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Simultaneous Congo red decolorization and electricity generation in air-cathode single-chamber microbial fuel cell with different microfiltration, ultrafiltration and proton exchange membranes

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ABSTRACT

Different microfiltration membrane (MFM), proton exchange membrane (PEM) and ultrafiltration membranes (UFMs) with different molecular cutoff weights of 1 K (UFM-1K), 5 K (UFM-5K) and 10 K (UFM-10K) were incorporated into air-cathode single-chamber microbial fuel cells (MFCs) which were explored for simultaneous azo dye decolorization and electricity generation to investigate the effect of membrane on the performance of the MFC. Batch test results showed that the MFC with an UFM-1K produced the highest power density of 324 mW/m^2 coupled with an enhanced coulombic efficiency compared to MFM. The MFC with UMF-10K achieved the fastest decolorization rate (4.77 mg/L h), followed by MFM (3.61 mg/L h), UFM-5K (2.38 mg/L h), UFM-1K (2.02 mg/L h) and PEM (1.72 mg/L h). These results demonstrated the possibility of using various membranes in the system described here, and showed that UFM-1K was the best one based on the consideration of both cost and performance.

1. Introduction

As a novel technology, microbial fuel cells (MFCs) have been explored to enhance the degradation of some recalcitrant pollutants with an instantaneous electricity generation (Luo et al., 2009; Morris et al., 2009; Zhang et al., 2009). The application of air-cathode single-chamber MFC which uses oxygen in air as cathodic electron acceptor substantially reduced the internal resistance and additional energy consumption and was shown to represent a simpler design with cost savings (Liu et al., 2003). Previous works in our laboratory have demonstrated that rapid decolorization of azo dye coupled with power generation can be achieved in such an air-cathode single-chamber MFC (Cao et al., 2010; Sun et al., 2009b).

Membrane is utilized to physically separate the anodic and cathodic chambers (or cathode for a single-chamber MFC). As an optional component, membranes affect both the performance and cost of MFCs. The disadvantages of membranes in MFCs are their high cost and diminishment of system performance. For example, the price of a proton exchange membrane (PEM) is $ 1400/m^2, and the power density substantially decreased when used in air-cathode single-chamber MFCs (Liu and Logan, 2004). However, the absence of a membrane could also cause several problems. One major constraint is the increased oxygen and substrate diffusion, which can lower the Coulombic efficiency and bioelectrocatalytic activity of the anode microorganisms (Kim et al., 2007; Liu and Logan, 2004). Another problem is that it makes cathode directly exposure in contaminated environment of anode, which leads to a rapid biofouling/deactivation of cathode catalyst and deteriorates MFC performance (Tartakovsky and Guiot, 2006). This is especially true for the long-term treatment of complex wastewaters. In addition, the increased oxygen diffusion from cathode into anodic chamber after omitting a membrane is not helpful for the bacterial decolorization of azo dye, since high rate decolorization of azo dyes are usually achieved under anaerobic condition (Mu et al., 2009). Therefore, how to overcome these challenges is the highest priority for the application of air-cathode single-chamber MFC for simultaneous azo dye decolorization and bioelectricity generation.

Recently, a variety of membranes which are widely applied in water purification and wastewater treatment industry have been extensively explored for MFCs, including cation exchange membrane, anion exchange membrane, bipolar membrane, microfiltration membrane (MFM), and ultrafiltration membrane (UFM) (Kim et al., 2007; Rozendal et al., 2007; Sun et al., 2009a; Ter Heijne et al., 2006; Zuo et al., 2008). These membranes are convenient

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for the diversified design and application of the MFC due to their easily availability, low cost and versatility.

In our previous study, MFM has been instead of PEM and was used in an air-cathode single-chamber MFC for simultaneous azo dye decolorization and electricity generation (Cao et al., 2010; Sun et al., 2009b). Although the MFM is efficient for isolating cathode catalyst from the anodic microorganisms and suspended pollutants but it still runs a risk for the degeneration of the cathode catalyst due to its high permeability to soluble pollutants presented in anodic chamber such as azo dye and its broken down products (Sun et al., 2009a).

