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Enhanced performance and hindered membrane fouling for the treatment of coal chemical industry wastewater using a novel membrane electro-bioreactor with intermittent direct current

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Abstract: A membrane electro-bioreactor (MEBR) embracing biological treatment, electrokinetic phenomena and membrane filtration was established by applying intermittent direct current (DC) to MBR. MEBR exhibited significant improvement of treatment performance and reduction of membrane fouling. COD and total phenols removal efficiencies increased to 83.53% and 93.28% at an exposure mode of 24’-OFF/6’-ON, compared to 71.24% and 82.43% in MBR. Trans-membrane pressure increment rate declined dramatically in MEBR, which was mainly attributed to the increase of sludge floc size and decrease of zeta potential, soluble microbial products and specific resistance to filtration, resulted from electrokinetic effects such as electrocoagulation, electrophoresis, electroosmosis and electromigration of ions. It was notable that DC exposure exerted distinct evolution on microbial community, with the improvement of microbial community richness and diversity. The relative abundances of functional genera were promoted noticeably in MEBR. An interactive relevance existed among microbial community structure, mixed liquor properties

and operational parameters.

Keywords: membrane electro-bioreactor; membrane fouling; electrokinetics; mixed liquor properties; microbial community; relationship analysis

1. Introduction

New coal chemical industry has been rapidly booming in China due to the stresses of environmental pollution and energy shortage according to the national energy development strategy, which is a comprehensive utilization of coal resource in a cleaner way. However, the coal gasification process, an indispensable procedure in coal chemical industry, discharges enormous amount of high-strength wastewater containing various of toxic compounds and a great number of refractory organic and inorganic pollutants such as phenolic compounds (main organic pollutants), nitrogen-containing heterocyclic compounds (NHCs), polycyclic aromatic hydrocarbons (PAHs), long chain hydrocarbons, ammonia, cyanide and thiocyanate et al (Xu et al., 2016). The high-strength wastewater has posed great pollution risk to ecological environment due to its toxicity and refractory, especially without appropriate treatment (Hou et al., 2015; Xu et al., 2018). Membrane bioreactor (MBR) integrates activated sludge process with membrane filtration and has been applied to the treatment of domestic wastewater and various kinds of industrial wastewater as well as wastewater reuse with the advantages of higher mixed liquor suspended solids (MLSS) and organic pollutant loading, superior effluent quality, independent control of hydraulic retention time (HRT) and solids/sludge retention time (SRT) et al (Barreto et al., 2017; Chen et al., 2017). The conventional MBR refers to aerobic MBR. With the development of MBR technology, anaerobic MBR (AnMBR) emerged for the complement of membrane biological system. AnMBR allows energy recovery by conversion of organic matter into methane-rich biogas during the anaerobic digestion(Martin Garcia et al., 2013; Ruigómez et al., 2017). Both types of MBR have been considered as the preferential available technology with potential application in water and wastewater treatment(Huang and Lee, 2015; Xie et al., 2016; Liu et al., 2018). However, membrane fouling remains the major issue to be solved to ensure the sustainability of this promising technology(Ahmed et al., 2018; Fan et al., 2018). The reduction of flux and rise of trans-membrane pressure (TMP) caused by membrane fouling resulted in the increase of operational costs and deterioration of effluent quality(Jia et al., 2014; Meng et al., 2017). Generally, membrane fouling was classified as reversible (recoverable by physical methods) and irreversible (including irreversible can be removed through maintenance cleaning, irreversible that can be removed through chemical cleaning and irrecoverable fouling) (Ibeid et al., 2013). Factors affecting membrane fouling include membrane properties, influent and sludge characteristics (soluble microbial products (SMP), floc size, colloids et al.) as well as operational conditions (MLSS, SRT, HRT et al.) (Tian et al., 2011). Several approaches had been attempted to hinder membrane fouling (Iorhemen et al., 2017). However, it can be concluded from the attempts that searching for an effective and powerful method to reduce membrane fouling at an acceptable range remains a formidable challenge. Recently DC was introduced to MBR for the enhancement of pollutant removal and fouling reduction (Bani-Melhem and Elektorowicz, 2011; Jiang et al., 2017). Synergic effects worked on the treatment processes in the integrated system. Electrodes were placed in MBR to form electric field as well as involve electrochemical reactions. The activity of most microbes could be promoted by adequate electric field stimulation, leading to the enhancement of treatment performance. Small-size soluble and colloid components were reduced by electrochemical effects to migrate membrane fouling. Some researchers inserted a cathode inside the membrane to form electric field with an anode outside the membrane module, in which the negatively charged extracellular polymeric substance (EPS) were forced to move away from the membrane surface and then effectively attenuated the precipitation of EPS on membrane surface and membrane fouling(Liu et al., 2012). However, this membrane module with built-in cathode was complex and constrained by the module form and adversely affected the integrity of membrane module. In addition, few studies focused on the treatment of industrial wastewater and microbial community dynamics induced by DC in the system were rarely conducted. The relationships among intermittent DC mode, properties of mixed liquor and microbial community dynamics were not clear.

