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A fluidized bed membrane bioelectrochemical reactor for energy-efficient wastewater treatment

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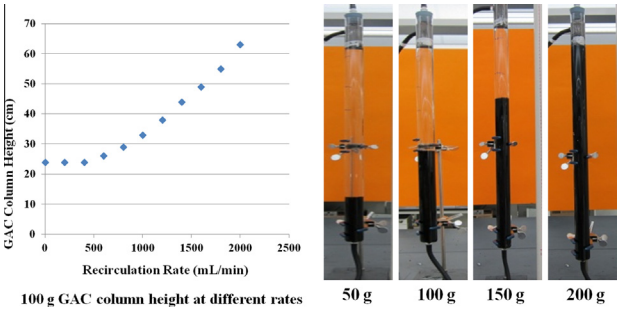
HIGHLIGHTS

- Fluidized granular activated carbon (GAC) helps control membrane fouling in a MBER.
- Fluidized GAC plays a minor role in direct contribution to electricity generation.
- Fluidized bed MBER can be coupled to a microbial fuel cell for improved treatment.
- The couple system could achieve theoretically energy neutral.

GRAPHICAL ABSTRACT



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ABSTRACT

A fluidized bed membrane bioelectrochemical reactor (MBER) was investigated using fluidized granular activated carbon (GAC) as a mean of membrane fouling control. During the 150-day operation, the MBER generated electricity with contaminant removal from either synthetic solution or actual wastewater, as a standalone or a coupled system. It was found that fluidized GAC could significantly reduce transmembrane pressure (TMP), although its function as a part of the anode electrode was minor. When the MBER was linked to a regular microbial fuel cell (MFC) for treating a wastewater from a cheese factory, the MFC acted as a major process for energy recovery and contaminant removal, and the coupled system removed more than 90% of chemical oxygen demand and >80% of suspended solids. The analysis showed that the ratio of energy recovery and consumption was slightly larger than one, indicating that the coupled system could be theoretically energy neutral.

1. Introduction

Sustainable wastewater treatment is of great importance to maintain a sustainable societal development, and its key features include high quality of treated effluent and energy-efficient treatment process. Those features can be realized separately using technologies such as membrane bioreactors (MBR) and microbial fuel cells (MFCs). MBR technology has been applied to treat both municipal and industrial wastewaters. It has several advantages over conventional activated sludge system (Judd, 2008); however excessive energy consumption due to aeration and antifouling control is still a hurdle for its application especially in some energy-shortage areas. MFC technology is an emerging concept and has been intensively studied as an alternative method for energy-efficient wastewater treatment (Wang and Ren, 2013). Comparing with conventional activated sludge technology, MFCs have less or no demand for aeration and produce much less sludge due to anaerobic treatment (Rabaey and Verstraete, 2005). Research has demonstrated that MFC treatment of domestic wastewater could be energy-neutral (Zhang et al., 2013).

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Integration of MFCs with a membrane filtration process such as MBRs may provide an ideal solution to achieve high-quality effluent, with less energy requirement (than aerobic treatment systems). An early effort used the biofilm formed on the stainless steel as filter materials to achieve low effluent turbidity and high removal of both organic matter and ammonium nitrogen (Wang et al., 2011, 2012). The use of micro/ultra filtration membranes as filtration media in MFCs was reported in a membrane bioelectrochemical reactor (MBER), in which the commercially available hollow fiber membranes were installed in the anodic chamber of a tubular MFC (Ge et al., 2013b). This MBER system effectively treated both synthetic and domestic wastewater but membrane fouling was a serious issue. To facilitate the application of proper fouling control and minimize its effects on the anode microbial activity, hollow-fiber membranes were installed in the cathode compartment of an MFC with either an aerobic or anoxic cathode (Li et al., 2014). In addition to internally installed membrane, MFCs were also directly linked to an MBR (Malaeb et al., 2013).