Ultrafiltration membranes (UFMs) are also widely applied in wastewater treatment, which were selected to separate organic matters from water based on their different molecular cutoff weights and have been successfully used in MFCs to reduce oxygen diffusion and substrate fluxion (Kim et al., 2007). In addition, UFM can be tailored in pore size and dimension which can meet various application requirements of MFCs.

In present study, effect of membrane types on performance of air-cathode single-chamber MFC for simultaneously azo dye decolorization and electricity generation was firstly investigated. We intended to investigate the UFMs with different molecular cutoff weights of 1 K (UFM-1K), 5 K (UFM-5K) and 10 K (UFM-10K) on the performance of the air-cathode single-chamber MFC through comparison with PEM and MFM. A representative azo dye of Congo red was selected as model azo dye. Permeability to oxygen and substrate, Coulombic efficiency, power density, and Congo red decolorization for all of the tested membranes were detailedly examined and the most suitable membrane was accordingly addressed.

2. Methods

2.1. Dye

Congo red ($C_{32}H_{22}N_6O_6S_2Na_2$, analytical grade) was manually prepared to the chosen concentrations with deionized water, which was purchased from Damao chemical reagent plant of Tianjin, China and used as received without further purification.

2.2. Microorganisms and medium

A mixture of aerobic and anaerobic sludge (1:1, v:v), collected from the Liede municipal wastewater treatment plant, Guangzhou, China, was used as the original anodic inoculums of the MFCs. The sludge was added into MFCs with a final concentration of 2 g of volatile suspended solids per liter. Before inoculation, the sludge was washed three times using deionized water to remove soluble carbon sources and was filtered through a 0.25 mm pore size sieve to remove impurities. Congo red (300 mg/L) and glucose (500 mg COD/L) as the mixed fuels were used in the experiments. The medium used for the MFCs anodes contained (per liter of deionized water): 5.544 g $NaH_2PO_4 \cdot 2H_2O$, 23.084 g $Na_2HPO_4 \cdot 12H_2O$, 0.62 g NH_4Cl, 0.26 g KCl, 12.5 ml trace metals solution and 5 ml vitamin solution as reported by Lovley and Phillips (1988).

2.3. MFC construction and operation

Air-cathode single-chamber MFCs were constructed as previously reported with a slight modification in dimension (Sun et al., 2009a). Total volume of the MFC was reduced to 512 mL (8 × 8 × 8 cm in height with an operating volume of approximately 400 ml plus a 112 ml headspace). Anodes were made of non-wet-proofed porous carbon papers (no catalyst) with a projected surface area of 3 cm × 3 cm on one side, while cathodes were similar to the anodes in dimension and prepared by coating 0.5 mg/cm^2 of Pt on wet-proofed porous carbon papers. The PEM-cathode MFC was constructed by binding of PEM (Nafion 117, Dupont Co., USA) on the surface of the air cathode based on published procedure (Liu et al., 2005). The MFM-cathode MFC and UFM-cathode MFC were constructed by directly applying the MFM and UFM onto the water-facing side of the cathode. For the UFM-cathode MFC, different molecular cutoff weights of UFM-1K, UFM-5K and UFM-10K were applied to individual MFC, respectively. MFM and UFMs were stored in deionized water and gently rinsed prior to use. The entire cathode was covered with a thick plexiglass cover with holes to allow oxygen to reach the cathode.

All MFCs were operated in a batch-fed mode at a fixed load of 1000 Ω and mixed by a magnetic stirrer to enhance mass transfer. At the beginning of each batch test, the reactors were flushed with nitrogen gas for 15 min to remove dissolved oxygen and maintain anoxic conditions in the anodic chamber. All the MFCs were initially fed with glucose containing medium solution alone (500 mg COD/L). The medium solution was refreshed when the voltage decreased to 50 mV and the suspended biomasses were reserved, forming a complete fed-batch cycle. Once reproducible maximum voltages were observed, the substrate in the anodic chamber was switched to a mixture of Congo red (100–300 mg/L) and glucose (500 mg COD/L). MFCs were operated for another several cycles until the reproducible maximum voltages were observed again to make the system fully acclimate to the dye. Experiments were conducted in a constant-temperature room (30 ± 1 °C) and the average value was reported for all data.

