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REVIEW

Membrane applications for microbial energy conversion: a review

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Abstract
Technologies for conversion of microbial energy have recently attracted interest to transform waste into bioenergy, thus addressing simultaneously environmental and energy issues. Nonetheless, actual microbial systems for energy conversion have limitations such as low rate of mass transfer, uneven energy distribution and strong inhibition of products and by-products. These technical bottlenecks can be alleviated by using membranes, which regulate the transfer of mass, heat and energy. Here we review applications of membranes for microbial energy conversion. We discuss mechanisms, functions and development of membranes for feedstock preparation, bioenergy production and bioproduct post-treatment. We present key membrane factors that control the efficiency of microbial fuel cells. We address membrane biofouling problems and anti-fouling approaches, in order to improve future commercialization.

Keywords Membrane · Microbial energy · Microbial fuel cell · Biohydrogen · Bioreactor · Biofouling

Introduction

Nowadays, human beings still rely on traditional fossil fuels like natural gas, coal and petroleum as predominant fuel types for activities. But limited reservoir, depleting supply and ever-increasing consumption restrict the dependency on traditional fossil fuels as major energy sources (Chang et al. 2020a, b; Kumari and Singh 2018). Besides, many environmental problems associated with fossil fuels combustion have proposed pressing needs to develop renewable and environmental-friendly energy sources which are derived from non-fossil sources in ways that can be replenished (Bouabidi et al. 2018; Fu et al. 2018; Guo et al. 2019). Among various renewable energy types, like solar, wind, hydro, geothermal and biofuels (Aravind et al. 2020), 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 (Srivastava et al. 2017).

According to feedstock types, microbial biofuels were mainly classified into the first-, second- and third-generation biofuels (Nigam and Singh 2011), as shown in Table 1. The first-generation biofuels are produced using edible substrates as feedstocks, like oleaginous crops or starch-containing crops, which required relatively simple pretreatment of feedstocks since the materials are easy to be degraded than lignocellulose. But the competition of arable land and freshwater with human beings' food for the first-generation biofuels production strongly restricted its application (Correa et al. 2017). The second-generation biofuels fulfill the gaps of the first-generation biofuels due to utilization of non-edible substrates from forestry and agricultural lignocellulose. But sophisticated pretreatment processes of feedstocks are necessary to hydrolyze tight crystalline structure of cellulose, leading to greatly increase the energy cost on

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Table 1 Various generations of biofuels (Correa et al. 2017; Leong et al. 2018; Nigam and Singh 2011; Kumari and Singh 2018)

Biofuel 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 fresh water 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, wastes 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

the second-generation biofuels (Kumari and Singh 2018). Comparing with the first- and second-generation biofuels, the third-generation biofuels deriving from microorganisms (microalgae, microbes, etc.) are considered as promising alternatives since they can avoid major disadvantages of food competition and non-biodegradability (Zhu et al. Zhu et al. 2018). Many microorganism species (like microalgae, yeast and fungi) have abilities to accumulate fatty acids or sugar in the cells, which can be used as substrates for biodiesel or biohydrogen production through downstream processing of the microbial biomass (Leong et al. 2018, Hu et al. 2018).