In a treatment system containing membrane, fouling/scaling is always a great challenge. A new method for fouling control was developed by using fluidized granular activated carbon (GAC) in an anaerobic membrane bioreactor (AnMBR) (Kim et al., 2011). This method required little physical or chemical cleaning of membrane module during 120-day operation, and it also had energy benefits for fouling control comparing to conventional MBR. This fluidized AnMBR has been advanced to pilot test that achieved satisfactory performance (Shin et al., 2014). A similar fluidized AnMBR was linked to a single-chamber MFC as a post-treatment for improving effluent quality (Ren et al., 2014). The concept of fluidized particle bed was also applied to prevent inorganic scaling deposit on the surface of cathode electrode in a microbial electrolysis cell (MEC) (Cusick et al., 2014).

Intrigued by the fluidized AnMBR concept, a fluidized bed membrane bioelectrochemical reactor (MBER) was developed here for energy-efficient wastewater treatment. This MBER aimed to take advantage of fluidized GAC as both fouling control media and partial anode electrode. The objectives of this study were: (1) to examine the feasibility of electricity generation and wastewater treatment in this fluidized bed MBER; (2) to investigate membrane fouling affected by the operating conditions; and (3) to formulate a treatment system by linking the MBER to an MFC for treating actual wastewater.

2. Methods

2.1. Reactor construction

2.1.1. MBER setup

The MBER was constructed as a tubular reactor (45 cm long and 5 cm in diameter) made of cation exchange membrane (CEM-Ultrex CMI 7000, Membrane International, Inc., Glen Rock, NJ, USA) (Fig. 1A). The main body of the anode electrode was a piece of carbon cloth supported by stainless steel mesh, which was installed inside the membrane tube (along the interior wall). Ten 38-cm PVDF hollow fiber membranes (15,000 Dalton, Litree Purifying Technology Co., China) were installed inside the membrane tube, which was then filled with 230 g of 8 × 30 mesh GAC (Calgon Carbon Corp., Pittsburgh, PA, USA), resulting in an anode liquid volume of 700 mL. The hollow fiber membranes had a pore size of 0.02 μm and the total membrane surface area was 0.021 m^2. Before use, carbon cloth was soaked in acetone solution overnight and heated for 30 min at 450 °C (Wang et al., 2009). The cathode electrode consisted of one layer of carbon cloth (Zoltek Corporation, St. Louis, MO, USA) coated with Pt/C powder (10%, Etek, Somerest, NJ, USA) with a loading rate 0.05 mg Pt cm^{-2}. The cathode electrode wrapped the membrane tube and was exposed in the air for passive oxygen supply as described in the previous studies (Zhang et al., 2010). The anode and cathode electrodes were connected by using titanium wires to an external resistor of 48 ohm (which was determined by polarization tests for high power output).

2.1.2. MFC construction

A tubular MFC was constructed with a CEM tube (45 cm long and 5 cm diameter), which contained a one-meter long carbon brush folded as an anode electrode. The anode liquid volume was about 1000 mL. The cathode electrode was a piece of carbon cloth treated and coated with Pt catalyst similarly to that of the MBER. The electrodes were connected to an external circuit using titanium wires.

2.2. Operation conditions

2.2.1. MBER operation

The MBER anode was inoculated with anaerobic digester sludge from a wastewater treatment facility (South Shore, Oak Creek, WI, USA) and was operated at room temperature of ~20 °C. The synthetic anode solution contained (per L of tap water): sodium acetate 0.5 g; NH_4Cl 0.15 g; NaCl 0.5 g; $MgSO_4$ 0.015 g; $CaCl_2$ 0.02 g; KH_2PO_4 0.53 g; K_2HPO_4 1.07 g and 1 mL trace element (He et al., 2006). The anolyte was recirculated at 800 mL min^{-1} unless elsewhere stated. This rate was determined according to a test that examined the height of the fluidized GAC column affected by recirculation rate, and found that 800 mL min^{-1} could fully fluidize the GAC in the MBER. The hollow fiber membranes were operated under an intermittent mode that extracted water for 4 min and then relaxed for 1 min. Tap water was used as a catholyte to rinse the cathode electrode from top to bottom and additional tap water was added to compensate for evaporation. The effects of the anolyte recirculation and hydraulic retention time (HRT) were examined. Organic loading rates varied when adjusting HRT: 0.40 kg COD m^{-3} d^{-1} at 24 h, 0.80 kg COD m^{-3} d^{-1} at 12 h, and 1.31 kg COD m^{-3} d^{-1} at 8 h. The anode of the MBER was acclimated with the seed sludge for about one week with varying the external resistance from 2000 to 10 Ω. After 100-day operation, the tubing for the anode feeding and the influent port of the CEM tube were clogged by GAC due to operating problems, and subsequently a new CEM tube was constructed (while other materials remained same) and used to the end of the study.

