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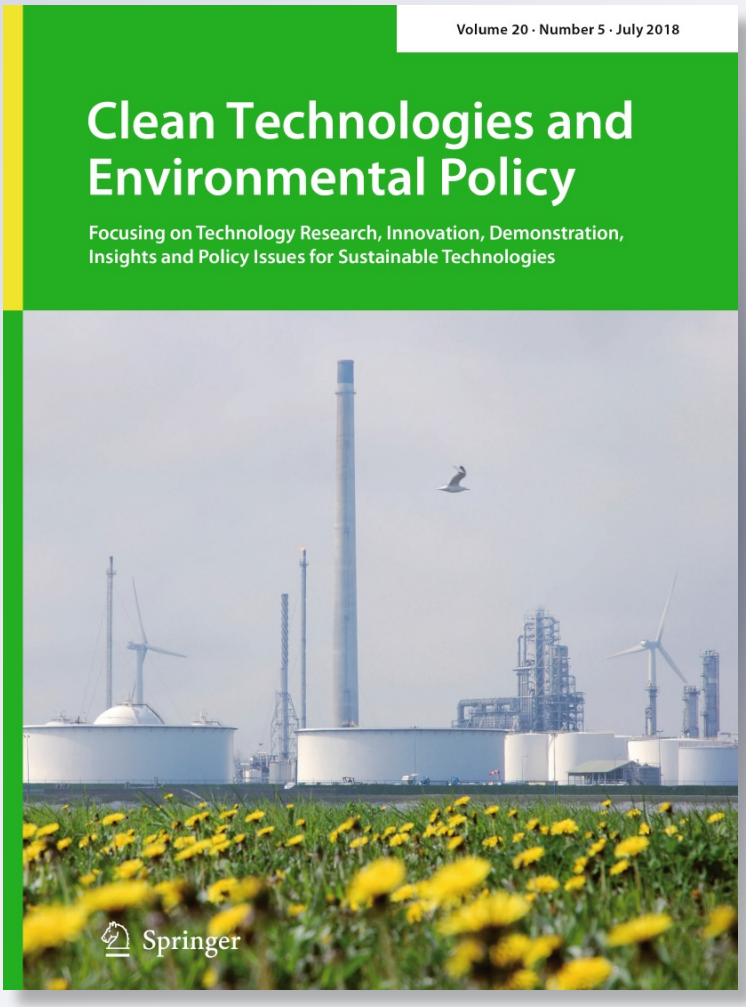

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REVIEW PAPER

Integrating bioelectrochemical systems for sustainable wastewater treatment

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Abstract

Current wastewater treatment processes such as activated sludge process and other aeration technologies are resource-consuming and are unsustainable. Novel and integrated processes are crucial to the development of sustainable wastewater treatment systems. In this context, anaerobic treatment technologies provide numerous opportunities for minimization of energy and resource consumption and maximization of beneficial products. Further, integration of anaerobic digestion augmented by co-digestion, fermentation, dark fermentation or photo-fermentation and other bioelectrochemical systems may result in resource-efficient waste management and environmental protection. This mini-review discusses various possibilities and highlights recent developments of integrated aerobic and anaerobic technologies with bioelectrochemical systems for sustainable wastewater treatment.

Keywords Anaerobic digestion · Bioelectrochemical systems · Biorefineries · Clean technologies · Fermentation · Microbial fuel cells · Sustainability

Introduction

Wastewater treatment is an absolute necessity to protect our environment (Gude 2016). Current wastewater treatment technologies are reliable from treatment perspective but still contribute to greenhouse gas emissions and other emerging concerns of water quality (Gude 2016; Arana and Gude 2018). Wastewater treatment processes can be broadly categorized into aerobic and anaerobic processes, application of which depends on the characteristics and suitability of the wastewater sources (Gude 2015a). Among the aerobic processes (activated sludge process, AS and trickling filter, TF), activated sludge process is the most robust and reliable technology dominating most of the technologies in the field. Anaerobic treatment processes (such as waste stabilization ponds, WSP and upflow anaerobic sludge blanket, UASB) are also desirable in specific applications where high-strength wastewaters are generated which are not suitable for aerobic treatment. A comparison of aerobic and anaerobic treatment technologies for wastewater treatment is shown in Table 1. As the global concerns for energy and nutrient sources become adverse in the recent years, there is a conscientious effort among the designers, planners, researchers and industrialists to develop resource-efficient technologies for sustainable wastewater treatment (Gude 2015b).

Wastewater is a concern and a challenge while at the same time it provides numerous opportunities for achieving a pathway toward sustainability. A preliminary examination of currently well-established technologies and emerging process configurations brings forth the anaerobic digestion (AD), fermentation and dark fermentation (DF) processes and bioelectrochemical systems (BES) such as microbial fuel cells (MFCs), microbial electrolysis cells (MEC) and other biorefinery configurations as promising technologies for developing sustainable wastewater treatment systems that are chemical-, energy-, and cost-efficient. Moreover, integration of these processes could be beneficial from treatment and resource recovery perspectives. Integration can be considered from energy recovery, resource (water and other chemicals) recovery and final water quality-based objectives. This may include integration of aerobic and anaerobic technologies or biochemical and bioelectrochemical operating principles. This mini-review article will discuss some of these promising technologies and integrated

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Table 1 Typical features of treatment technologies for domestic wastewater (rating: ++… excellent, +… positive, − … negative). Reproduced with permission from Haandel and Lettinga (1994), Mara and Pearson (1998), Metcalf and Eddy (2014), von Sperling et al. (2002) and Wett and Buchauer (2003)

	UASB		WSP		TF	AS
Environmental conditions	Anaerobic	Anaerobic	Facultative	Maturation	Aerobic	Aerobic
Suited for raw sewage	+	++	++	–	+	++
Suited for settled sewage	++	++	++	–	++	++
Suited for wastewater temperatures (°C)	> 20	> 5	> 5	> 5	> 5	> 5
BOD removal efficiency (%)	> 70	> 50	> 70	> 50	80–90	> 90
Nutrient (N, P) removal efficiency	–	–	–	–	+ (++)	+ (++)
Coliform removal (%)	90	90	90–99	> 99	90–95	90–98
Typical HRT	~ 6 h	> 1 d	> 4 d	> 3 d	~ 6 h	~ 15 h
Odor nuisance	++	+	+	++	++	++
Energy demand and gas production	++	++	++	++	+ (++)	–
Land requirement	++	+	–	–	+	+
Investment cost	++	++	+	+	+	–

process applications for energy-neutral or energy-positive and resource-efficient sustainable wastewater treatment.

Energy producing wastewater treatment processes

Anaerobic digestion

Microbial degradation and stabilization of organic matter under anaerobic conditions produces biogas which is a mixture of carbon dioxide and methane (Gude 2015b). Hydrolysis, acidogenesis, acetogenesis and methanogenesis are the main steps that result in the end products of methane and carbon dioxide. AD is widely used across many agricultural, food, industrial and municipal sectors for treating a variety of wastewater sources due to its advantages such as low biomass yields and biogas production. However, poor operational stability still prevents AD from being widely commercialized (Dupla et al. 2004). In AD, microorganisms responsible for acid and methane formation and their composition vary significantly. This is mainly influenced by many factors such as physiological conditions, nutrient requirements, rate of growth and other environmental stressors (Dupla et al. 2004). An optimum balance among the microbial composition is essential for stable operation of the digestion process. Failures related to AD process are mainly caused by the inhibitory effects by the chemical products and those toxic compounds present in the wastewater sources. The inhibitory substances and conditions can be listed as: high concentrations of ammonia, pH, temperature, the presence of ions such as Na^+, K^+, Ca^{2+} and Mg^{2+}, heavy metals, sulfides, competition between the anaerobes (hydrolytic, acidogenic bacteria, acetogens, hydrogenotrophic and aceticlastic methanogens) and sulfate-reducing bacteria, organic substances such as chlorophenols, halogenated aliphatics, lignins and lignin-related compounds. For more details, refer to other contributions (Gunaseelan 1997; Chen et al. 2008).

