial pressure of hydrogen in the gaseous units.

In general, the membrane bioreactor can be mainly classified into two types: submerged membrane bioreactor and side-stream membrane bioreactor (as shown in Fig. 9.2). Membrane modules are usually submerged in the liquid phase of the reactor for the submerged membrane bioreactor, while they are set outside of the reactor as a separate unit for the side-stream membrane bioreactor (Łukajtis et al.

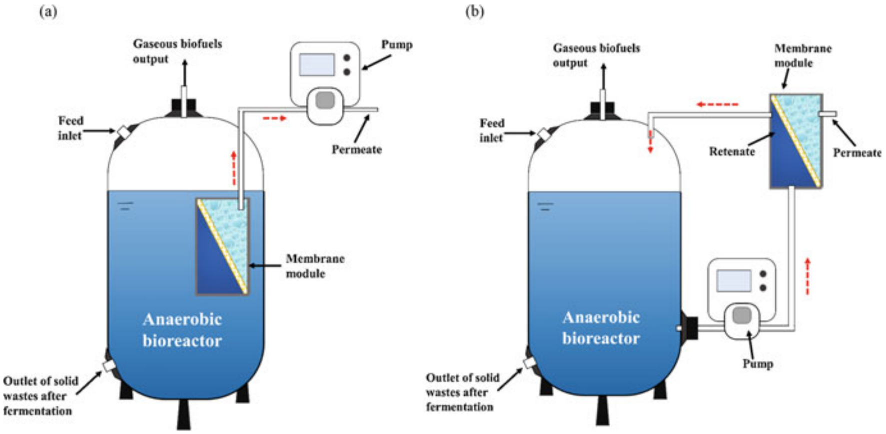


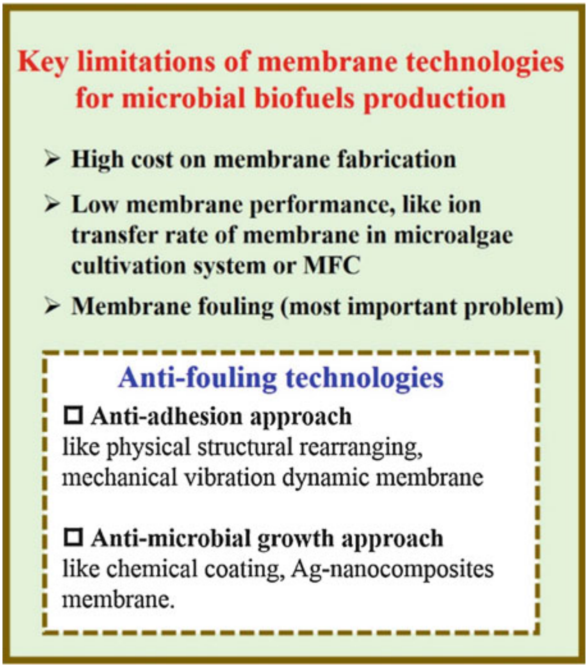
Fig. 9.2 Configurations of (**a**) the submerged membrane bioreactor (MBR) and (**b**) the side-stream MBR for gaseous biofuels production

2018). The side-stream membrane bioreactor is characterized by small exchange area of the membrane and easy conduction of membrane washing. However, a high energy cost is required to supply enough transmembrane pressure for the filtration of fermentative broth. On the contrary, the energy cost in the submerged membrane bioreactor is much lower than the side-stream membrane bioreactor, but larger membrane exchange area is necessary (Aslam et al. 2018a). Recently, many derived types of membrane bioreactor are proposed for high-efficiency biohydrogen production. Bakonyi et al. (2015) established a double-membrane bioreactor, in which a commercial microfiltration membrane module was added into a membrane hydrogen fermenter, which realized simultaneous biohydrogen production and purification. A dynamic membrane bioreactor integrating a self-forming dynamic membrane with a continuous fermenter was constructed by Park et al. (2017). In the dynamic membrane bioreactor, the membrane module successfully retained effective hydrogen-producing–bacterial consortia, resulting in a maximum hydrogen production rate of 51.38 L/L/day. Saleem et al. (2018) adopted a side-stream dynamic membrane bioreactor using dynamic membrane as a solid–liquid separation media and significantly improved the dark-fermentative biohydrogen production under mesophilic conditions.

9.3.3 Membranes Used for Biohydrogen Purification

Another important role of membrane in biohydrogen production system is purification of the gaseous products to obtain high-quality hydrogen fuel. During biohydrogen production via photo- or dark fermentation, large quantities of by-products are generated along with hydrogen gas, like CO_2, CO, SO_x, and NO_x, which have great negative effects on combustion property of biohydrogen as fuel (Khan et al. 2018). It is important to remove the impurities with CO_2 as a major target for gas upgradation. Membrane technology for biohydrogen purification is a feasible approach because it avoids chemical conversion of the mixed gas.