2.2.2. Integrated MFC + MBER operation

After four-month operation, the fluidized MBER was linked to an MFC to treat an industrial wastewater (Schreiber Foods, Inc., WI, USA). The wastewater collected from the effluent of DAF (dissolved air flotation) unit was first fed into the MFC anode and then the MFC effluent was supplied to the MBER (Fig. 1B). The coupled system was operated under an overall HRT 19.6 h (11.6 h in the MFC and 8 h in the MBER). The organic loading rate was about 1.00 kg COD m^{-3} d^{-1}. The anolyte recirculation rates were 200 and 600 mL min^{-1} for the MFC and the MBER, respectively.

2.3. Measurement and analysis

The voltage was recorded every 3 min by a digital multimeter (2700, Keithley Instruments, Cleveland, OH). The pH was measured using a benchtop pH meter (Oakton Instruments, Vernon Hills, IL, USA). The concentration of Chemical Oxygen Demand (COD), ammonium, nitrite and nitrate concentration were measured according to the manufacture's procedures (Hach DR/890, Hach Company, Loveland, CO, USA). The trans-membrane pressure (TMP) was manually recorded 3 times daily and the average value was reported in this study. The turbidity was measured using a

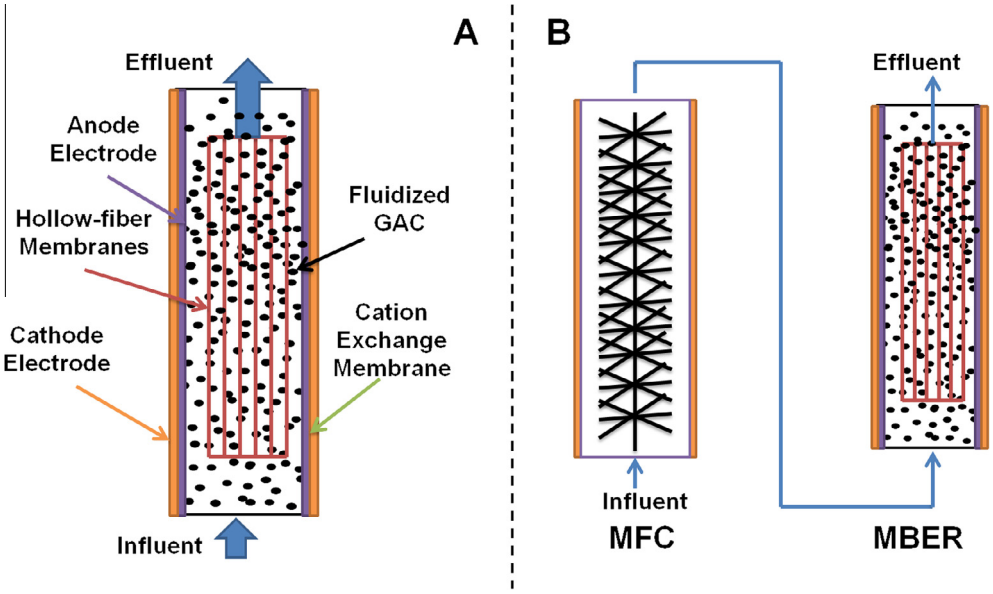

Fig. 1. The schematics of the system: (A) an individual MBER reactor; (B) the coupled MFC–MBER system.

turbidimeter (DRT 100B, HF Scientific, Inc., Fort Meyers, FL, USA). The polarization testing was performed by a potentionstat (Reference 600, Gamry Instruments, Warminster, PA, USA) at a scanning rate of 0.2 mV s^{-1}. The current and power density was normalized to the anode liquid volume. The Coulombic Efficiency was calculated according to the previous study (Logan et al., 2006).