Fermentation

Fermentation is a process of chemical breakdown of organic substrates by bacteria, yeasts or other microorganisms, typically involving release of heat. Fermentation products such as mixed liquors, carbon dioxide, hydrogen and methane have been found to provide many beneficial uses. Hydrogen production via fermentation has been investigated due to its high energy density. Anaerobic or facultative bacteria can perform fermentation as a result of metabolic pathways (De Gioannis et al. 2013). Fermentation products can be monitored or optimized by controlling the operating conditions which establish suitable metabolic pathways (Singh et al. 2015). In this manner, volatile fatty acids (VFAs) and alcohols production including acetate, propionate, butyrate, lactate and ethanol can be tailored to the specific needs. An understanding of various metabolic pathways involved in fermentation process is important. For instance, fermentation of carbohydrates involving acetate and butyrate production pathways will release 4 and 2 mol of H_2 mol^{-1} of glucose, respectively. It should be noted that propionate, ethanol and lactic acid may also be produced in the same mixed microbial consortium which may inhibit H_2 production. Because propionate is a metabolite of a H_2-consuming pathway, while ethanol and lactic acid are associated with zero-H_2 pathways (Guo et al. 2010). The current challenge in this research and process development is to accomplish optimum hydrogen production with low operating costs and yet

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produce an acceptable quality effluent for further treatment. To achieve this, operational parameters including temperature, pH, reactor configuration, substrate concentration and organic loading rate should be studied for their effect and interdependence on the optimization of process for hydrogen production.

Dark fermentation

Organic substrates originating from various sources both simple and complex carbohydrates can be converted to hydrogen gas and fatty acids using anaerobic or facultative bacteria including mixed anaerobic consortia (Zong et al. 2009). Due to formation of fatty acids, the maximum theoretical hydrogen yield in DF process is relatively low which depends on the composition of fatty acids produced (Li and Fang 2007). Yet, DF is considered the most commercially feasible method for hydrogen production from cellulose, due to a relatively high hydrogen production rate (Wang et al. 2011). Cellulose is first hydrolyzed to hexoses (similar to organic matter in wastewater), which can then be fermented to hydrogen and acetate according to (Wang et al. 2011)

$$C_6H_{12}O_6 + 3H_2O \rightarrow CH_3COO^- + CH_3CH_2OH + 3H^+ + 2H_2 + 2HCO_3^- \quad \left(\Delta G^\circ = -206.3 \frac{kJ}{mol}\right)$$

For organics in wastewater, assuming an organic substance such as glucose, other possible reactions are:

$$C_6H_{12}O_6 + 12H_2O \rightarrow 6HCO_3 + 6H^+ + 12H_2 \quad \left(\Delta G^\circ = 241 \frac{kJ}{mol}\right)$$

$$C_6H_{12}O_6 + 4H_2O \rightarrow 2CH_3COO^- + 2HCO_3 + 4H^+ + 4H_2 \quad \left(\Delta G^\circ = -48 \frac{kJ}{mol}\right)$$

$$C_6H_{12}O_6 + 2H_2O \rightarrow CH_3CH_2CH_2COO^- + 2HCO_3 + 3H^+ + 2H_2 \quad \left(\Delta G^\circ = -137 \frac{kJ}{mol}\right)$$

Photo-fermentation

This process employs a diverse group of photosynthetic bacteria that use sun light as energy to convert organic compounds into hydrogen and CO_2 (Azwar et al. 2014). For instance, photo-fermentation with purple non-sulfur (PNS) bacteria can be used to convert fatty acids into hydrogen and other gases including small molecule compounds under anoxic or anaerobic conditions with sunlight. Photo-hydrogen production was performed mainly through four species of PNS bacteria. Other bacteria that were found in photo-fermentation process are *Rhodobacter sphaeroides*, *Rhodopseudomonas palustris*, *Rhodobacter capsulatus* and *Rhodospirillum rubrum* (Sasikala et al. 1991; Barbosa et al. 2001; Hillmer and Gest 1977; Miyake et al. 1982). Small chain organic acids like acetate, lactate and butyrates are used as carbon source, while light is used as energy source to produce hydrogen (Uyar et al. 2009). As a comparison, DF takes place in the absence of light with a series of biochemical reactions under anaerobic conditions.

The need for photosynthetic fermentation is realized due to the fact that the produced volatile fatty acids in the effluent of DF may carry a potential threat to the environment. In an effort to eliminate this problem, further decomposition of the fatty acids and reduction of chemical oxygen demand (COD) in the fermentation effluent is required. Converting the fatty acids to methane is another feasible option. Several studies also focused on the two-step hydrogen production process of sequential dark- and photo-fermentation which achieved higher hydrogen yields from various substrates compared to hydrogen yields by DF or photo-fermentation alone (Tao et al. 2007). For example, hydrogen yield from sucrose increased by 80% in a two-step process, from 3.67 mol H_2 mol^{-1} sucrose in DF to 6.63 mol H_2 mol^{-1} sucrose in a two-step process (Zong et al. 2009).

Bioelectrochemical systems

The two common BES are microbial fuel cells and microbial electrolysis cells. A microbial fuel cell is a galvanic cell in which exoelectrogenic (electron generating) electrochemical reactions take place in the anode chamber which flow through a conducting circuit and are received by an electron acceptor in the cathode chamber (Gude 2016; Schröder 2007). The standard free energy can be expressed in a standard cell voltage term or electromotive force, emf as E. The theoretical cell voltage or electromotive force (emf) of the overall reaction (the difference between the anode and cathode potential) determines if the system is capable of electricity generation as shown below (Schröder 2007).

$$\Delta E^\circ_{Cell} = \Delta E^\circ_{Cathode} - \Delta E^\circ_{Anode}$$

A negative free reaction energy leads to a positive standard cell voltage. This distinguishes a galvanic cell from an electrolysis cell, as the latter, associated with a positive

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free reaction energy and thus with a negative cell voltage, requires the input of electric energy. The standard cell voltage can also be obtained from the biological standard redox potentials of the respective redox couples, as shown below. In MFCs, the Gibbs free energy of the reaction is negative. Therefore, the emf is positive, indicating the potential for spontaneous electricity generation from the reaction. For example, if acetate is used as an organic substrate ($[CH_3COO^-] = [HCO_3] = 10$ mM, pH 7, $T = 298.15$ K, $pO_2 - 0.2$ bar), with oxygen reduction, the combined redox reaction would be (Rozendal et al. 2008) as shown in oxidation and reduction reactions below (Rozendal et al. 2008):

Anode:

$$CH_3COO^- + 4H_2O \rightarrow 2HCO_3 + 9H^+ + 8e^-$$

($E^\circ = -0.289$ V vs. SHE)

Cathode:

$$2O_2 + 8H^+ + 8e^- \rightarrow 4H_2O \quad (E^\circ = 0.805 \text{ V vs. SHE})$$

Overall:

$$CH_3COO^- + 2O_2 \rightarrow 2HCO_3^- + H^+ \quad (E^\circ = 1.094 \text{ V vs. SHE})$$

BES can be used as a suitable technology in applications of clean electricity production, waste remediation, resource recovery and valuable chemical production under different process configurations (microbial electrolysis cells) and process conditions (microbial electrosynthesis). For example, BES can be designed to utilize solar energy for excess biomass and electricity production through plants and photosynthetic microorganisms such as microalgae (Bajracharya et al. 2016).