In general, a membrane is a semipermeable separator which acts as a selective mass transfer barrier to realize separation of different compositions (Bakonyi et al. 2018). According to membrane type (porous or nonporous membrane), gas transfer mechanisms of the membrane mainly include (1) viscous flow, (2) surface diffusion, (3) Knudsen diffusion, (4) capillary condensation, (6) molecular sieving, (7) solution diffusion, and (8) facilitated transport, which are elaborately described in the previous paper (Bakonyi et al. 2018; Li et al. 2015a; Lundin et al. 2017). Superior permeability and selectivity are two key criteria for the membrane applied in gas purification, but it is unfortunate that these two factors are usually not compatible with each other. This limits application of most available membrane types in industrial production of biohydrogen. Many researchers have been dedicating so much effort to enhance the gas separation characteristics of membranes for biohydrogen purification. Ahmad et al. (2016) constructed a nearly superhydrophobic and microporous membrane by blending amorphous poly-



Key limitations of membrane technologies for microbial biofuels production

- ➢ High cost on membrane fabrication
- ➢ Low membrane performance, like ion transfer rate of membrane in microalgae cultivation system or MFC
- ➢ Membrane fouling (most important problem)

Anti-fouling technologies

☐ Anti-adhesion approach
like physical structural rearranging, mechanical vibration dynamic membrane

☐ Anti-microbial growth approach
like chemical coating, Ag-nanocomposites membrane.

Fig. 9.3 Key limitations of membrane application in microbial biofuels production process (Buitrón et al. 2019)

benzimidazole and semicrystalline polyvinylidene fluoride, which removed 67% of CO_2 in gas mixture of H_2 and CO_2 at highest CO_2 flux of 4.16×10^{-4} mol/m^2/s across the membrane. Wu et al. (2017a) synthesized a membrane made of glassy polymers, polyetherimide-coated bio-cellulose nanofibers, and a coconut shell active carbon as adsorbent carriers for CO_2 separation in dark-fermentative gas mixture. The synthesized membrane was convinced to have CO_2 permeability of 16.72 Barrer and corresponding CO_2/H_2 selectivity of 0.15. Abd. Hamid et al. (2019) proposed a synthesized polysulfone–polyimide membrane with the highest permeability of 348 GPU (gas permeation unit, 1 GPU equal to 1×10^{-6} cm^3(STP)/(cm^2•s•cm Hg)) for H_2 and 86 GPU for CO_2, H_2/CO_2 selectivity of 4.4, and H_2 purification efficiency of 80%.

However, many previous literatures also reported that the equipment cost, reliability, and energy efficiency of the membrane bioreactor are unable to compete with the traditional CSTR. Among various influencing factors, membrane fouling is one of the most important problems, as seen in Fig. 9.3 (Buitrón et al. 2019). During microorganism growth and metabolism, a quantity of soluble microbial products and extracellular polymeric substances which consists of complex biopolymer mixtures like proteins, polysaccharides, lipopolysaccharides, and lipoproteins, is produced in the cultures (Zhang et al. 2015). With assistance of the excretive soluble microbial products and extracellular polymeric substances, the biomass flocs are easily attached and accumulated on membrane surface since the biomass flocs are usually

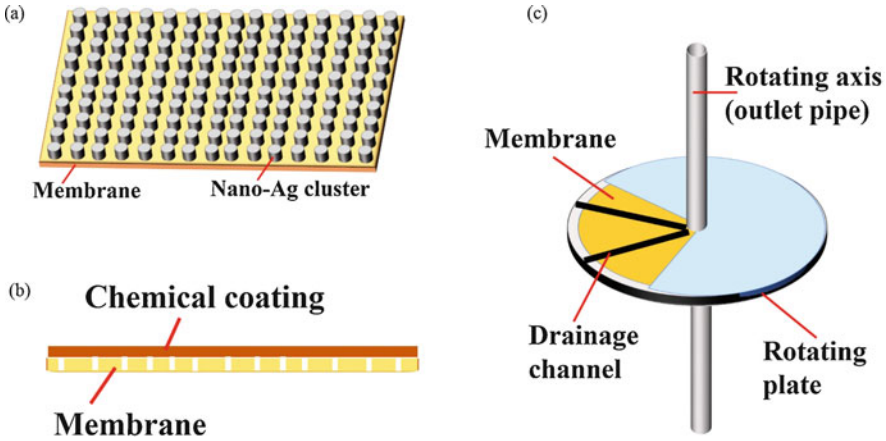


Fig. 9.4 Typical antifouling membrane system. (**a**) Membrane surface modification with nano-Ag cluster, (**b**) chemical coating of membrane and (**c**) dynamic membrane system with rotating unit (Qin et al. 2018)

larger than the membrane pore size, resulting in pore blocking and membrane fouling (Khan et al. 2019; Zhang et al. 2015).