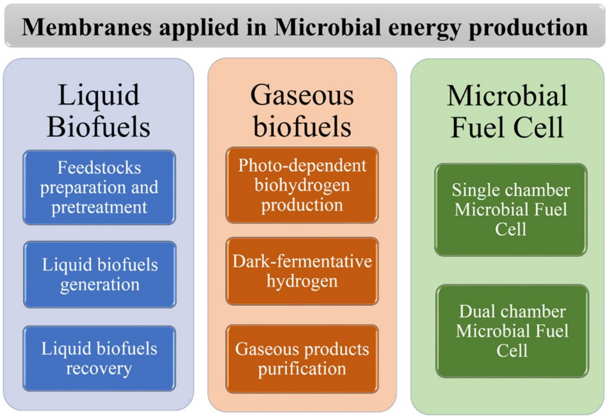


Fig. 1 Major application of membranes in microbial energy production processes

Until now, the biofuel productivity and quality are poor due to many technical bottlenecks like low utilization and conversion efficiency of feedstocks, difficulty on products separation and purification (Hajilary et al. 2019; Hu et al. 2019). During microbial energy conversion processes, mass transfer and heat distribution usually determine the operating conditions of system (temperature, pH, material proportion, retention time, etc.), which ultimately affects direction and rate of the chemical reactions, like lignocellulose hydrolysis to produce sugars, sugars fermentation to produce bioethanol or biohydrogen (Pei et al. 2017). The introduction of membrane modules in microbial energy conversion system can obviously regulate the mass and heat transfer by acting as physical barrier, internal bridge, inhibitors separator or products extractor, which thus avoid many technique limitations of the system. The functions of membrane vary with its utilizing occasions. In the paper, major application of membranes on microbial energy conversion processes, i.e., liquid biofuels, gaseous biofuels and microbial fuel cell, is discussed and illustrated in Fig. 1. In detail, we reviewed (a) membrane application on liquid biofuels production, mainly on biomass cultivation and harvesting, liquid biofuel generation and liquid product refining, (b) membrane application on gaseous biofuel production, mainly on photo-dependent biohydrogen production, dark fermentative biohydrogen production and gaseous product purification, (c) membrane application on microbial fuel cell, (d) membrane biofouling and anti-biofouling technologies. This article is an abridged version of the chapter by Chang et al. (2020a, b).

Membrane application on liquid biofuel 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. The process of liquid biofuels production mainly includes feedstock preparation like microalgae cultivation and harvesting, liquid biofuels generation like fermentation and products refining like bioethanol and biodiesel recovery. Among these steps, membrane plays important role on enhancement of liquid biofuels production over the membrane-less approaches. Major application of membranes in liquid biofuels production and its advantages are shown in Table 2.

Membranes for microalgae cultivation and harvesting

Abundant biodegradable feedstocks are prerequisites for economy feasible liquid biofuel production (Hajilary et al. 2019). Among various feedstocks, microalgae biomass is considered as a promising candidate owing to more than ten times higher photosynthesis efficiency than land plants (Chang et al. 2018). Besides, microalgae can be cultivated on non-arable land with wastewater and exhausted gas as nutrients and carbon source to produce intracellular sugar and lipid (Guo et al. 2019). However, there are many limitations that need to be addressed for the microalgae biomass production system, like poor light penetration, low carbon transfer rate and inappropriate nutrients feeding. From these aspects, membranes are useful to enhance the performance of the microalgae production system.

As is known, final microalgal biomass concentration is affected by CO_2 transfer rate, light and nutrients (Chang et al. 2016; Sun et al. 2018). 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) were successfully adopted in their works. Results demonstrated that the carbon availability in microalgae suspensions was effectively improved and microalgae biomass was enhanced. To exploit inorganic salts in wastewater as nutrients for microalgae cultivation, Chang et al. (2018) designed an annular photobioreactor based on ion exchange membranes for selectively transfer of cations and anions from wastewater chamber to microalgae cultivation chamber but prevented transport of suspended solids in wastewater, ensuring high light penetration and proper nutrients availability in microalgae culture. Furthermore, 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).

Besides microalgae biomass cultivation, membrane is also used in microalgae harvesting to reduce energy cost on microalgae biomass enrichment. Filtration with microfiltration or ultrafiltration membrane is known as 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 membrane-free methods (Wei et al. 2018). But 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).

Table 2 Major application of membranes in liquid biofuels production

Process	Examples	Advantages
Feedstock 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 Biofuel generation	Fermentation for liquid biofuel generation (bioethanol, biolipids, etc.)	For enzyme recovery: enzyme recovery without damage enzymatic activity, like microfiltration or ultrafiltration membrane For sugar concentrating and inhibitor removal: simultaneously realize sugar concentrating and inhibitors removal with low energy cost, like ultrafiltration, nanofiltration, reverse osmosis and membrane distillation
Liquid biofuel recovery	Liquid product 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 non-porous membrane for pervaporation Hybrid membrane process: realize more functions at the same time, like distillation–pervaporation system