Integrated processes

Anaerobic systems such as AD usually discharge effluents which are enriched with high concentrations of organic contaminants and nutrient. The soluble components of these contaminants include several volatile fatty acids (VFAs), which are odorous and increase biological oxygen demand in the receiving environment. Post-processing to remove these contaminants could be coupled with energy producing technologies. AD-integrated BES can be envisioned at the domestic levels, especially in decentralized wastewater treatment systems such as septic tanks and cess pools. Many centralized systems also include anaerobic digesters for biomass/excess sludge management which provide an ideal opportunity for various applications. Microbial and microalgae-based wastewater treatment systems are also receiving increasing attention due to the multiple benefits they provide (Otondo et al. 2018; Blair et al. 2014; Kokabian and Gude 2013). These systems can provide additional microalgae biomass which could be processed in anaerobic digesters and the effluent from which could be fed to bioelectrochemical systems (Gude et al. 2013; Kokabian and Gude 2015; Kokabian et al. 2018a, b, c). The following sections discuss the various possibilities for integrating AD and bioelectrochemical systems.

Septic tanks

Septic tanks and other decentralized wastewater treatment systems are ideal for remote, under-privileged and developing communities (Yazdi et al. 2015). Current septic systems design does not include an energy recovery option. However, they can be designed to produce bioelectricity which could be a valuable source for these communities. Such configurations are receiving increasing interest in recent years as the need for energy recovery and sanitation is increasing around the world. Yazdi et al. (2015) developed a pluggable MFC-septic tank configuration in which several MFC units can be included in series or parallel settings. A maximum power density of 142 ± 6.71 mW m^{-2} was reported in a laboratory study when 3 MFC units were connected in parallel which was sufficient to power a LED bulb of 6 W capacity for 4 h in a day. Septic tanks can be designed to produce methane both by AD and by electromethanogenesis process as shown in Fig. 1a (Zamalloa et al. 2013).

Stacked MFC configuration is usually considered to increase the power production in a septic tank system. A septic tank of 2.93 m^3 with a useful volume of 2.44 m^3 was integrated with stacked MFCs which consisted of 15 individual cells distributed in the second chamber (Alzate-Gaviria et al. 2016). Under a continuous flow mode, 15 cartridges of MFCs were installed in a septic tank. Three MFCs were chosen to further tests which produced 109.40 ± 34.25, 131.58 ± 27.75 and 124.01 ± 27.57 mW m^{-3}, respectively, when fed an organic loading of rate of 200, 500 and 1000 ppm of COD, respectively. Total COD removal and total coulombic efficiency were 89.67 ± 5.19 and $48.07 \pm 2.33\%$. It was reported that the internal resistance of the unit was mainly due to higher anode resistances due to electron transfer losses.

In developing communities, latrines can be designed to integrate the microbial fuel cell technology. A three-chamber MFC unit was demonstrated around the latrine system in Ghana (Castro 2014). The system consisted of membraneless anode and biocathode chamber system separated by a middle chamber which provided for nitrification of ammonia in the anode effluent (see Fig. 1b). Anode compartment essentially removed carbonaceous compounds, while the biocathode chamber was designed to provide for denitrification process, thus providing a complete nitrogen removal system along with electricity production. Nitrogen and organic matter removal was observed during various operational conditions in Phase I before the MFC began treating

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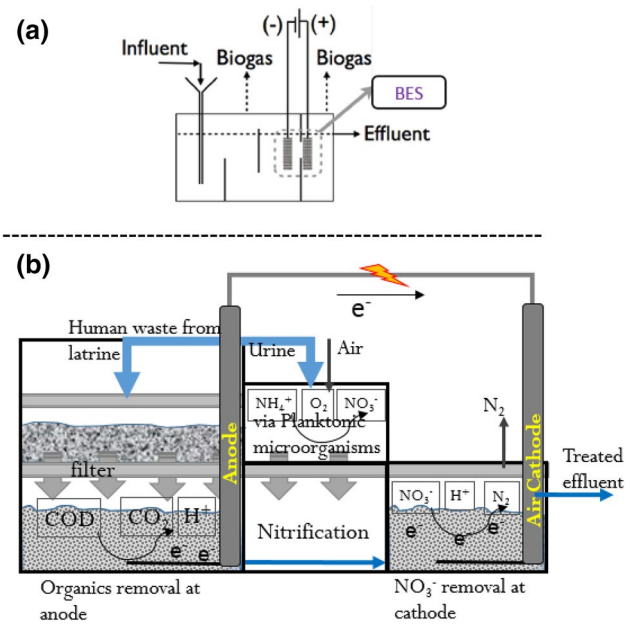

Fig. 1 **a** A septic integrated with a bioelectrochemical system, BES (a microbial fuel cell system could produce electricity, whereas a microbial electrolysis system produces additional biogas); **b** A latrine-MFC (in Agona, Nyakrom, Ghana) design consisting of an anode and a biocathode chamber for simultaneous carbon and nitrogen removal from human wastes (after Castro 2014)

synthetic feces and urine solutions during Phase II. During all of the operational conditions, COD removal was greater than 90%. Nitrate removal in Phase I reached up to 76.8 ± 7.1%, while nitrogen removal during phase II was 68.4 ± 2.8 mg NL^{-1}. Power production reached an average 3.40 ± 0.01 nW m^{-2} during the Phase I and decreased to 0.66 ± 0.02 nW m^{-2} in Phase II indicating the potential evidence of AD occurring in the anode, which limited power production by anode respiring bacteria.

Urine-fed MFCs were also investigated for electricity production. An estimated annual global production of ~ 6.4 trillion liters (based on a world population of 6.97 billion and average daily urine production of 2.5 L/adult human) of urine may provide an opportunity for potential energy recovery embedded in the form of nutrients (Ieropoulos et al. 2012). Field-scale demonstrations such as use in refugee camps and other outdoor events like music festivals were reported recently including pilot-scale studies at wastewater treatment plants (Ieropoulos et al. 2012; Winfield et al. 2012; Heidrich et al. 2014; Martinucci et al. 2015). Similarly, human feces could be used as a substrate (Fangzhou et al. 2011). A two-chamber MFC achieved removal efficiencies for total chemical oxygen demand, soluble chemical oxygen demand, and NH_4^+ reached 71, 88 and 44%, respectively, over 190 h treatment time producing a maximum power density of 70.8 mW m^{-2}.