The energy performance was evaluated by analyzing energy production and consumption. The estimation of energy consumption (by the pumping system for feeding, recirculation and membrane extraction) was based on power consumption, calculated by the following equation (Kim et al., 2011):

$$P = \frac{Q\gamma E}{1000} \quad (1)$$

where P is power requirement (kW), Q is flowrate (m^3 s^{-1}), γ is 9800 (N m^{-3}) and E (m H_2O) is head loss. The energy consumption for membrane filtration was calculated based on an average vacuum pressure of 37 kPa and permeate flow of 0.8 mL min^{-1}. Energy conversion efficiency was assumed as 61.2% from electrical energy to pumping energy (Kim et al., 2011). Energy recovery was calculated by normalizing average operation power to unit volume of treated wastewater (kWh m^{-3}) or based on the amount of organic contaminants removal (kWh kg COD^{-1}) (Xiao et al., 2014).

3. Results and discussion

3.1. Feasibility of the fluidized bed MBER

The feasibility of the fluidized bed MBER was demonstrated by examining its electricity generation, contaminant removal, and membrane pressure during a 100-day operation. The MBER produced a current density generally varying between 6 and 8 A m^{-3} (Fig. 2A). Adjusting HRT and the anolyte recirculation rates did not obviously affect current generation, possibly because substrate distribution was affected by the presence of GAC and the selected recirculation rate could not significant change the distribution situation; the exact reasons warrant further investigation.

The hypothesis that GAC may act as a part of the anode electrode was investigated by compared the MBER with and without GAC. The results of polarization tests showed that the maximum power density of the MBER containing GAC was 1.8 W m^{-3}, 50% higher than that (1.2 W m^{-3}) of the MBER without GAC (Fig. 3). Likewise, the MBER with GAC produced a maximum current density of 16.4 A m^{-3}, 53% higher than 10.7 A m^{-3} in the one without GAC. Because the power/current densities were calculated based on the liquid volume and the presence of GAC reduced the liquid volume from 1000 to 700 mL, it is also necessary to compare the absolute power/current output between those two systems. The maximum absolute power production was 1.26 mW with GAC, slightly higher than 1.20 mW in the absence of GAC. The maximum current generation in the MBER with GAC was 11.5 mA, whereas 10.7 mA was produced when no GAC was in the MBER. The difference in the normalized energy recovery (NER) (Ge et al., 2013a) was more obvious: the MBER with the fluidized GAC recovered 0.0146 kWh m^{-3}, much higher than 0.0095 kWh m^{-3} when GAC was absent; however, the same HRT for those two systems (the MBER with GAC treated less wastewater than the other system) decreased the significance of the difference in NER. Those results do not provide a strong proof that the presence of GAC could greatly improve electricity production and the fluidized GAC could function as a part of the anode electrode, which is different from a recent study using fluidized bed as the anode electrode (Wang et al., 2014). The possible reason for that difference may be that the liquid velocity of the MBER at 800 mL min^{-1}, about 6.7 mm s^{-1}, may not be the optimal velocity for power output. According to Wang's work, the maximum power density of their MFC reached the highest at 5–6 mm s^{-1} and would become less at either lower or higher velocity. Due to the difference between the present MBER system and their MFC system, it will not be reasonable to use their optimal value to evaluate the present MBER performance; however, their results reveal the relationship between the liquid velocity and electricity generation, which will help to identify the optimal liquid velocity for the MBER. If the future studies can demonstrate the role of the fluidized GAC as a part of the anode electrode, it will make the concept of fluidized bed MBER more attractive and competitive.