Membranes used for liquid biofuel fermentation

Saccharification and fermentation are important steps for biomass conversion to liquid biofuels, directly determining biofuel productivity and quality. Before fermentation, the macromolecular organic matters in the biomass should be first hydrolyzed into simple sugars by enzyme. In detail, the hexose sugar monomer contained in cellulose and the pentose sugar monomer contained in hemi-cellulose should be released and hydrolyzed into simple sugars like glucose. The complex lipid- and protein-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). Enzymatic hydrolysis of feedstocks is commonly used methods attributing to many advantages, like mild operation, low energy cost and low inhibitors formation, but the economy input is high due to enzyme consumption. Recovery and reuse of the hydrolysis enzyme using membranes are a promising approach to reduce economic cost on enzyme utilization since membrane can retain the catalytic activity of the enzyme, ensuring high-efficiency and low cost of biomass conversion to fermentative sugars (Saha et al. 2017). According to pore size, membrane used for enzyme recovery mainly includes microfiltration and ultrafiltration types, in which microfiltration membrane is usually made of cellulose, acetate and polysulfone, and functioning as barrier to remove remaining biomass from enzyme solution (Singh and Purkait 2019), while ultrafiltration membrane is always made of polyethersulfone or polysulfone and is frequently used to extract enzyme from the hydrolysis solution (Enevoldsen et al. 2007).

Considering low sugar concentration and high inhibitors content of hydrolysate, membranes are also used on sugar enrichment and inhibitors removal for highly efficient fermentation. Comparing with conventional methods for sugar concentrating and inhibitors removal, like physical adsorption, thermal evaporation, solvent extraction and ion exchange, the membrane-based technology has additional benefits of low energy cost and biocompatibility (Tanaka et al. 2019). Up to date, the commonly used membrane technologies for sugar concentrating and inhibitors removal are ultrafiltration, nanofiltration, reverse osmosis and membrane distillation. The characteristics of different membrane technologies have been reviewed by previous authors (Zabed et al. 2017).

Membranes for recovery of liquid biofuels

In general, final biofuel purity is usually poor attributing to many factors, like sugar concentration of hydrolysate, activity of fermentation microbial, operating conditions. For example, bioethanol concentration is usually lower than 5% (in w/w) when using cellulose as feedstocks, indicating that recovery and enrichment of biofuels from hydrolysate are necessary. Among various biofuels' recovery processes, membrane-assisted biofuel recovery has particular advantages of low energy demand, pure products and mild operating conditions (Balat et al. 2008). The known membrane-based bioethanol recovery technologies include ultrafiltration, reverse osmosis, membrane distillation, membrane pervaporation and hybrid process, among which membrane distillation and membrane evaporation are two well-established methods nowadays (Bayrakci Ozdingis and Kocar 2018).

Working mechanism of membrane distillation is based on the vapor pressure differential at microporous hydrophobic membrane surface, which acts as the driving force for biofuels separation (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). For the membrane evaporation process, non-porous membrane is usually used for biofuels selectively recovery from hydrolysate via partial vaporization based on the solution–diffusion model (Trinh et al. 2019). Pervaporation membrane can be roughly classified into hydrophilic membrane and hydrophobic membrane, for which the former is mainly used to remove water from the mixed solution while the latter is mainly used to extract biofuels from the liquid stream (Huang et al. 2008). Therefore, the hydrophobic membrane is more energy efficient for the case with low product concentration, especially for the bioethanol recovery with concentration less than 10% (in w/w). In recent years, the hybrid processes integrating various unit operations together have attracted attentions for continuous biofuel production. For example, the hybrid fermentation–pervaporation process can remove the produced bioethanol in situ to offset product inhibition and avoid microbial 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 (Novita et al. 2018).

Membrane applications for gaseous biofuel production

Gaseous biofuels, like biohydrogen and biogas, are also important renewable energy types which have been widely used in practical. For example, the biogas digester is commonly constructed in medium or small size for household cases attributing to simple configuration and low investment (Chen et al. 2017; Xiao et al. 2020). The membrane bioreactor with sophisticated structure is not suitable for biogas production attributing to high investment but is frequently used in biohydrogen production. Biohydrogen production is a technology to produce hydrogen gas with microorganisms, which can be roughly classified into three types: (a) photo-dependent biohydrogen production via photolysis of water by algae and cyanobacteria, (b) photo-fermentation by decomposing organic matters with photosynthetic bacteria, and (c) dark fermentation for hydrogen production with facultative or obligate anaerobic bacteria (Trchounian et al. 2017; Srivastava et al. 2020). Among many emerging approaches for biohydrogen production, membrane integrated biohydrogen production system is for sure a promising technology allowing for dealing with various kinetic inhibitions, like biomass washout, substrate or product inhibition, as shown in Table 3.