Integration with wastewater treatment processes

MFCs can be conveniently integrated within existing centralized wastewater treatment systems (see Fig. 2a). As reported earlier, the configurations could vary depending on the influent wastewater characteristics (Pham et al. 2006). One of the options, especially for high-strength (COD) wastewaters, would be to directly feed the anaerobic digester with the influent followed by treatment of low COD products in the AD effluent in MFCs prior to supplying the effluent to the conventional aeration-based wastewater treatment systems. The other option would be to separate the suspended COD matter and other particulates to feed AD and MFC units followed by treatment in conventional wastewater treatment systems. In both processes, energy recovery could be enhanced due to MFC integration. Figure 4 shows an integrated configuration including AD and MFC/MEC units in a conventional wastewater treatment scheme. Other options for integrating BES with AD for solid waste treatment include (Cheng and Kaksonen 2017): (1) BES as a separate downstream process to convert AD effluent into electricity; (2) BES as a biosensor to monitor AD process stability; (3) BES to improve digestibility of AD substrate; (4) Direct integration of BES in an AD reactor to facilitate in situ electromethanogenesis; (5) BES as a separate system to improve AD biogas quality and yield; and (6) BES as an add-on unit for toxicity removal and resource recovery from AD processes.

Integration with agricultural and other industrial wastewaters

Finally, anaerobic digesters in various applications including agricultural wastes, dairy and food industry, and other industrial wastes can produce effluents suitable for anolyte use in MFCs (See Fig. 2b). Integration with anaerobic digesters may enhance energy recovery benefits as well as the overall process economics. Because many of these industries are remote, in situ linkage of MFCs with digesters can prove to be an ideal combination (Li et al. 2015). Table 2 provides a summary of few studies utilizing agricultural, farm and industrial wastewaters for bioelectricity production (Schievano et al. 2016; Zuo et al. 2015; Min et al. 2005; Sciarria et al. 2013, 2015; Fradler et al. 2014). Pandey et al. (2016) provided a critical summary of numerous substrates, both of fermentable and non-fermentable nature, used in microbial fuel cells.

AD of microalgae biomass

Microalgae-based wastewater treatment systems have become a promising alternative for conventional energy

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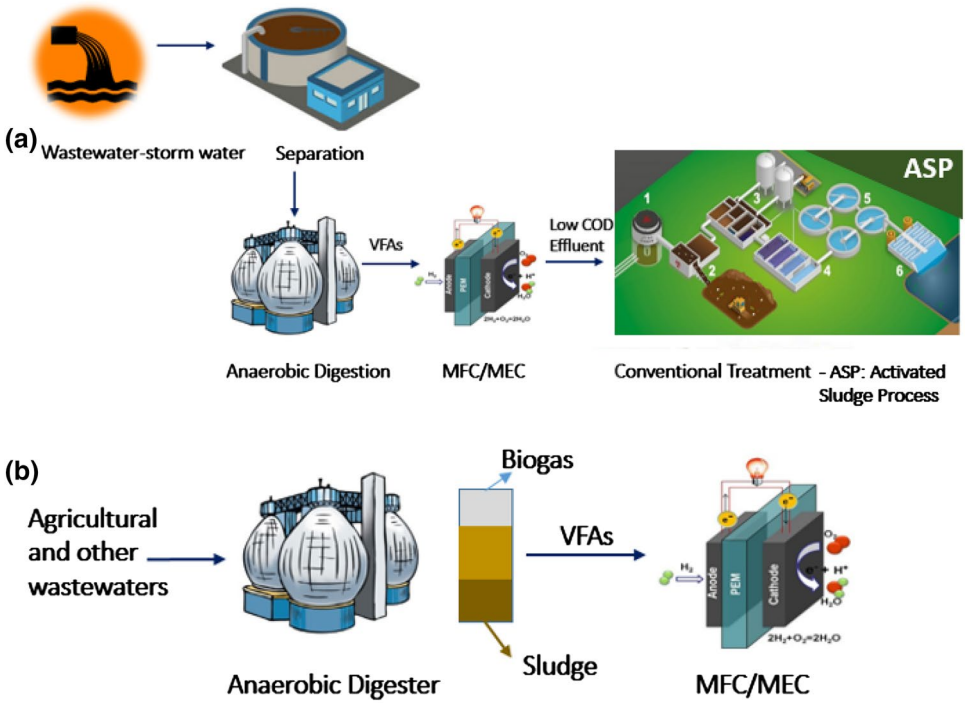

Fig. 2 **a** Integrated configuration including AD and MFC/MEC units in a conventional wastewater treatment scheme—1. Wastewater pumping station, 2. Grit removal, 3. Primary sedimentation, 4. Aeration tanks, 5. Secondary clarifiers, 6. Disinfection and pH/DO adjustment before discharge; **b** MFC integrated with an anaerobic digester as a post-treatment technology

Table 2 Current generation, voltage potential and energetic yields in MFC reactors

Type of MFC reactor	Voltage mV (Ω)	Volumetric power density (reactor volume) W m^{-3}	Energy recovery from input wastewater Wh L^{-1}	Coulombic efficiency (%)	COD removal gCOD L^{-1} (%)	References
Brewery WW Fed-batch	628 (500)	5.1	–	10	87	Schievano et al. (2016)
Corn stover Fed-batch	1446 (250)			19–29	60–70	Schievano et al. (2016)
Swine WW Fed-batch	357 (1 k)			8	27	Zuo et al. (2015)
Swine WW Fed-batch		–	–	26	86	Zuo et al. (2015)
Food/dairy WW Fed-batch	600 (1 k)	–	–	2.5	77	Min et al. (2005)
(OMW + DW) Fed-batch	380 (1 k)	5.17	1.0 ± 0.14	29	60	Sciarria et al. (2015)
Red wine lees Fed-batch	340 (1 k)	3.1	0.45 ± 0.1	9	27	Sciarria et al. (2013)
White wine lees Fed-batch	420 (1 k)	8.2	1.3 ± 0.3	15	90	Sciarria et al. (2013)
Two-stage digestate Continuous 4-modules tubular	500 (1 k)	2.3–3.4	0.093	60	4.4	Fradler et al. (2014)
Two-stage digestate Fed-batch	530 (1 k)	13.3	0.369 ± 0.03	21	21.7	Schievano et al. (2016)
One-stage digestate Fed-batch	520 (1 k)	12.1	0.271 ± 0.02	19	17.6	Schievano et al. (2016)

intensive aeration systems. An energy-positive treatment scheme is possible with this configuration as discussed in many previous reports (Gude 2015a, b). In microalgae-based wastewater systems, the microbial and microalgae communities can function in a synergistic effect to produce high lipid containing biomass suitable for biofuel production which includes biogas, biodiesel and other biofuel products (see Fig. 3) (Blair et al. 2014; Gude 2015b). Heterotrophic (bacteria) and photoautotrophic (microalgae) microorganisms can be used purposely for carbon and nutrient removal,

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respectively, in an integrated wastewater system. The mixed biomass produced from the system can be fed as organic source to a microbial fuel cell system. Electricity generation potential and biodegradation capacities could vary depending on the microalgae species. For example, when *Chlorella vulgaris* and *Ulva lactuca* were fed in two separate microbial fuel cells, *C. vulgaris* produced more energy per substrate mass (2.5 kWh kg^{-1}), but *U. lactuca* was degraded more completely over a batch cycle (73% COD) (Velasquez-Orta et al. 2009). Maximum power densities of 0.98 W m^{-2} (277 W m^{-3}) and 0.76 W m^{-2} (215 W m^{-3}) were reported for *C. vulgaris* and *U. lactuca*, respectively. Co-digestion is another promising alternative for enhancing biogas production. Co-digestion of microalgae biomass and or other food and agricultural wastes can result in enhanced energy recovery.