In this regard, enhancement of physical–chemical properties of the membrane to reduce foulant attaching on the membrane surface is a primary objective to prevent membrane fouling. Membrane modifications with physical structural rearranging, chemical coating, and functional material embedding are promising approaches for antifouling membrane development (López-Cázares et al. 2018; Qin et al. 2018; Shan et al. 2018). Schematic of some typical membrane modification methods for antifouling technology is shown in Fig. 9.4, like physical structural modification with nano-Ag cluster (Fig. 9.4a) and chemical solvents coating on the membrane (Fig. 9.4b). For example, López-Cázares et al. (2018) enhanced the anti(bio)fouling of cation exchange membranes (Nafion and Ultrex membranes) by immobilizing nanocomposites of nanoparticles on graphene oxide as a thin film using a polydopamine adhesive. Shan et al. (2018) explored a facile and biomimetic method of amphiphobic surface with special structure and controllable wettability, which enhanced the flux and antifouling performances of the membrane. Li et al. (2018a) grafted thermo-responsive polymer chains on the surface of polyethersulfone, developing a modified membrane with rich porosity and well antifouling property.

Another important antifouling approach is dynamic membrane technology which uses a physical barrier to prevent formation of cake layer on the membrane surface (Yang et al. 2018). Compared with the conventional approaches to control membrane fouling by air bubbling, the dynamic membranes can provide stronger shear force on the phase interface of the liquid and membrane by mechanical vibration, like rotating, vibrating, and oscillating (Bagheri and Mirbagheri 2018; Qin et al. 2018). The typical dynamic membrane system, like membrane rotating system, is shown in Fig. 9.4c. Ruigómez et al. (2017) proposed a physical cleaning strategy based on membrane rotation in a submerged anaerobic membrane bioreactor and improved the fouling removal effectiveness, achieving a stable net permeate flux of 6.7 L/m^2 h. Chatzikonstantinou et al. (2015) employed high-frequency powerful vibration technique in both hollow fiber and flat sheet modules to prevent membrane fouling. They reported that the strategy of high-frequency powerful vibration is capable of reducing membrane fouling and is promising with respect to energy savings. These emerging antifouling technologies provide great potential to reduce membrane manufacturing and operating costs, which then enhance the commercial feasibility of biohydrogen application as energy sources.

9.4 Membrane Application in Microbial Fuel Cells

Microbial fuel cells (MFCs), which are bioelectrochemical devices, have attracted a particular interest in the energy field due to its environmental-friendly characteristic by using microorganism as electrocatalyst to conduct an oxidation–reduction reaction and convert chemical energy in wastewater into electrical energy (Leong et al. 2013; Zhong et al. 2018). The configuration of MFCs generally contains three parts, anode, cathode, and electrolyte layer, in which the MFCs can be roughly classified into two types, i.e., dual chamber MFC and single chamber MFC (as shown in Fig. 9.5). The dual chamber MFC contains an anode and a cathode chamber, which are separated by a proton exchange membrane that acts as electrolyte bridge. In contrast, the single chamber MFC contains only anode chamber, with air as the cathode of the system. The MFC has dual advantages of simultaneous electricity generation and treating wastewater, but commercialization of this technology is still hindered by high cost (Tender et al. 2008) and low power density (Tender et al. 2002).

The membrane is a major part of the MFC acting as separator that physically divides the anode and cathode but keeping them chemically and ionically connected, which significantly influences the MFCs' overall investment and power density.

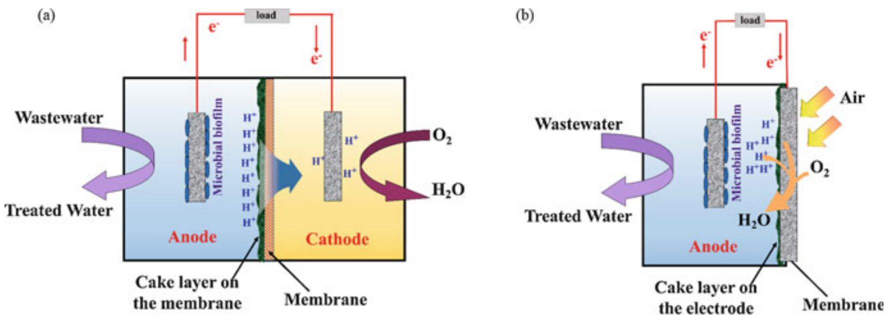


Fig. 9.5 Schematic diagram of (**a**) the dual chamber microbial fuel cell (MFC) and (**b**) the single chamber MFC