2.4. Analytics and calculations

The substrate diffusion coefficients (Ds) and oxygen diffusion coefficients (Do) were measured and calculated as described by Kim et al. (2007). Dissolved oxygen (DO) was measured using an YSI Model 550A meter (Yellow Springs Instruments, Yellow Springs, USA).

An UV–visible scanning spectrophotometer (BECKMANDV640) was used for the measurements of Congo red concentrations at 496 nm and recording of UV–visible absorption spectra over a wavelength range from 200 to 600 nm.

Cell voltages across the resistance were recorded every 660 s using a multimeter and a data acquisition system (Model 2700, Keithly Instruments, USA).

Electrode potentials were measured using Ag/AgCl reference electrodes (0.197 vs. normal hydrogen electrode; NHE).

Power density normalized by the anode surface (mW/m^2) and coulombic efficiency were calculated as previous described (Sun et al., 2009a).

Azo dye diffusion through the different membranes was tested by applying membrane as a separator in a two-chamber MFC in which one chamber was filled with 300 mg/L of Congo red and another chamber was filled with deionized water. Sample was regularly obtained from the deionized water containing chamber for UV–visible absorption spectra measurement.

3. Results and discussion

3.1. Diffusion coefficients based on membranes

As presented in Table 1, the Ds and Do for the MFM were 1.02×10^{-6} cm^2/s and 9.96×10^{-6} cm^2/s, respectively, which were higher than those measured for other membranes. The PEM had the lowest Ds (1.47×10^{-11} cm^2/s) and Do (3.89×10^{-6} cm^2/s) and the three UFMs have relatively intermediate values. These results showed that the Ds and Do are directly proportional to the

Table 1
Mass transfer and diffusion coefficients of oxygen and substrate measured for each membrane. MFM, microfiltration membrane; PEM, proton exchange membrane; UFM-10K, UFM-5K and UFM-1K, ultrafiltration membrane with molecular cutoff weights of 10, 5 and 1 K; Ko, mass transfer coefficient of oxygen; Do, diffusion coefficient of oxygen; Ks, mass transfer coefficient of substrate; Ds, diffusion coefficient of substrate.

Membrane property	PEM	UFM-1K	UFM-5K	UFM-10K	MFM
Thickness (cm)	0.019	0.04	0.04	0.04	0.02
Ko (cm/s)	2.05×10^{-4}	2.11×10^{-4}	2.14×10^{-4}	2.24×10^{-4}	4.98×10^{-4}
Do (cm^2/s)	3.89×10^{-6}	8.44×10^{-6}	8.57×10^{-6}	8.95×10^{-6}	9.96×10^{-6}
Ks (cm/s)	7.72×10^{-10}	6.18×10^{-9}	1.13×10^{-7}	3.27×10^{-7}	5.08×10^{-5}
Ds (cm^2/s)	1.47×10^{-11}	2.47×10^{-10}	4.51×10^{-9}	1.31×10^{-8}	1.02×10^{-6}

pore sizes of membranes. A larger pore size could result in higher Ds and Do. The high value of Do indicates the potential to allow more oxygen into the anode chamber through these membranes per unit time while the high value of Ds will lead to increased diffusion of substrate from anode chamber to cathode surface and simultaneously facilitate overall charge transfer. Thus, both the Ds and Do should be considered during selecting of membranes for diversified application of MFCs.

3.2. Electricity generation by MFCs with different membranes

As showed in Fig. 1, the similar voltages generated with all membranes were observed (441–488 mV) except the UFM-10K (maximum voltage of 331 mV) in the air-cathode single-chamber MFC. The internal resistance were varied over a relative small range (118–212 Ω) for all membranes other than UFM-10K. UFM-10K produced a clearly higher internal resistance of 713 Ω (Table 2). These results demonstrated that all membranes except for the UFM-10K were well suited for the system described here.