Integration with fermentation processes

Use of fermentation for MFC operation was also well studied (Schröder 2007). As discussed earlier, numerous fermentative and photo-heterotrophic processes result in the production of various energy-rich reduced metabolites such as hydrogen, ethanol or formate which can be oxidized in the bioanodes. Karube and co-workers immobilized hydrogen-producing cultures as biocatalysts and platinum as an electrocatalyst for hydrogen oxidation (Karube et al. 1977). The platinum electrode poisoning due to corrosion and related lower power densities were addressed by developing polyaniline-coated platinum electrodes in later studies (Schröder et al. 2003; Niessen et al. 2004). Heterotrophic, photo-heterotrophic and even purely photosynthetic microorganisms and the access to complex carbohydrates like starch and cellulose have been exploited in number of ways for current generation in MFCs (Rosenbaum et al. 2005, 2006). A photosynthetic microbial electrochemical cell (PMEC) for hydrogen production was developed using photosynthetic microorganisms and heterotrophic bacteria confined to anode and cathode compartments, respectively (see Fig. 4). In this process, hydrogen production could be enhanced by solar energy utilization, in addition to integrating the DF anaerobic digester and energy harvesting by microbial fuel cells (Bensaid et al. 2015). Harvesting of solar energy in photosynthetic MEC provides the heat required for enhancing biogas production in AD and the carbon dioxide as well as low COD products produced in AD can be utilized by photosynthetic microorganism to grow biomass (Colombo et al. 2017).

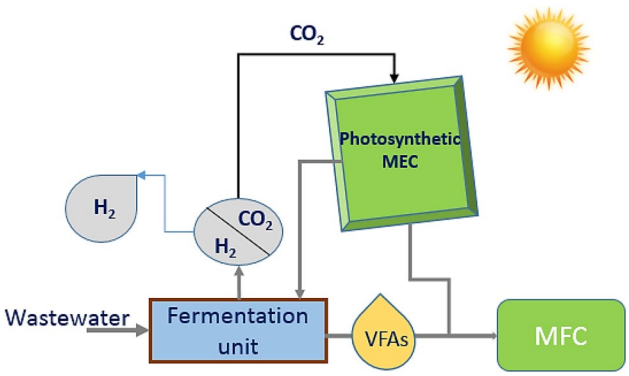

Fig. 4 Photosynthetic microbial electrochemical cell integrated with dark fermentation and microbial fuel cell for wastewater treatment and energy recovery. Reproduced with permission form Bensaid et al. (2015)

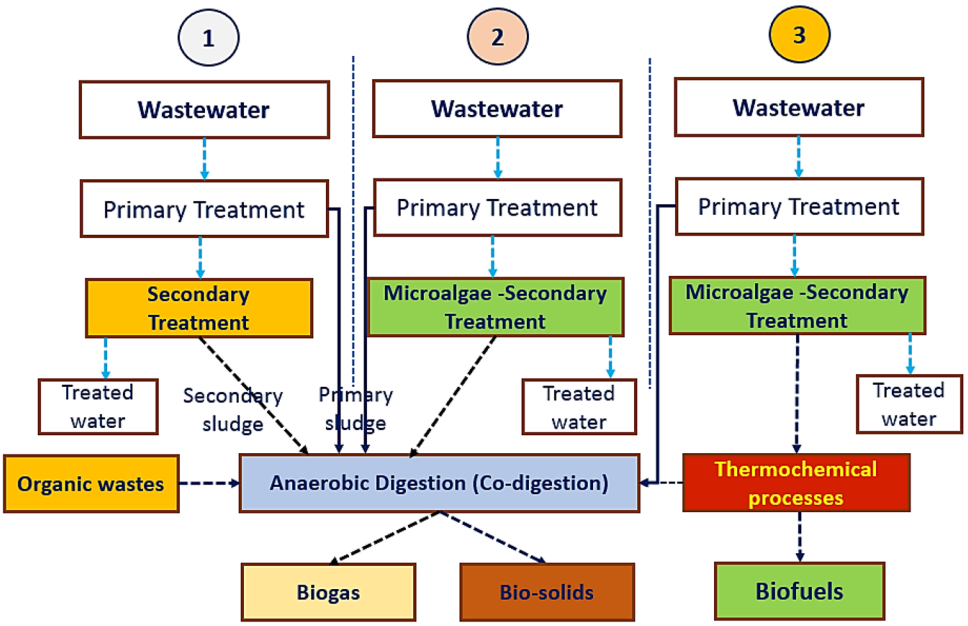

Fig. 3 Approaches for energy-positive wastewater treatment: 1. Enhanced energy recovery through primary sludge collection and co-digestion of organic wastes for biogas production, 2. Microalgae–bacteria mixed culture system for energy conservation and co-digestion of mixed biomass and 3. Separate microalgae cultivation system or microalgae–bacteria system for wastewater treatment and thermochemical biofuel production. After Gude (2015b)

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Integrating DF, AD and MFCs

Several combinations of fermentation, AD and BES are possible (see Fig. 5). Microbial fuel cells have been utilized to treat the supernatant fraction of digestates, to simultaneously degrade residual soluble/suspended organic matter, reduce nitrogen content and produce bioelectricity (Fradler et al. 2014). In particular, air–cathode MFCs were demonstrated to have potentially high COD removal efficiency with a variety of liquid streams (Sciarria et al. 2015; Fradler et al. 2014; Kim et al. 2015; Pant et al. 2016) and, additionally, to act simultaneously as nitrification–denitrification systems (Virdis et al. 2010).

Coupling MFC to DF and AD is a recently developed technological concept which was demonstrated in various combinations such as AD + MFC or DF + AD + MFC. Premier et al. (2013) evaluated the first configuration of this kind followed by Fradler et al. (2014) who studied the performance of a 4-module tubular MFC applied after a DF + AD system as a polishing stage. With hydraulic residence time of 8 h, and the influent concentration of 1029 mg COD L^{-1}, the COD removal remained below 10%, with a relatively low energy recovery per raw influent volume (92 J L^{-1}). Higher concentrations of COD, to avoid high dilutions of the AD effluent, resulted in even lower efficiencies.

Many other integrated processes such as biogas–biohydrogen production, biogas–MFC or biogas–biofertilizers production were developed to increase the spectrum of products with higher increased value and waste reduction (Fradler et al. 2014; Premier et al. 2013; Ledda et al. 2013; Schievano et al. 2012). Of particular interest is a recent study in which DF, AD and microbial fuel cells and solid–liquid separation processes were integrated to co-produce hydrogen, methane, bioelectricity and biofertilizers (Fig. 6a). Two integrated systems, AD + MFC and DF + AD + MFC were compared to a traditional one-stage AD system in converting a mixture (COD = 124 ± 8.1 gO_2 kg^{-1} fresh matter) of swine manure and rice bran. AD + MFC produced a methane yield of 182 LCH_4 kg^{-1} COD, while DF + AD + MFC produced biohydrogen and biomethane of 27.3 ± 7.2 LH_2 kg^{-1} COD and 154 ± 14 LCH_4 kg^{-1} COD, respectively. The liquid fraction was treated in MFCs which produced power densities of 12–13 W m^{-3} and average bioelectricity yields