Membranes 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) within 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). Biohydrogen production via photolysis is regarded as the cleanest way for hydrogen production because of high-efficiency carbon mitigation and quick solar energy conversion efficiency, but its application is severely inhibited by low hydrogen productivity, oxygen inhibition and strict light requirement (Argun and Kargi 2011).

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. 2018), while 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 optimize light distribution and light conversion efficiency, researchers have proposed many approaches like optical fibers (Zhong et al. 2019), light guide plates (Fu et al. 2017) and cell immobilization techniques (Tian et al. 2010), which improved biohydrogen productivity to some extent.

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. Since some membranes have ability to selectively separate gas and liquid components as well as regulate mass and heat transfer, membrane-integrated photobioreactors for biohydrogen production are expectable to enhance photo-biohydrogen production.

Membranes for dark fermentative biohydrogen production

Comparing with biohydrogen production via photolysis or photo-fermentation, dark fermentative biohydrogen production occupies higher proportion nowadays. Since light is unnecessary for dark fermentation, reactors' design is more flexible and the volume utilization of the bioreactors can be fully exploited. In addition, since oxygen inhibition is no

Table 3 Major application of membranes in gaseous biofuel production

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
Product 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; Lundin et al. 2017) Key criteria for the membrane: (1) permeability and (2) selectivity

longer a problem under anaerobic conditions, dark fermentative biohydrogen production shows more reliable and faster hydrogen production rate.

For conventional dark fermentation process, low biomass density in continuous stirred tank reactor caused by high biomass washout rate and by-products or product inhibitions is crucial shortcomings for hydrogen output (Kariyama et al. 2018). The introduction of membrane modules in anaerobic membrane bioreactor can effectively enhance the conversion efficiency from raw material to biohydrogen by ensuring high solid retention time and selectively removing inhibitors (Shin and Bae 2018). Besides, the mass transfer of metabolites in the system can be regulated with membrane for highly efficient biohydrogen production (Park et al. 2017). For example, 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 submerged membrane bioreactor and side stream membrane bioreactor (as shown in Fig. 2). Membrane modules are always submerged in the liquid of the reactor for the submerged membrane bioreactor, while they are set outside of the reactor for the side stream bioreactor. The submerged membrane bioreactor is typically characterized by low energy cost but high membrane utilization and high complexity on membrane washing than the side stream membrane bioreactor. To solve the shortcomings of these two types of membrane bioreactor, many derived membrane bioreactors were recently 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), which successfully retained bacterial consortia and resulted 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.

Membranes for biohydrogen purification

Another important role of membrane in biohydrogen production system is purification of the gaseous products to obtain high-quality hydrogen fuel. To eliminate effect of by-products (CO_2, CO, SO_x) on combustion property of biohydrogen, membrane technologies for biohydrogen purification are a feasible approach because it avoids chemical conversion of the mixed gas (Zhang et al. 2020).

The membrane used for biohydrogen purification is usually a semi-permeable separator acting as a selective mass transfer barrier to separate various compositions (Bakonyi et al. 2018). Superior permeability and selectivity are two key criteria for the membrane applied in gas purification, but it is unfortunately 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 to enhance the gas separation characteristics of membranes for