Coulombic efficiencies were also varied with the membranes. Table 2 showed that the MFM resulted in a lowest coulombic efficiency of 7.25% while the highest coulombic efficiency of 19.9% was achieved for PEM. For the three UFMs, the coulombic efficiencies increased from 8.3% to 14.5% when the molecular cutoff weights decreased from 10 to 1 K.

A clear difference in power generation was observed for all tested membranes. As can be seen from Fig. 2, the maximum power density was highest for the UFM-1K (324 mW/m^2), followed by the MFM (237 mW/m^2), PEM (186 mW/m^2), UFM-5K (138 mW/m^2), and UFM-10K (54 mW/m^2). These results indicated that variations in power generation during azo dye decolorization are associated with membrane used in MFCs. The membrane-affected differences during electricity generation could attribute to the internal resistances affected by the diffusion coefficients of different membranes. It should be noted that the power densities obtained here are lower than that of previously reported by using an acetate-fed two-chamber air-cathode cubic MFCs with different membranes (Kim et al., 2007). This case was most possibly due to the competition between azo dye and anode for electrons from co-substrate (glucose) or the activities inhibition of electricity-producing bacterial communities caused by azo dye and its broken down products (Sun et al., 2009b).

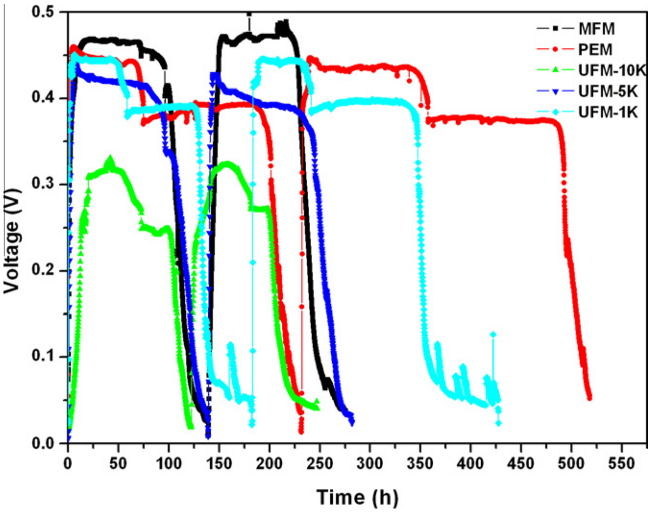

Fig. 1. Voltage output for MFCs with different membranes using Congo red (300 mg/L) and glucose (500 mg COD/L) as co-substrate at an external resistance of 1000 Ω. PEM, proton exchange membrane; MFM, microfiltration membrane; UFM-10K, UFM-5K and UFM-1K, ultrafiltration membrane with molecular cutoff weights of 10, 5 and 1 K.

Although PEM resulted in highest coulombic efficiency, it also significantly diminished power density due to high internal resistance. Compared with PEM, application of UFM-1K achieved both higher power density and coulombic efficiency. The power density produced by using UFM-1K (324 mW/m^2) is 74% higher than that by using PEM (186 mW/m^2).

Because application of UFM-1K and MFM resulted in higher power densities than PEM, the open circuit potentials (OCP) and the electrode potentials of the MFCs with the three membranes were further examined. As showed in Fig. 3, the MFM-cathode MFC had similar cathode working potentials to the UFM-1K-cathode MFC at various currents. However, there were obvious differences in the OCP and working potentials of cathode between UFM-1K-cathode MFC and PEM-cathode MFC. Application of UFM-1K in the cathode of the MFC resulted in an OCP of 0.255 V which is 77% higher than that of using PEM (0.144 V). The difference in potential as observed over a range of working potentials up to a current of 2.3 mA which could be a result of two reasons. First, heating press PEM onto the surface of cathode to bond them together could cause carboxylation and degeneration of the catalyst the cathode, resulting in a less efficient cathode (Liu and Logan, 2004). Second, relative big pore size of the UFM-1K make it more convenient for protons and oxygen diffusion than PEM and thus result in a lower internal resistance and higher power density (Table 1). The cathode potentials of the MFC with UFM-1K obtained here (from 36 to 452 mV versus SHE) remain much lower than the theoretical cathode potential of 804 mV, indicating that further improvement in cathode performance is possible (Logan et al., 2006).