Until now, the possible types of membranes that can be used in the MFC include cation exchange membrane (Daud et al. 2018), anion exchange membrane (Elangovan and Dharmalingam 2017), porous membrane (Li et al. 2015b), polymer/composite membrane (Ahilan et al. 2018), etc. Each type of membrane has its advantages and disadvantages. For example, cation exchange membrane is the preferential separator used in MFC since it directly conducts H^+ from anode to cathode, which enhances coulombic efficiency of the MFC (Chaudhuri and Lovley 2003). pH splitting between the anode and cathode chamber of the MFC easily happened, attributing to transfer competition of other cations (like K^+, Na^+, NH_4^+, and Ca^{2+}) with H^+ across the cation exchange membrane, which may cause H^+ accumulation in anolyte (Chae et al. 2008). The anion exchange membrane can effectively diminish pH splitting since the AEM conduct OH^- or carbonate anions transfer from cathode to anode, promoting H^+ transfer by acting as H^+ carrier (Varcoe et al. 2014; Ye and Logan 2018). However, the substrate crossover through the AEM is a major drawback for MFC performance (Hernández-Flores et al. 2017). Though the internal resistance of porous membrane is low, it is not a good candidate for the MFC, attributing to high crossover rate of oxygen and substrate through the pores, except for cases when aerobic bacterium in anode is intended to be cultivated for removal of some specific organic matters, like azo bonds during azo dyes treatment (Slate et al. 2019). Polymer/composite membrane is a newly emerging type which combines merits of polymers and inorganic or organic fillers to realize more abundant functions, but it is in cost of larger surface roughness, resulting in higher possibility of biofouling (Antolini 2015). In general, the membrane affects MFCs' performance and cost from aspects of membrane internal resistance, oxygen diffusion, substrate loss across the membrane, pH splitting, and membrane biofouling (Dharmalingam et al. 2019; Leong et al. 2013).

The membrane with high resistance is not conducive to proton diffusion from anode to cathode due to low ion-exchange capacity of the membrane, resulting in poor MFC performance, while low resistance membrane with porosity like microfiltration membrane can also reduce the power density of the MFC, attributing to high crossover rate of oxygen and substrate through the pore on the membrane (Zhao et al. 2009). Therefore, the membrane with low internal resistance and low oxygen and substrate crossover rate is an ideal type for improving coulombic efficiency and power density of the MFC (Ji et al. 2011). Gao et al. (2018) developed a novel carbon-based conductive membrane that had a lower internal resistance (752 Ω) relative to the proton exchange membrane (937 Ω) and enhanced the power density of the MFC to 228 mW/m^3. Wu et al. (2017b) adopted an electroconductivity aerated membrane (EAM) as biocathode in the MFC to enhance power density and wastewater treatment. The EAM had superior property in controlling oxygen and substrate diffusion as well as proton transfer, resulting in a power density of 4.20 ± 0.13 W/m^3 at a current density of 4.10 ± 0.11 A/m^2.

Oxygen and substrate diffusion across the membrane are important issues for MFC which can significantly reduce MFC's power density and coulombic efficiency (Do et al. 2018). Oxygen transfers from cathode to anode and then competes with the anode to accept electrons since oxygen is a more favorable electron acceptor. In contrast, the substrate transfers across the membrane from anode to cathode chamber, which is in opposite direction of oxygen diffusion. The substrate is then oxidized by aerobic bacteria, and extra electrons are generated for the oxygen reduction reaction at the cathode, leading to an internal short circuit inside the MFC and reducing coulombic efficiency (Kim et al. 2013). Thus, the occurrence of oxygen and substrate diffusion across the membrane diminishes the power density of the MFC. The membrane in the MFC acts as a physical barrier for oxygen and substrate diffusion during operation. From this view, the performance of the MFC with membrane is usually better than the membraneless MFC. For example, it was reported that the coulombic efficiency of the MFC with membrane was 20% higher than the membraneless one (Li et al. 2018b; Slate et al. 2019). Unfortunately, a membrane that can totally avoid oxygen and substrate diffusion is still not yet developed. Some auxiliary approaches are necessary to minimize negative effects of oxygen and substrate crossover on MFC performance. For example, Ahilan et al. (2018) modified ceramic membrane with montmorillonite–$H_3PMo_{12}O_{40}/SiO_2$ composite to reduce the oxygen mass transfer coefficient to 5.62×10^{-4} cm/s, which is near the commercia polymeric Nafion membrane. Logan et al. (2005) used chemical oxygen scavenger, i.e., cysteine, in the anode chamber to remove the oxygen by reacting with oxygen to form disulfide dime (cystine). Yousefi et al. (2018) assembled a chitosan/montmorillonite nanocomposite film layer-by-layer over the surface of commercial unglazed wall ceramics to be utilized as the separator of MFC, in which the oxygen diffusion coefficient was one-sixth of the blank ceramic membrane. To avoid substrate diffusion, a membrane which is nonporous and has high selectivity for cations but does not allow anions transfer is the preferred approach (Leong et al. 2013).