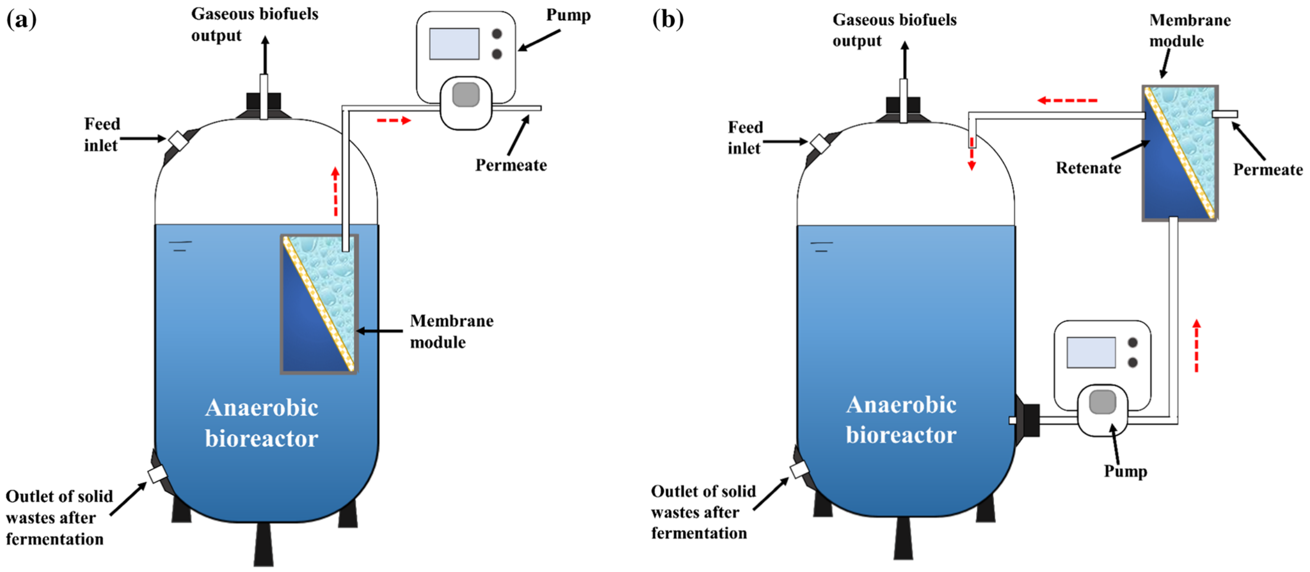
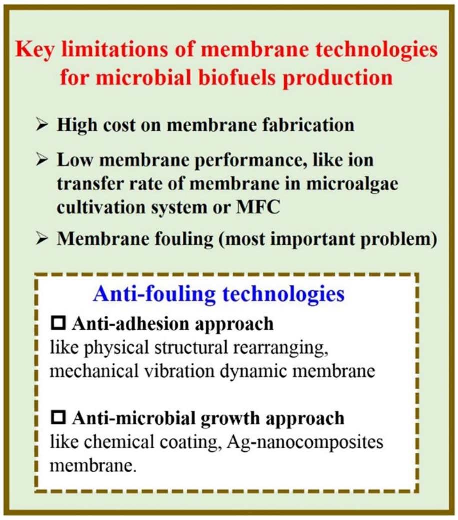


Fig. 2 Configurations of **a** the submerged membrane bioreactor (MBR) and **b** the side stream MBR for gaseous biofuels production

biohydrogen purification. Ahmad et al. (2016) constructed a nearly superhydrophobic and microporous membrane by blending amorphous poly-benzimidazole and semi-crystalline 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 adsorbents 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 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%.

Among various influencing factors for the cost of membrane bioreactor, membrane fouling is one of the most important problems, as shown in Fig. 3 (Buitrón et al. 2019). During microbial growth, soluble microbial products and extracellular polymeric substances consisting of complex biopolymer mixtures like proteins, polysaccharides, lipopolysaccharides and lipoproteins are produced in the cultures, which triggered biomass flocs formation and accumulation on membrane surface, resulting in membrane pore blocking and fouling (Shan et al. 2018). Membrane modification with physical structural rearranging, chemical coating and functional material embedding is considered as promising approaches for anti-fouling membrane development (López-Cázares et al. 2018; Qin et al. 2018; Shan et al. 2018). Schematic of some typical membrane modification methods for anti-fouling technology is shown in Fig. 4, like physical structural modification with nano-Ag cluster (Fig. 4a) and chemical solvents coating on membrane (Fig. 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 anti-fouling performances of the membrane.