In the current study, membrane electro-bioreactor (MEBR) coupling of MBR and intermittent DC with iron anode was constructed to treat coal chemical industry wastewater. The primary objective of this research was to evaluate the performance of efficiency enhancement and fouling reduction. The variations of sludge mixture properties (such as zeta potential, floc size, SMP, specific resistance to filtration (SRF) et al.) were inspected to analyze the possible mechanism in MEBR. The microbial community in MEBR was investigated preliminary by the high-throughput sequencing technology. Moreover, the relationships among intermittent DC mode, current density, properties of mixed liquor, membrane fouling and microbial community dynamics were analyzed, which was conductive to a deeper understanding of the mechanism and optimization control.

2. Materials and methods

2.1 Materials

Real coal chemical industry wastewater was obtained from the full-scale wastewater treatment facility of a coal chemical industry plant in China, and the wastewater samples used in this study were the effluent of anaerobic treatment process. The wastewater characteristics were as follows: COD 1486.4±102.4 mg/L, BOD_5 253.3±18.2 mg/L, total phenols 233.8±21.2 mg/L, TOC 335.6±22.3 mg/L, NH_4-N 127.2±8.5 mg/L. The inoculum sludge was collected from the A/O tank, with SVI of 88.

2.2 Experimental procedure

A cylindrical Plexiglas bioreactor with the volume of 3.0 L was used as the reaction medium. A hollow fiber membrane module (normal pore size 0.4 μm, effective surface area 0.02 m^2) was positioned at the center of reactor to construct MBR. In MEBR, stainless steel plate and graphite plate were installed inside the bioreactor as anode and cathode. Both electrodes with the same dimension of 3.0 × 3.0 cm were fixed on two plastic brackets with the distance of 10 cm. The electrodes were connected with a DC power supply which was controlled by a timer to achieve intermittent DC. Current density was maintained at 1.33 mA/cm^2. Air was bubbled from diffuser at the bottom of the reactor to supply oxygen and shear stresses on the membrane surface. The effluent from the membrane module was withdrawn via a peristaltic pump at the mode of suction 9.0 min followed by a 1.0 min rest. HRT was maintained at 18 hours. The bioreactors were operated at complete SRT, which was to say, no sludge was discharged from the bioreactors during the whole process except the samplings for the properties measurements of mixed liquor. A MEBR and a MBR (controlled trials) were operated simultaneously. Electrical exposure mode was defined as the ratio of time without current to time with current in a cycle of 30 min. In the trials for relationship analysis, current density varied in the range of 0.5-1.33 mA/cm^2, and exposure mode ranged in 2-5.

2.3 Analytical methods

Floc particle size was measured as the mean particle size diameter (PSD) using the laser diffraction particle size analyzer. Zeta potential was measured with the Zeta Sizer 3000 analyzer. The specific oxygen uptake rate (SOUR) was calculated from the linear regression slope of the DO drop vs. time divided by the VSS concentration in the test chamber (Hou et al., 2014). SRF was calculated with the Carman equation (Formula S1) based on the data of filtrate volume versus time in vacuum filtration test. Proteins and polysaccharides contents in SMP were measured calorimetrically following Dubois et al. (Dubois et al., 1956) and Lowry et al. (Lowry et al., 1951) methods. Microbial community was analyzed via high-throughput sequencing technology using Miseq Illumina followed by DNA extraction and PCR according to the methods in the literatures (Ma et al., 2015; Jia et al., 2016). Membrane fouling rate was evaluated by measuring the TMP increment over time. When TMP increased to >10 kPa, membrane module was disassembled from the reactor and cleaned thoroughly with tap water and then returned back to the reactor. The number of physical cleanings represented the severity of the reversible fouling and was also used to evaluate membrane fouling.

3. Results and discussion

3.1 Treatment performance and organics removal

Treatment performances of MBR and MEBR at various electrical exposure modes were illustrated in Fig. 1. Average COD and total phenols removal efficiencies in MBR were 71.24% and 82.43% respectively during 10 days stable operation. It can be seen that performances in MEBR at all modes were higher than MBR except mode 2, demonstrating that intermittent DC with iron anode enhanced the treatment performance of MBR. Average COD and total phenols removal efficiencies in MEBR at mode 4 increased to 83.53% and 93.28% respectively. At mode 2, COD and total phenols removal efficiencies decreased to