The contaminant removal focused on the removal of COD, which was affected by two separate factors, HRT and the anolyte recirculation rate (Fig. 2C). At an HRT of 24 h (water flux of 1.38 L m^{-2} h^{-1}) and a recirculation rate of 800 mL min^{-1}, the MBER

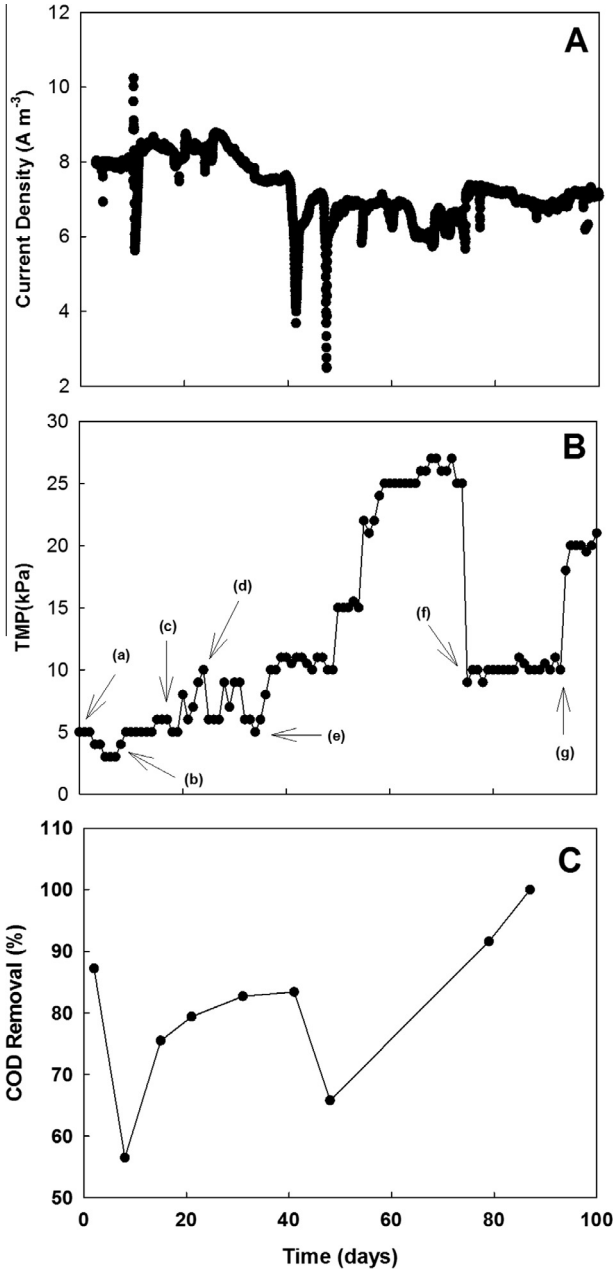

Fig. 2. The performance of the MBER with the synthetic solution: (A) electricity generation; (B) TMP variation: the arrows (a), (b), (d), (f) and (g) indicate HRT 24, 12, 8, 8, and 5 h, respectively, under the recirculation flow rate of 800 mL min^{-1}, and the arrows (c) and (e) indicate HRT 12 and 8 h with no recirculation; (C) the COD removal efficiency.

removed 87.1% of COD. Reducing the HRT to 12 h (water flux of 2.77 L m^{-2} h^{-1}) at a recirculation rate of 800 mL min^{-1} decreased the COD removal to 56.0%, likely due to the increased COD loading rate at a lower HRT. Stopping the recirculation (0 mL min^{-1}) at the HRT 12 h improved the COD removal to 79.5%; this interesting phenomenon was possibly related to the interaction between fluidized GAC and substrate distribution. However, because of the short testing period for this situation (10 days), it was also possible that the MBER was adapting to the more COD input in the first few days of decreasing the HRT and exhibited improved performance after adaptation. A longer term operation (nearly 70 days) was used to investigate the effect of recirculation at a shorter HRT of 8 h (water flux of 4.17 L m^{-2} h^{-1}). The MBER removed 82.7% of COD in the first nine days after the HRT change at an anolyte recirculation of 800 mL min^{-1}. A lower COD removal was obtained in the following forty days without recirculation. After the recirculation was restarted the MBER removed 91.6% of COD. One can see from the variation of COD removal efficiency that decreasing HRT could instantly decrease the COD removal efficiency because of a higher organic loading rate; however, once the MBER was adapted to the new organic loading rate, the COD removal efficiency could be substantially improved, indicating a strong response and capacity of the MBER for organic variation and removal. Those results also indicate that recirculation is important to COD removal, especially at a higher COD loading rate (shorter HRT) that requires better substrate distribution. The turbidity of the membrane permeate was 1.0 ± 0.7 NTU, within the range of the permeate turbidity from an MBR (typically lower than 1 NTU). It was observed that the microbial growth inside the tubing used to collect the permeate affected the turbidity measurement.