Another important anti-fouling approach is dynamic membrane (like rotating membrane in Fig. 4c) using a physical barrier to prevent biomass accumulation on membrane surface (Yang et al. 2018). Comparing with the anti-fouling method of air bubbling, the dynamic membranes provide stronger shear force on the membrane by mechanical vibration, like rotation, vibration and oscillation (Bagheri and Mirbagheri 2018; Qin et al. 2018). The structure and function of dynamic membrane systems are also continually upgraded by researchers. 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 in reducing membrane fouling and is promising with respect to energy savings. These emerging anti-fouling technologies provide great potential to reduce membrane manufacturing and operating costs, which then enhance the commercial feasibility of biohydrogen application.



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. 3 Key limitations of membrane application in microbial biofuel production process (Buitrón et al. 2019)

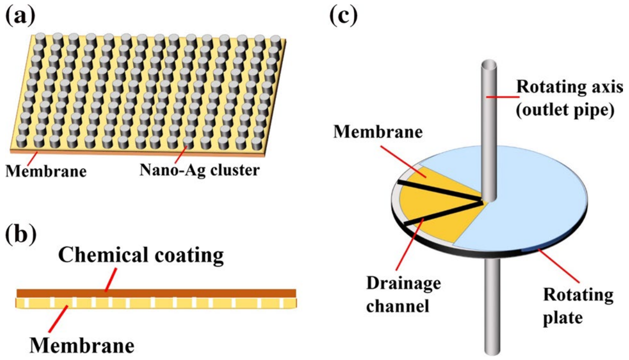


Fig. 4 Typical anti-fouling membrane system **a** membrane surface modification with nano-Ag cluster, **b** chemical coating of membrane and **c** dynamic membrane system with rotating unit (Qin et al. 2018)

Membrane applications in microbial fuel cells

Microbial fuel cell (MFC) as a bioelectrochemical device has attracted particular interests in environmental and energy fields (Liu et al. 2019). It uses microorganism as electrocatalysts to conduct oxidation–reduction reaction and convert chemical energy in wastewater into electrical energy (Leong et al. 2013; Wen et al. 2019). MFC can be mainly classified into single chamber and dual chamber MFC (Fig. 5) according to the structure. The dual chamber MFC contains an anode and a cathode chamber separated by a membrane as electrolyte bridge, while only anode chamber was included in the single chamber MFC. Membrane is an important component of the MFC, which physically divides but chemically and ionically connects the anode and cathode chamber, significantly influencing overall performance of the MFC. The possible membrane types used for the MFC include cation exchange membrane (Daud et al. 2018), anion exchange membrane (Elangovan and Dharmalingam 2017), porous membrane (Li et al. 2015), polymer/composite membrane (Ahilan et al. 2018; Filiz 2017), etc. Each type of membrane has its advantages and disadvantages. For example, cation exchange membrane can enhance columbic efficiency of the MFC since they directly conduct H^+ transfer from anode to cathode, but pH splitting of the MFC with cation exchange membrane is easily happened (Chaudhuri and Lovley 2003). The anion exchange membrane can diminish pH splitting of the MFC, but substrate crossover is a major drawback (Varcoe et al. 2014). Though low internal resistance of porous membrane is beneficial for H^+ transfer of the MFC, high crossover rate of oxygen and substrate through the membrane pores is detrimental to MFC performance (Slate et al. 2019). Until now, an ideal membrane taking all aspects into account is yet to be developed.

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. (Leong et al. 2013). The membrane with high resistance is not conducive to proton transfer due to low ion exchange capacity and reduce MFC power output. But low resistance membrane with porosity like microfiltration membrane can also reduce the power density attributing to high crossover rate of oxygen and substrate (Slate et al. 2019). Therefore, the membrane with low internal resistance and low oxygen and substrate crossover rate is an ideal type for 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 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 membrane are also important problems for the MFC. Oxygen could compete with the anode for electrons since oxygen is more favorable electron accepter, and substrate transfer could lead to an internal short circuit inside the MFC and reduce columbic efficiency (Kim et al. 2013). An important role of membrane in the MFC is to prevent oxygen and substrate crossover. It is reported that the coulombic efficiency of the MFC with membrane was 20% higher than the membrane-less one (Slate et al. 2019). Unfortunately, a membrane that can totally avoid oxygen and substrate diffusion is not developed, and some auxiliary approaches to minimize negative effects of oxygen and substrate were proposed. Ahilan et al. (2018) modified ceramic membrane with montmorillonite-$H_3PMo_{12}O_{40}/SiO_2$ composite to reduce the oxygen mass