The anode potentials of the UFM-1K and PEM were totally similar. Only a slightly increase in anode potential with PEM was observed when the current up to 1.82 mA which probably due to the fact that the potential is unstable at high current (Gil et al., 2003). However, there were significant differences in the anode potentials between the UFM-1K-cathode MFC and the MFM-cathode MFC. The MFM-cathode MFC produced more positive anode potentials than that of UFM-1K-cathode MFC over the current range of 0–2.7 mA. The potential difference could attribute to the oxygen diffusion, because higher Do of MFM (Table 1) allows more oxygen diffuse from the cathode into the anode chamber which

Table 2
Comparison of the performance of MFCs with different membranes for the simultaneous electricity generation and azo dye decolorization in terms of maximum voltage, maximum power density, internal resistance, COD removal and coulombic efficiency. MFM, microfiltration membrane; PEM, proton exchange membrane; UFM-10K, UFM-5K and UFM-1K, ultrafiltration membrane with molecular cutoff weights of 10, 5 and 1 K.

Membrane	Maximum power density (mW/m^2)	Internal resistance (Ω)	Batch test (1000 Ω resistor)			
			Maximum voltage (mV)	COD removal (%)	Coulombic efficiency (%)	Decolorization rate (mg/L h)
MFM	237	118	488	68.9	7.25	3.61
PEM	186	166	460	61.2	19.9	1.72
UFM-10K	54	713	331	57.4	8.3	4.77
UFM-5K	138	212	441	60	11.7	2.38
UFM-1K	324	101	451	52	14.5	2.02

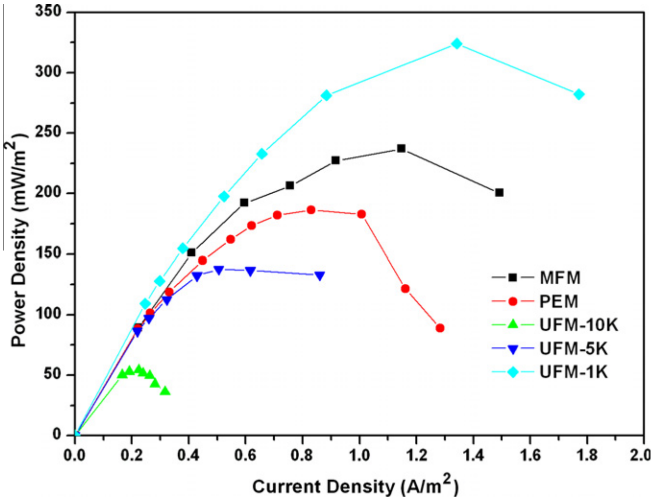

Fig. 2. Power density for air-cathode single-chamber MFCs with different membranes. PEM, proton exchange membrane; MFM, microfiltration membrane; UFM-10K, UFM-5K and UFM-1K, ultrafiltration membrane with molecular cutoff weights of 10 K, 5 and 1.

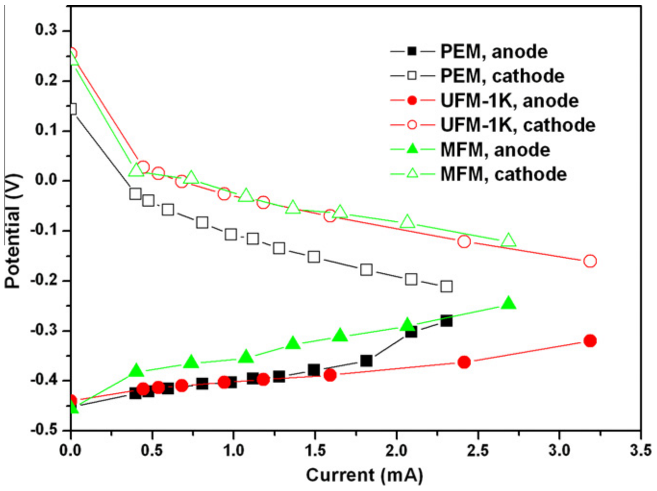

Fig. 3. Electrode potentials (vs. Ag/AgCl reference electrode) for air-cathode single-chamber MFCs with PEM, UFM-1K and MFM. PEM, proton exchange membrane; UFM-1K, ultrafiltration membrane with molecular cutoff weights of 1 K; MFM, microfiltration membrane.