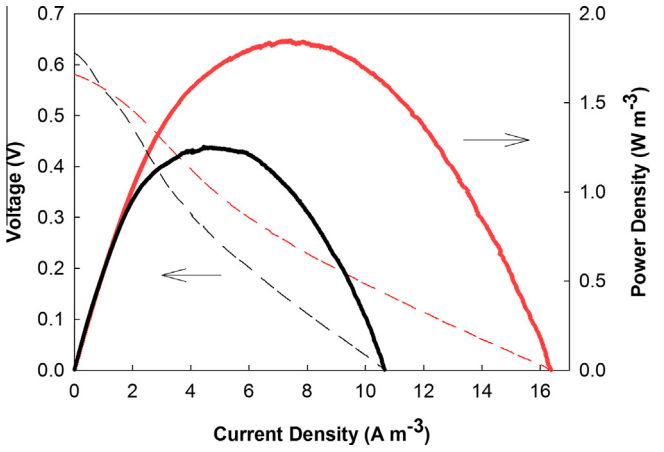

Fig. 3. The polarization results for the MBER: with fluidized GAC (red line), and without GAC (black line). Solid lines are for power output and dashed lines are for voltage. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

The TMP of the hollow-fiber membranes remained below 30 kPa during the operation, and was influenced by HRT and/or anolyte recirculation rate (Fig. 2B). Adjusting HRT will change organic loading rate and water flux through the hollow-fiber membranes, both of which could significantly affect TMP. Increasing organic loading rate at a shorter HRT would bring more organic/inorganic compounds into the reactor and increased the chance of adsorption of those compounds by hollow-fiber membranes, thereby accelerating membrane fouling and increasing TMP. Increasing water flux with a lower HRT will demand a larger pressure difference across hollow-fiber membranes and thus increase TMP. In more details, decreasing HRT from 24 to 8 h when the recirculation was kept at 800 mL min^{-1} resulted in TMP fluctuation between 5 and 10 kPa. On day 33 (arrow e in Fig. 2B), the recirculation was turned off and it was observed that TMP started to increase to above 25 kPa in the following 20 days; when the recirculation was restarted on day 76 (arrow f in Fig. 2B), the TMP quickly dropped to 10 kPa. Those results suggest that the recirculation of anolyte and thus fluidized GAC was a key factor to maintain a low TMP and control membrane fouling/scaling, which confirms the findings in the previous study (Kim et al., 2011). The fluidized GAC removed the deposits from the surface of hollow-fiber membranes via abrasion, thereby reducing membrane fouling and decreasing TMP. However, it should also be noted that the long-term and/or strong abrasion could damage the membrane. Further decreasing the HRT to 5 h (water flux of 6.67 L m^{-2} h^{-1}) on day 93 (arrow g in Fig. 2B) elevated the TMP to 20 kPa, because of a higher water/organic flux.