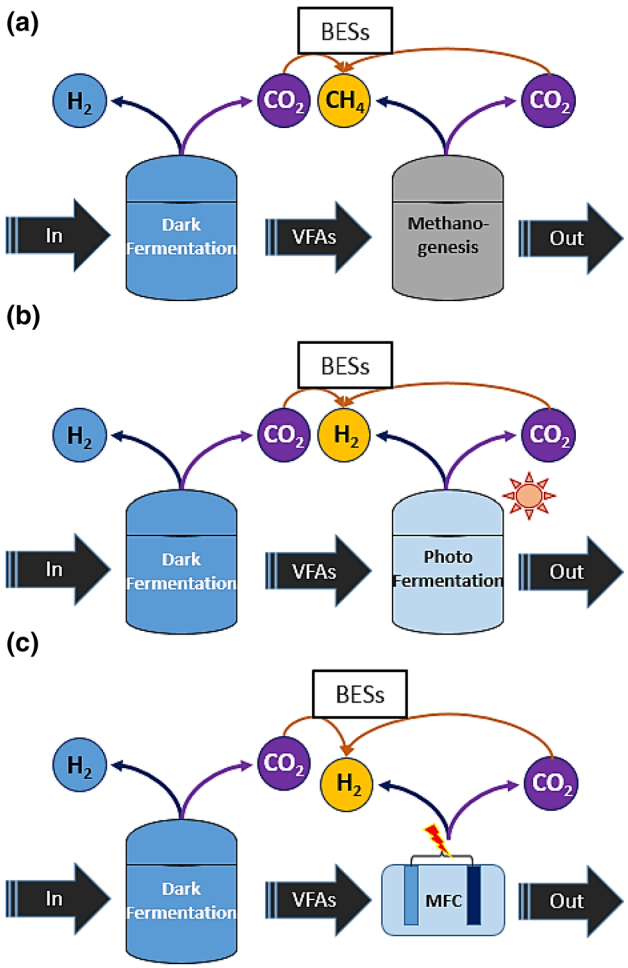

Fig. 5 Various possibilities for integrating AD, fermentation and BES

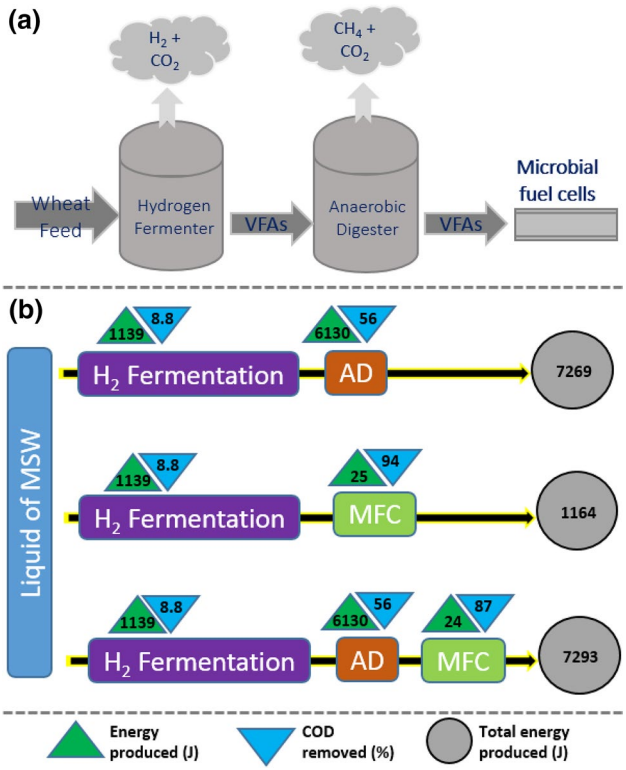

Fig. 6 An experimental unit scheme (**a**) and **b** energy production and COD removal efficiency comparison for different biochemical processes for high-strength wastewater from swine manure and rice bran (data taken from Schievano et al. 2016)

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of 39.8 Wh kg^{-1} COD to 54.2 Wh kg^{-1} COD, respectively (Schievano et al. 2016).

Further, utilization of a high-strength organic wastewater derived from the municipal solid waste was processed through different biochemical processes including AD, fermentation and microbial fuel cells processes (Rózsenberszki et al. 2017). Single-stage, double-stage and three-stage treatment processes were investigated. For the single-stage processes, COD removal efficiency was the highest in MFC (92.4%) followed by AD (50.2%) and hydrogen fermentation (8.8%) process. However, the energy density (J g^{-1} COD removed d^{-1}) was the lowest for MFC (0.43) followed by AD (205) and hydrogen fermentation (2277). The cumulative energy production for other integrated, double-stage and three-stage processes is shown in Fig. 6b. COD removal efficiency and energy generation trends are similar to the single-stage processes. It should be noted that the three-stage process produced a slightly excess total energy. From these results, as already been discussed MFC technology has low power production capacity despite its high COD removal capacity. This is mainly due to the energy conversion and transfer losses in the microbial bioelectrochemical system which need to be addressed in the future research efforts.

Carbon sequestration

All of the biological processes described in previous sections produce carbon dioxide which represents a valuable resource for many beneficial uses as shown in Fig. 7 and by the electrochemical reactions below (Bajracharya et al. 2017). Most of the electrochemical approaches require external catalysts and high energy input to facilitate CO_2 reduction. Highly active, selective and stable electrocatalysts are required for these applications. In addition, overpotentials should be overcome to improve energy efficiency. BES which utilize microorganisms and their metabolic products as biocatalysts could be attractive considering the aforementioned issues. There are several advantages associated with BES applications (Bajracharya et al. 2017): (1) low energy input needed to activate CO_2 reduction due to the biological intervention, (2) selective for reactions, even on multi-step reactions, (3) adaptability of microbes for producing different products, (4) low-cost design and operation, (5) reaction at ambient conditions, (6) recyclability of the biocatalyst and (7) possibility of high value uplift in the market.

Carbon $CO_2 + 4H^+ + 4e^- = C + 2H_2O$ ($E = 0.21$ V)

$CO_2 + 2H_2O + 4e^- = C + 4OH^-$ ($E = -0.627$ V)

Methane $CO_2 + 8H^+ + 8e^- = CH_4 + 2H_2O$ ($E = 0.169$ V)

$CO_2 + 6H_2O + 8e^- = CH_4 + 8OH^-$ ($E = -0.659$ V)

Ethanol $2CO_2 + 12H^+ + 12e^- = CH_3CH_2OH + 3H_2O$ ($E = 0.084$ V)

$2CO_2 + 9H_2O + 12e^- = CH_3CH_2OH + 12OH^-$ ($E = -0.744$ V)

Ethylene $2CO_2 + 12H^+ + 12e^- = CH_2CH_2 + 4H_2O$ ($E = -0.064$ V)

$2CO_2 + 8H_2O + 12e^- = CH_2CH_2 + 12OH^-$ ($E = -0.764$ V)

Methanol $CO_2 + 6H^+ + 6e^- = CH_3OH + 4H_2O$ ($E = 0.016$ V)

$CO_2 + 5H_2O + 6e^- = CH_3OH + 6OH^-$ ($E = -0.812$ V)

Formaldehyde $CO_2 + 4H^+ + 4e^- = CH_2O + H_2O$ ($E = -0.07$ V)

$CO_2 + 3H_2O + 4e^- = CH_2O + 4OH^-$ ($E = -0.898$ V)

Formic acid $CO_2 + 2H^+ + 2e^- = HCOOH$ ($E = -0.25$ V)

Formate $CO_2 + H_2O + 2e^- = HCOO^- + OH^-$ ($E = -1.078$ V)