could inhibit the activity of the electricity-producing bacteria and result in aerobic oxidation of substrates. As a result, less electrons are transferred to the anode and the anode potential is elevated. The reasons are also responsible for the lower coulombic efficiency of the MFM-cathode MFC. The high Ds of the MFM could also partially contribute to the lower coulombic efficiency in the MFM-cathode MFC due to increased substrate diffusion from the anodic chamber to the cathode surface. As Harnisch et al. (2009) reported that the diffused organic matters through membranes can be directly oxidized on a platinum cathode, thus providing electrons for oxygen reduction reaction, resulting in the internal short circuit reaction. The continuous electron flow and thus permanent substrate and oxygen turnover on Pt particle may significantly lower the coulombic efficiency of the MFC.

The coulombic efficiencies obtained here using different membranes were much lower than those of in similar MFC systems. In those systems, 40–55% of coulombic efficiencies were achieved (Liu and Logan, 2004). This was most likely due to that there exist a competition between Congo red decolorization and the electricity generation since electrons were all needed in both the processes (Li et al., 2010).

3.3. Congo red decolorization in the anode of MFCs with different membranes

Membrane could be a key factor for the successfully decolorization of azo dye using air-cathode single-chamber MFC. As previous described, a membrane with a high Ds has also a high Do. However, the two coefficients have opposite effects on azo dye decolorization. On the one hand, high Ds could accelerate azo dye decolorization because it results in a lower internal resistance which may increase substrate conversion rate as compared to a high internal resistance during MFC operation (Gil et al., 2003). As a result, more electrons are provided for the reduction of azo dye, and a faster decolorization rate is achieved. On the other hand, the high Do allows increased diffusion of oxygen from the cathode into the anode chamber, which is not favorable for the azo dye decolorization because high rate decolorization of azo dyes are usually achieved under anaerobic condition (Mu et al., 2009). Therefore, how to strike a balance between internal resistance and oxygen diffusion should be a vital procedure for selection of membrane in air-cathode single-chamber MFC used for simultaneous azo dye decolorization and electricity generation.

Decolorization of Congo red (300 mg/L) was investigated in MFCs with different membranes at an external resistance of 1000 Ω using glucose (500 mg COD/L) as a co-substrate. It can be seen from Fig. 4 that a clear effect of membrane type on Congo red decolorization in air-cathode single-chamber MFC was observed. In present work, increase of Ds from 1.47×10^{-11} cm^2/s (PEM) to 1.31×10^{-8} cm^2/s (UFM-10K) by using different membranes resulted in continuous increase of decolorization rate of Congo red from 1.72 mg/L h to 4.77 mg/L h, despite the fact that Do was increased simultaneously. This result indicated that the positive effect of higher Ds exceeded the negative effect of higher Do on Congo red decolorization. However, further increase of Ds to 1.02×10^{-6} cm^2/s (MFM) led to a decrease of decolorization rate from 4.77 mg/L h to 3.61 mg/L h while the Do was increased from

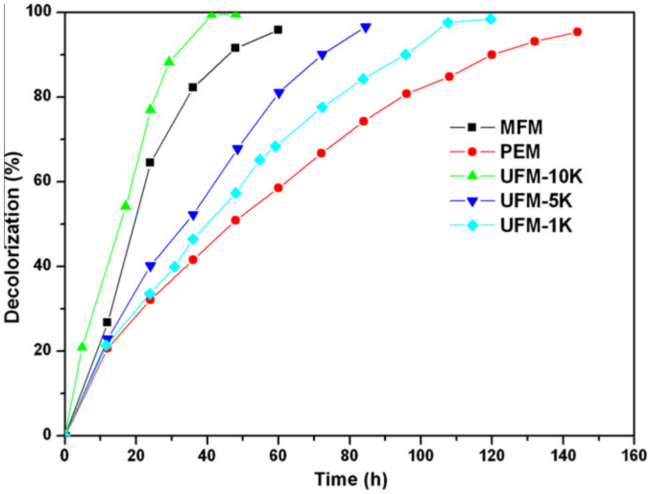

Fig. 4. Decolorization of Congo red (300 mg/L) in MFCs with different membranes using glucose (500 mg COD/L) as co-substrate at an external resistance of 1000 Ω. PEM, proton exchange membrane; MFM, microfiltration membrane; UFM-10K, UFM-5K and UFM-1K, ultrafiltration membrane with molecular cutoff weights of 10, 5 and 1 K.