3.2. The MFC+MBER system

The MBER was hydraulically connected to an MFC for treating an actual industrial wastewater. This coupled system showed great performance in contaminant removal (Fig. 4). The MFC acted as a major removal process, in which the TCOD was reduced from 559 ± 172 mg L^{-1} to 228 ± 34 mg L^{-1} representing removal efficiency of 59.2%. The MBER further decreased the TCOD to 28 ± 11 mg L^{-1}, thereby achieving a total removal efficiency of 95.0% by the coupled system. The suspended solids, both TSS and VSS, were largely reduced in the MFC, which decreased the TSS and VSS concentrations from 115 ± 142 and 108 ± 148 mg L^{-1} to 23 ± 8 and 13 ± 8 mg L^{-1}, respectively, representing 80.0 and 87.9% of reduction. The final TSS and VSS in the MBER permeate were 6 ± 8 mg L^{-1} and 3 ± 3 mg L^{-1}, respectively. The concentrations of suspended solids in the permeate were higher than expected, possibly because of microbial growth in the tubing that collected water from hollow-fiber membranes. The turbidity of the membrane permeate was 1.6 ± 0.8 NTU. The coupled system had negligible ammonia and nitrate removal, related to the low concentrations of those compounds (NH_4^+-N, 2 ± 1 mg L^{-1}, and NO_3^--N, 1 ± 0 mg L^{-1}) in the wastewater sample. The nitrite concentration decreased from 26 ± 31 mg L^{-1} to 4 ± 3 mg L^{-1}, likely due to denitrification process occurred in the anodes of the MFC and the MBER. The significant removal of COD and suspend solids by the MFC could help to alleviate the treatment burden on the MBER and thus benefited membrane filtration. During the operation of the coupled system, the TMP of the MBER was around 37 kPa, resulting from the actual wastewater that had a more complex composition than the synthetic solution.

The coupled system successfully generated electricity and theoretically achieved a slightly positive energy balance. The MFC was a major energy producer in the system, with a current density of 13 A m^{-3} (Fig. 5), and energy recovery of 0.036 kWh m^{-3} or 0.109 kWh kg COD^{-1}. The MBER produced a much lower current density of 7 A m^{-3}, and its energy recovery was lower as well, at 0.011 kWh m^{-3} or 0.060 kWh kg COD^{-1}. The energy performance of the coupled system was summarized in Fig. 6, and the energy balance was analyzed by comparing energy production and consumption in both the MFC and the MBER. The total energy recovery per unit of the treated wastewater was 0.047 kWh m^{-3}. Energy was consumed by the pumping system including feeding, recirculation and permeation. Assuming energy conversion efficiency from electrical to pump was 61.2%, the total energy consumption was 0.046 kWh m^{-3}, of which the MFC consumed 0.003 kWh m^{-3} and the MBER used 0.043 kWh m^{-3}. The ratio of energy production/consumption is 1.02, indicating a theoretically neutral energy balance.

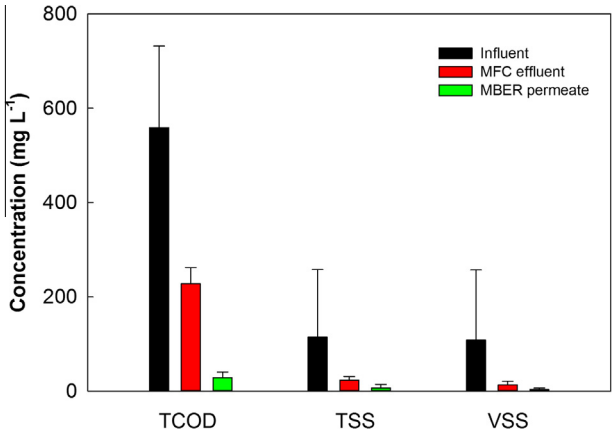

Fig. 4. The contaminants removal from cheese wastewater by the coupled MFC–MBER system. TCOD: total COD; TSS: total suspended solids; VSS: volatile suspended solids.