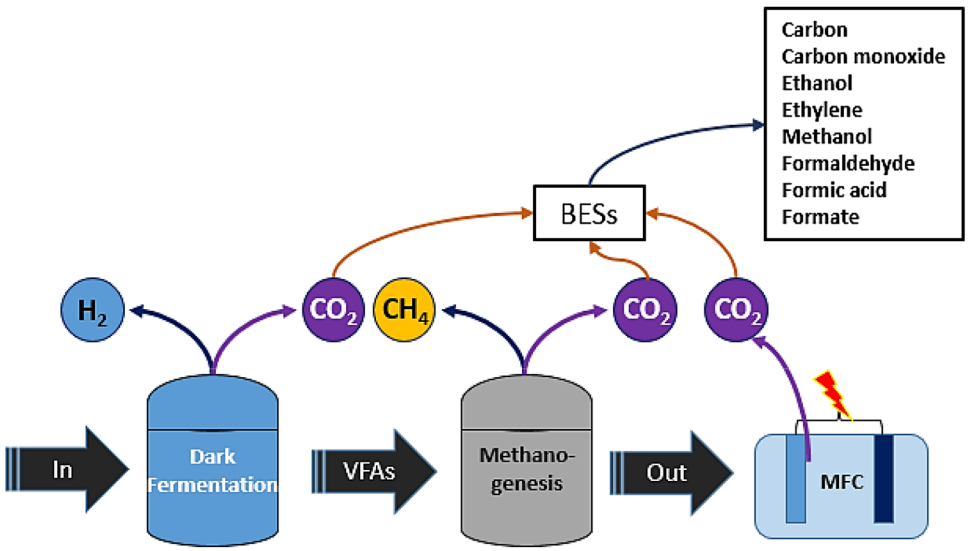

Fig. 7 Carbon sequestration in integrated fermentation, AD and BES

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Other integrated processes for BES

BES offer a wide flexibility to be incorporated into other beneficial processes due to their simple nature and elegant operation. Integrating BES with other conventional aeration and membrane processes may provide additional energy, water and resource recovery benefits.

Integration with membrane processes

BES provide an energy-efficient approach for wastewater treatment. However, the effluent still requires further treatment for direct discharge or reuse. Integration of membrane filtration in MFCs can be beneficial to achieve high-quality effluents. Dynamic, ultrafiltration and forward osmosis membrane with different pore sizes were evaluated for integration in MFCs. The integration can be accomplished through either internal or external configurations. In an internal configuration, membranes can act as a separator between the electrodes, or be immersed in the anode/cathode chamber as a filtration component. The external configuration allows for MFC and membrane module to be operated independently (Yuan and He 2015). For example, by integrating a forward osmosis membrane as a separator in an MFC, osmotic MFCs were created (Zhang et al. 2011). The performance of this osmotic MFC was examined using sodium chloride solution or synthetic seawater. In parallel, an MFC with a cation exchange membrane (CEM-MFC) was used for comparison purposes. Osmotic MFC produced more electricity than the CEM-MFC. Water flux from the anode chamber to cathode chamber was observed in osmotic MFC but not in CEM-MFC. The solute concentration in the catholytes had a significant effect on electricity generation and water flux potentials (Zhang et al. 2011). This process can be used as a pre-treatment technique for seawater desalination processes.

Integration with aeration tank in conventional wastewater treatment plant

Integrating MFCs into an aeration tank does not require additional space but offers several other potential benefits. In MFC, wastewater can be treated under an anaerobic condition reducing energy consumption with much less suspended solids concentrations, thus less sludge production. MFCs can produce some electricity, which can be potentially applied to offset the energy consumption by the treatment process. A real-world application of MFCs was reported in a recent study by Zhang and co-workers (Zhang et al. 2013). Two 4-L tubular MFCs were installed in a municipal wastewater treatment plant which treated primary effluents for more than 400 days. Both MFCs removed 65–70% COD at a hydraulic retention time of 11 h and reduced about 50% suspended solids. This study showed that the MFCs integration with existing wastewater treatment plants may allow for energy-positive treatment. Integration of a denitrifying MFC with this process improved the total nitrogen removal rate from 27.1 to 76.2%. Several other field applications of MFCs include a ceramic cascade temporarily installed in a municipal wastewater treatment plant (Winfield et al. 2012), a multi-electrode MFC system for contaminant removal (Heidrich et al. 2014) as well as for winery wastewater treatment (Cusick et al. 2011), and more recently, floating MFCs at the Nosedo, Milan wastewater treatment plant (Martinucci et al. 2015).

Integration with other bioelectrochemical systems

Microbial electrolysis cells (MECs) have been investigated for hydrogen production using various organic substrates. An external voltage must be applied to recover hydrogen from the fermentation end products and to make the reaction thermodynamically favorable in the presence of high concentrations of hydrogen (Pasupuleti et al. 2015). This energy can be provided by photobiological or bioelectrochemical processes. In MECs, the electrical voltage needed (0.110 V in theory, >0.2 V in practice) is significantly lower than the theoretical voltage needed for water electrolysis (1.8 V in practice) (Logan et al. 2008) which may be provided by a microbial fuel cell fed by same wastewater source. It may actually result in maximized resource utilization. Hydrogen gas production from cellulose was investigated using an integrated hydrogen production process consisting of a DF reactor and microbial fuel cells as power supply units for a microbial electrolysis cell (Fig. 8) (Wang et al. 2011). Two MFCs (each 25 mL) connected in series to an MEC (72 mL) produced a maximum of 0.43 V using fermentation effluent as a feed, achieving a hydrogen production rate from the MEC of 0.48 m^3 H_2 m^{-3} d^{-1} (based on the MEC volume), and a yield of 33.2 mmol H_2 g^{-1} COD removed in the MEC. The overall hydrogen production for the integrated system (fermentation, MFC and MEC) was increased by 41% compared with fermentation alone to 14.3 mmol H_2 g^{-1} cellulose, with a total hydrogen production rate of 0.24 m^3 H_2 m^{-3} d^{-1} and an overall energy recovery efficiency of 23% (based on cellulose removed) without the need for any external electrical energy input. The voltage produced by the MFCs for the MEC was relatively stable at 0.441 ± 0.010 V for the initial 52 h and then decreased to 0.384 V over the next 20 h as the cathode potential increased from 0.877 ± 0.010 to 0.816 V. The anode electrode potential of the MEC remained relatively constant at -0.434 ± 0.008 V over the 72 h cycle. When the same MEC unit was connected to an external power source at an applied (higher) voltage of 0.8 V, it resulted in an additional

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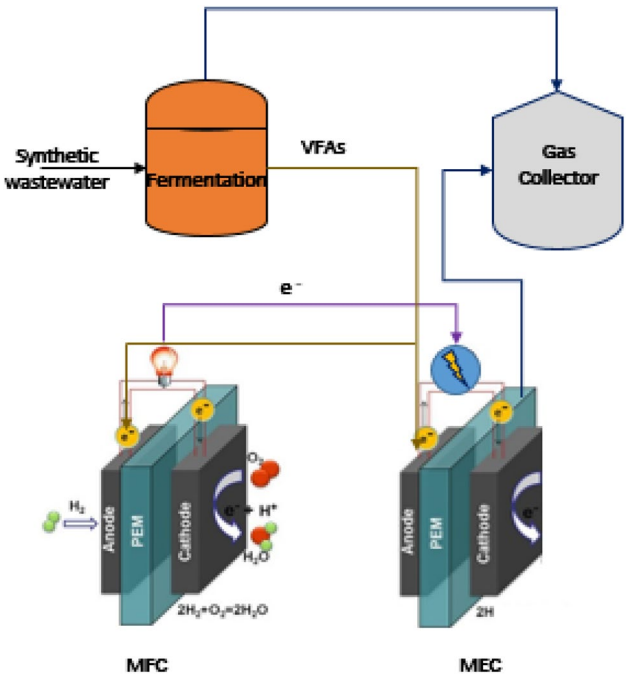

Fig. 8 Integrated MFC–MEC system for hydrogen production

production of 267 mL of hydrogen gas. This suggested that MFCs should produce more voltage for higher volumes of hydrogen production. In addition, hydraulic retention times of the reactors should be optimized to maximize the energy conversion efficiencies.