8.95×10^{-6} cm^2/s (UFM-10K) to 9.96×10^{-6} cm^2/s (MFM). In this case, the negative effect of higher Do began to prevail over the positive effect of higher Ds on Congo red decolorization. Thus, UFM-10K could be the most suitable membrane in terms of Congo red decolorization in this study. The fast Congo red decolorization rate may relate to the low power density in the UFM-10K-cathode MFC. The azo dye could be more competitive than the anode for accepting the electrons from co-substrate (glucose).

During the operation of MFCs, the azo bonds of Congo red were broken using protons and electrons from glucose oxidized by anodic bacteria, resulting in the formation of colorless products, such as aromatic intermediates (Isik and Sponza, 2005; Dos Santos et al., 2006). The removal efficiencies of COD in MFCs with different membranes using Congo red (300 mg/L) and glucose (500 mg COD/L) as co-substrate were ranged from 52% to 68.9% which were much lower than that (more than 92%) in the MFCs using readily degradable compounds as sole fuel (Sun et al., 2009a). This indicated that the decolorization products could not be degraded completely (Sun et al., 2011). Because the bacterial biodegradation of the decolorization products is an aerobic process, the microaerophilic condition resulted from the oxygen diffusion from the cathode into the anode chamber in an air-cathode single-chamber MFC provided a chance for a further biodegradation of the decolorization products (Xu et al., 2007). However, the Do of each membrane was different, so the degree of the further biodegradation of the decolorization products was supposed to be various. Based on the assumption, additional work will be needed to better understand the effect of different membranes on the further biodegradation of the decolorization products.

3.4. Practical application consideration based on membranes in MFCs

In addition to improvements in performance, the continued development of low-cost materials is essential for scale-up of MFCs. As an important component of air-cathode single-chamber MFC, membrane affects not only the performance but also the cost of an MFC. The purchased price of either a UFM ($ 160/m^2) or a MFM ($ 20/m^2) used in this work are much lower than the PEM ($ 1400/m^2). The high cost of PEM coupled with reduction in power output and decolorization rate suggest that it is not suitable for the simultaneous azo dye decolorization and electricity generation in air-cathode single-chamber MFC. Although both UFM and MFM achieved relative lower cost and improved performance compared to PEM, MFM is also not recommended due to its high permeability to soluble pollutants present in the anodic chamber which could run a risk for the degeneration of the cathode catalyst (Sun et al., 2009a). As showed in Fig. S1, the diffusion rate of the Congo red under the application of MFM is much higher than that of other membranes according to the relative peak height of the absorbance peak of Congo red at 496 nm. It can be also evidently observed in Fig. S1 that use of UFM-1K can successfully restrain Congo red diffusion. Although UFM-10K played outstandingly in decolorization, the low power density suggesting that it is not an appropriated choice. UFM-5K gave a mediocre performance in both electricity generation and Congo red decolorization. Thus, giving a comprehensive consideration in terms of cost, power generation and decolorization of Congo red, UFM-1K exhibited the best performance.

4. Conclusions

PEM are not suitable due to its reduction in power output, decolorization rate and high cost. MFM is also not recommended due to its high permeability to soluble pollutants present in anode chamber such as dye and its broken down products. Giving consideration to both power generation and decolorization of Congo red, UFM-1K exhibited the best performance among all of the three UFMs. In addition, UFM represented an order of magnitude decrease in cost compared to PEM. Therefore, the UFM-1K was the best one based on the consideration of both cost and performance in the system described here.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.biortech.2010.12.092.

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