The oxygen and substrate diffusion can also induce biofouling of the membrane and pH splitting of the MFC, which cause negative effects on MFC performance. The membrane biofouling usually occurs on the membrane surface facing the anode chamber due to the attachment of microbial and organic matter as a biofilm (Chae et al. 2008). Besides, oxygen near the membrane in the anode side that transferred from the cathode triggered biofilm formation of aerobic bacteria, which acts as barrier for proton diffusion between the anode and cathode (Li et al. 2018b). Thus, the produced H^+ in the anode accumulates in the anolyte, making the anolyte more acidic and the catholyte more alkaline. The phenomenon of pH splitting may deteriorate bacterial growth and metabolism and then reduce power density and coulombic efficiency. To ensure high performance of the MFC, the fouled membrane must be replaced with new one for proton diffusion, but this dramatically improved operating investment of the MFC. In recent years, researchers proposed some approaches to reduce membrane biofouling, like antimicrobial approach and anti-adhesion approach (Chatterjee and Ghangrekar 2014; Noori et al. 2018; Sun et al. 2016b; Yang et al. 2016). Chatterjee and Ghangrekar (2014) constructed antifouling MFC using vanillin as biocide. Yang et al. (2016) coated the membrane with a silver nanoparticle–polydopamine to mitigate biofouling of the membrane by taking advantage of antimicrobial effect of nano-Ag particle. Sun et al. (2016b) used well-ordered multi-walled carbon nanotubes and its derivative modified with the carboxyl-modified to prevent microbial adhesion. However, the effectiveness of these antifouling methods drastically reduced after a certain period of operation. Until now, biofouling is still one of the biggest limitations for membrane application in MFC field, which will deteriorate membrane performance and durability and then negatively affect the power output and operational cost (Do et al. 2018; Gajda et al. 2018).

In conclusion, the membrane is a very important component for the MFC. The properties of mass transfer like H^+, oxygen, and substrate; energy transfer like thermal, chemical, and electrical; and energy conversion between chemical, electrical, and thermal power in the MFC system are closely related to the function and structure of the membrane modules, which ultimately affects MFC's performance. Among various available membranes, the choice of an ideal type for the MFC requires certain criteria, including internal resistance; ion conductivity; permeability; physical, chemical, and thermal stability; biofouling; and cost (Dharmalingam et al. 2019; Rabaey and Verstraete 2005). A superior membrane with characteristics of high ionic conductivity and high antibiofouling property but with low internal resistance, low oxygen, low substrate diffusion rate, and low cost is needed to be developed for large-scale application of MFC.

9.5 Conclusions

Microbial energy conversion technology is a potential method for simultaneous realization of environmental remediation and energy production. Membranes play very important roles in bioenergy production processes for enhancement of bioenergy productivity and quality. This chapter presents a review on the roles and mechanisms of membranes on bioenergy production processes, and the important influencing factors are discussed. For liquid biofuels production, membranes can enhance microalgae biomass productivity, concentrate sugar concentration, remove inhibitors from the hydrolysate, and recover liquid biofuels from solution. For gaseous biofuels production, the membranes can enhance bioenergy output by ensuring high solid retention time (SRT) and purify the produced biogas for high-quality fuel generation. For the microbial fuel cell, the membrane can avoid internal short circuit and increase power density by acting as physical barrier and electrolyte bridge. But biofouling of membrane caused by microbial attachment is a vital problem that needs to be addressed. Antifouling technologies, like anti-adhesion approach or antimicrobial growth approach, are discussed in the work. For future prospect, antifouling technology of membranes is still the primary target to reduce membrane cost. Some versatile membrane types coated with functionalized groups or materials should be developed to fulfill various occasions. In addition, further application of membrane on microbial energy conversion should be explored, like membrane application on photo-dependent hydrogen production.

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