Integrated MEC and fermentation process can achieve superior results (Lu and Ren 2016). For example, hydrogen yields increase from 14 to 27% for fermentation alone and 23–71% for MEC alone to > 85% when combined. Similarly, the production rate (m^3 H_2-m^{-3} reactor-d^{-1}) could increase from 0.2 to 10 for fermentation alone and 0.1 to 3.4 for MEC alone to > 10 when combined. A novel scheme of integrating microbial desalination and MEC processes to achieve removal of nutrients, metals and salts from municipal wastewater, industrial wastewater and seawater, respectively, was demonstrated in a recent study (Li et al. 2017). The combined unit was able to produce 293.7 mW m^{-2} of power combined with 64% of salt removal, while 99.5% of metals and 95% of nitrogen removals were accomplished from industrial and municipal wastewaters simultaneously. The MDC–MEC system has shown a net positive energy potential of 0.027 kWh m^{-3} confirming the benefits of MDC process.

Kokabian and Gude (2013) have reported an energy perspective of a MDC process integrated with microalgae biocathode system. MDCs produce bioelectricity while desalinating the saline water through which an estimated 1.8 kW h of bioelectricity can be generated for every 1 m^3 of wastewater. The lowest energy requirements for a seawater reverse osmosis process require 2.2 kW h of electricity. When the two functions of wastewater treatment and desalination performed by MDCs are considered, the potential energy savings in MDCs are reported as 4 kW h m^{-3} of both wastewater and seawater. In addition, lipid production of 0.04 kg m^3 d^{-1} from the microalgae biomass (with a specific energy value of 48 MJ kg^{-1} and an electric conversion efficiency of 40%), a maximum electrical energy of 0.21 kW h m^{-3} of treated wastewater can be obtained which further increases the net energy benefit of the MDC and microalgae integrated process to 4.21 kW h m^{-3} or 2.01 kW h m^{-3}, respectively, with and without the desalination energy credit. In systems integrated with algal harvesting, the energy recovery benefits could be even higher since microalgae could have an energy content of 5–8 kW h kg^{-1} dry weight which can be recovered in the form of biofuels.

Biorefinery configuration

Wastewater treatment in MFCs can be integrated with microalgae and other photosynthetic oleaginous microorganisms to serve both as electron-acceptor-producing biomass which could serve as feedstock for biofuel production (Baicha et al. 2016). Anaerobic treatment of wastewater in anode chamber produces carbon dioxide which can be utilized by microalgae in the cathode chamber as a carbon source. Microalgae in return generate oxygen which can be available in situ conditions to serve as electron acceptor (Gude et al. 2013). Microalgae can be used to produce biodiesel, bioethanol, methane or hydrogen. Dry microbial biomass (dead cells) can be used as feed for MFCs (Blair et al. 2014; Velasquez-orta et al. 2009; Gude 2015c; Martinez-Guerra et al. 2014, 2018; Martinez-Guerra and Gude 2016). In addition to electricity production, electrons generated in the microbial fuel cell at the anode can also be used to produce chemical fuels, such as hydrogen gas (Logan et al. 2008; Wang et al. 2011). However, an external voltage supply is required to overcome the thermodynamic barrier for the conversion from protons to hydrogen gas. Utilizing solar energy, a number of solar-assisted microbial fuel cells were demonstrated recently. This can be done by coupling the conventional MFC with photosynthetic bacteria, semiconductor photo-electrodes, solar cell or photo-electrochemical cell and photosynthetic microbial desalination cells (Kokabian and Gude 2013, 2015; Kokabian et al. 2018a, b; Rosenbaum et al. 2006; Bensaid et al. 2015). In these devices, solar energy was utilized to facilitate bioelectricity or hydrogen generation. The demonstration of these new solar-assisted MFC devices opens up new opportunities in the recovery of chemical energy in wastewater for chemical fuel production.

In recent years, the field of renewable fuel production from lignocellulosic biomass, such as agricultural residues

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and woody biomass, has made great advancements. The main components of lignocellulosic biomass are cellulose, hemicelluloses and lignin. Lignocellulosic biomass contains a small amount of carbohydrates that are derived from the following monosaccharides: L-rhamnose, L-fructose, D-fucose and D-ribose (Catal et al. 2008). Several studies investigated the possibility of hydrogen production from cellulose and other lignocellulosic biomass in microbial electrolysis cells. Use of twelve monosaccharides originated from lignocellulosic biomass as feedstock in air–cathode MFCs was evaluated (Catal et al. 2008). The substrates included six hexoses (D-glucose, D-galactose, D(−)-levulose (fructose), L-fucose, L-rhamnose, and D-mannose), three pentoses (D-xylose, D(−)-arabinose and D(−)-ribose), two uronic acids (D-galacturonic acid and D-glucuronic acid) and one aldonic acid (D-gluconic acid). The maximum power density obtained from these carbon sources ranged from 1240 to 2770 mW m^{-2} at current density range of 0.76–1.18 mA cm^{-2}. Mannose resulted in the lowest maximum power density, whereas d-glucuronic acid generated the highest maximum power density.

In a biorefinery configuration, the potential use of MFCs to treat the inhibitors, sugars and lignin generated in the corn stover ethanol production plant was evaluated (Borole et al. 2009). This process assumed a solids loading of 30%. Acetic acid, 2-furfural, HMF and glucose and its oligomers are present in the recycle stream at a concentration of 6.5, 1.5, 0.23 and 2.1 g L^{-1}, respectively. Glucose was consumed prior to the removal of the inhibitor molecules by the MFC consortium. A minimum of three different MFCs were presumed to be needed, each with a certain substrate specificity (namely sugars, acetate or organic acids and lignin- + sugar degradation products). Assuming a 60% CE, the process yielded 2.5 megawatts of power which is equivalent to one quarter of the total power needed for the biorefinery plant. However, MFCs need to overcome several challenges to meet this target. Hydrogen production via MECs was identified as a better alternative for the overall biorefinery process in terms of economic benefits.

Conclusions

The role of AD, fermentation and BES toward developing sustainable wastewater treatment systems was discussed in detail with case studies. Integrated processes are being conceived as a potential platform for enhancing energy conversion efficiency in these systems. Despite many recent advancements in these areas, there are still many obstacles that need to be overcome to make the integrated processes viable for practical applications. Many studies are evaluated at laboratory scale which need pilot- or field-scale demonstrations. Detailed mass and energy balances and proper accounting of energy losses are warranted to better understand the feasibility of these technologies. Some fundamental studies are still warranted in research areas such as microbial fuel cells and microbial electrolysis cells to better understand the energy conversion efficiencies and to improve the process performances. Inhibitory effects that dominate the AD and fermentation processes should be investigated, especially for biorefinery applications involving lignocellulosic biomass-related products. Life cycle assessment and techno-economic analysis should be developed to determine the sustainability of these process configurations.

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