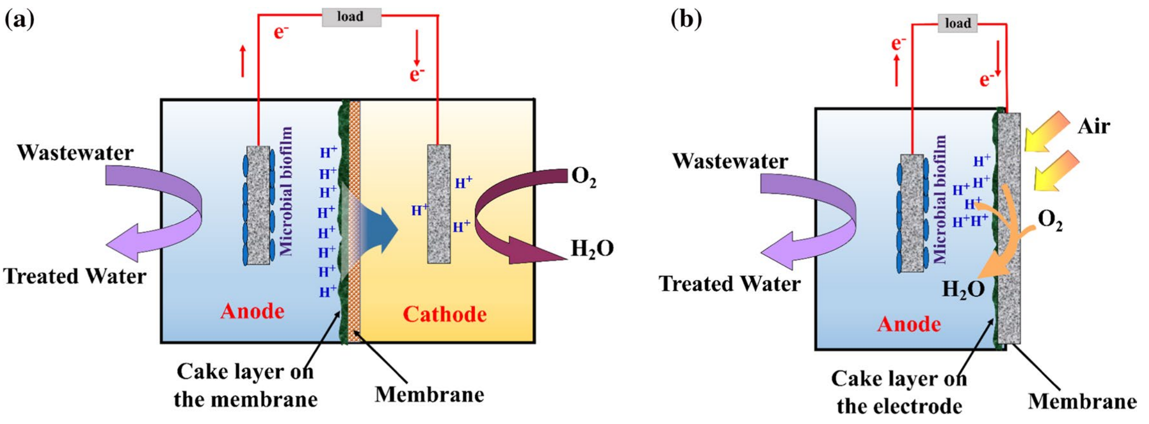


Fig. 5 Schematic diagram of **a** the dual chamber microbial fuel cell (MFC) and **b** the single chamber MFC

transfer coefficient to 5.62×10^{-4} cm/s, which is near the commercial 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, which reduced the oxygen diffusion coefficient to one-sixth of the blank ceramic membrane. Besides electrons competition and internal short circuit, oxygen and substrate diffusion can also induce biofouling of the membrane and pH splitting of the MFC. On the one hand, the diffused substrate provides food for the microbial biofilm, and diffused oxygen triggered biofilm formation of aerobic bacterial. On the other hand, the formed microbial biofilm acts as barrier for proton transfer and deteriorates pH splitting of the MFC. To ensure high performance of the MFC, the fouled membrane must be replaced with new one for proton diffuse, but this dramatically improved operating investment of the MFC. In recent years, researchers proposed some approaches to reduce membrane biofouling, like anti-microbial approach and anti-adhesion approach (Chatterjee and Ghangrekar 2014; Sun et al. 2016; Yang et al. 2016). Chatterjee and Ghangrekar (2014) constructed an anti-fouling MFC using vanillin as biocide. Yang et al. (2016) coated the membrane with a silver nanoparticle polydopamine to mitigate biofouling by taking advantage of anti-microbial effect of nano-Ag particle. Sun et al. (2016) used well-ordered multi-walled carbon nanotubes and its derivative modified with the carboxyl modified to prevent microbial adhesion. However, effectiveness of these anti-fouling 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 (Gajda et al. 2018).

Conclusion

Microbial energy conversion technology is a potential method for simultaneous realization of environmental remediation and energy production. Membranes play very important roles on enhancement of bioenergy productivity and quality. This paper presents a review on roles and mechanisms of membrane on bioenergy conversion processes and discussed important factors influencing the overall performances. For liquid biofuel 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 and purify the produced biogas for high-quality fuel generation. For the microbial fuel cell, membrane can decrease internal short circuit and increase power density by acting as physical barrier and electrolyte bridge. But biofouling of membrane caused by microbial attachment is vital problem that needs to be addressed. Anti-fouling technologies, like anti-adhesion approach or anti-microbial growth approach, are discussed in the work. For future prospects, anti-fouling technology of membranes is still 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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