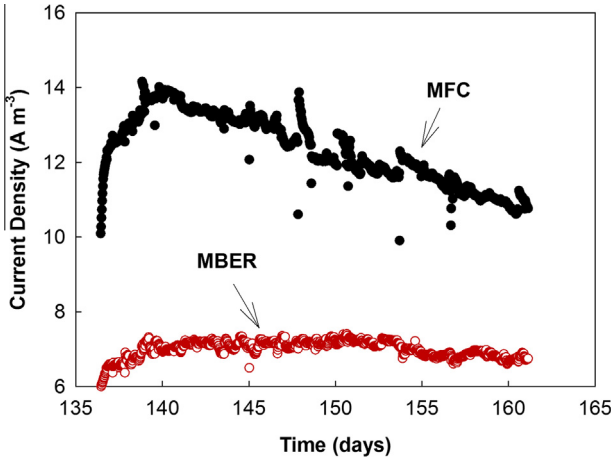

Fig. 5. The current generation by the individual unit in the coupled MFC–MBER system.

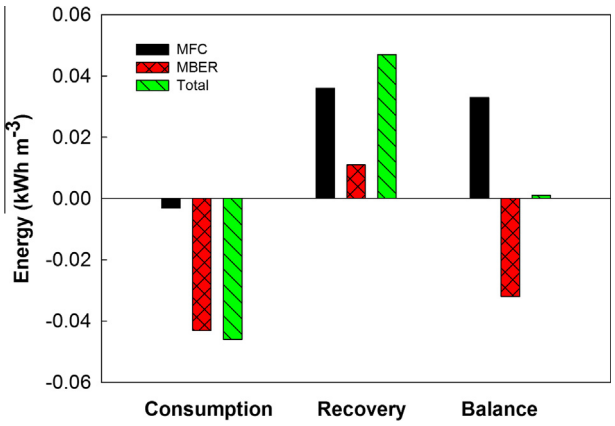

Fig. 6. The energy analysis of the individual unit and the overall coupled MFC–MBER system.

The results of both contaminant removal and energy performance demonstrate different role of each unit in this coupled system: the MFC is the major treatment process with most energy recovery, and the MBER functions as post-treatment to improve the effluent quality. The performance of the MFC is critical to the successful operation of the MBER, in the aspects of contaminant removal (to reducing membrane fouling) and energy recovery (to offset energy consumption). More than 90% of energy consumption was due to the MBER operation, especially the anolyte recirculation. Therefore, to make the couple system more energy positive, it is necessary to reduce energy consumption by recirculation through further evaluating fouling condition and recirculation rates.

Despite the successful proof of concept with a bench-scale system in this study, one must also note the challenges and limitations with further development of the coupled system for practical application. For example, the effect of fluidized GAC on membrane surface properties should be further investigated, because GAC may damage hollow-fiber membranes through long-term abrasion. Proper control of the anolyte recirculation (and thus fluidized GAC bed) is a key factor to minimize the abrasion, with simultaneous effects on fouling control and energy consumption; the interaction among those effects will be critical to

the successful operation of the fluidized MBER. The present study had two reactors (MFC + MBER) linked only through the anolyte stream; a practical operation of the coupled system will also need to consider the catholyte, which could be the treated anolyte; in that way, hydraulic coordination between the two reactors will be very important to maintain healthy operation and achieve satisfactory performance. Nutrient removal has not been well addressed in the present study; with aerobic cathodes that receive the treated anolyte, ammonia may be removed via nitrification and this should be further studied for its effects on cathode oxygen reduction and electrode fouling. The credibility of the energy analysis with a bench-scale system must be further verified with stepwise development of the proposed system, because of significant difference between bench- and large-scale systems. However, the energy analysis presented here exhibits the promising advantage of the coupled system, and has important implications for further studies; for example, the results of different role of each reactor in the couple system can guide the development of large-scale treatment systems, and the amount of MFC modules in the coupled system should be more than that of MBER modules, because of MFCs' major function in contaminant removal and energy recovery.

4. Conclusions

This study has demonstrated a new bioelectrochemical system through incorporating fluidized GAC into an MBER. Membrane fouling was effectively controlled with a low TMP. However, the function of the GAC as a part of the anode electrode was not proved. When coupling this MBER with an MFC, the treatment of actual wastewater was improved compared with a standalone MBER, and the overall energy balance was neutral. The coupled system presents an example of accomplishing low energy consumption and high-quality effluent for wastewater treatment. Those results encourage further research to address several key challenges such as economical feasibility and scaling up.

Acknowledgements

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