62.61% and 73.19%. Enhancement of DC declined with lower electrical exposure (mode 5, 77.16% for COD, 89.58% for total phenols). Appropriate electrical exposure promoted treatment performance, while excess electrical exposure (mode 2) constrained the effectiveness. Large amounts of iron ion were generated when MEBR run at high electrical exposure, and high concentration and precipitation of iron ion inhibited the activity of microbes in the sludge, thus exerting adverse effect on the treatment performance. Moreover, it was found that pollutant removal efficiency in MEBR exceeded the sum of removal efficiency by MBR and that by single electrochemical effect (only electric field worked). For example, the sum of COD and total phenols removal efficiencies by MBR and single electrochemical effect were 73.62% (71.24%+2.38%) and 86.21% (82.43%+3.78%), lower than that in MEBR at mode 4. The comparison implied that a synergic effect existed between biodegradation and electrochemical effect in MEBR. Performance improvement at appreciate electrical exposure mainly resulted from the changes of the mixed liquor and microbial community in activated sludge. Therefore, changes in the properties of the mixed liquor and microbial community were discussed in the subsequent section. Mode 4 was selected to conduct the following tests as comprehensive consideration of performance, energy consumption as well as electrode dissolution. In addition, less foam was observed in MEBR, which prevented the biomass from reducing due to dehydration of sludge caused by foam, guaranteeing the high treatment performance.

Organic compositions in the wastewater were detected via GC-MS (Table 1). The contents of various organic compounds decreased in the effluents of MBR and MEBR. Phenolic compounds were nearly completely removed, which was consistent with the high removal efficiencies both in MBR and MEBR. Long-chain hydrocarbons and part of nitrogenous heterocyclic compounds were residual in the effluent. Moreover, the contents and number of main organic compounds in the effluent of MEBR were less than that in the effluent of MBR, which was probably attributed to the synergic effect of MBR and electrochemical effects and the variation of microbial community in MEBR. This also verified the enhancement efficiency of MEBR.

3.2 Membrane fouling

TMP variation with operation duration in MBR and MEBR with MLSS of 4500 mg/L was depicted in Fig. 2a. The increase of TMP in MBR and MEBR was similar at the first 25 days. However, the first washing time in MBR was the 33rd day, and that in MEBR delayed to the 47th day. The membrane in the MBR was cleaned 3 times compared to only 1 time for MEBR. Membrane fouling rates (expressed by TMP increment over time) in MBR during each membrane washing were 0.23 kPa/d and 0.55 kPa/d, and that in MEBR was 0.20 kPa/d, exhibiting significant fouling reduction with the addition of intermittent DC. The comparison of TMP variation demonstrated that membrane fouling can be recovered better via physical cleaning in MEBR than MBR. This is mainly attributed to the effect of intermittent DC on microbes and properties of the mixed liquor, which lowered the contents of fouling compounds (SMP, colloidal materials, VSS et al.) contributing to irreversible membrane fouling. Moreover, TMP variation in MBR and MEBR with MLSS of 3000 mg/L was also investigated (Fig. 2b). Membrane washing in MBR was 2 times compared 1 time for MEBR. The comparison of different MLSS suggested that membrane fouling reduction by MEBR with high MLSS (high fouling potential) was more significant than that with low MLSS (low fouling potential). The superiority of MEBR was less pronounced when the sludge had a low fouling potential, though noticeable.

3.3 Changes in the properties of the mixed liquor

Performance improvement and membrane fouling reduction in MEBR were associated with significant changes in the properties of the mixed liquor. It was reported that SMP, sludge floc size, SRF were the key components determining the sludge fouling potential(Ibeid, Elektorowicz et al., 2013). The changes in the properties of the mixed liquor were analyzed, which provide a quantifiable basis for estimating membrane fouling.

Zeta potential determines the capability of microbial flocs to aggregate. The zeta potential of flocs in MBR increased slightly during the operation (average value from -17.6 mV to -24.7 mV), while a significant zeta potential reduction was observed in MEBR (Fig. 3a). The average value decreased to -12.1 mV in MEBR, which was attributed to the Fe ions released from the anode via electrolysis. Fe ions neutralized the negative charge on the solid surfaces and microbial flocs and thereby reduced the repulsive forces between the flocs. Fe ions led to the coagulation of colloidal particles and sludge flocs. This change of zeta potential reflected the possibility of bio-flocculation and the formation of larger floc size, as discussed subsequently.

Floc PSD changed in a way that reflected the impact of DC field on floc morphology and structure. Fig. 3b illustrated the PSD variations in MBR and MEBR. Floc PSD in MBR maintained at 123.6 μm. In contrast, floc PSD increased from 96.6 μm to nearly 157.1 μm, and then decreased and stabilized at 134.1 μm. The initial increase was caused by the electrocoagulation due to the suppression of diffuse double layer as the reduction in zeta potential, during which organic particles formed larger flocs and colloidal solids were reduced. The increase of floc PSD was also corroborated by enhanced movement of the charged solid particles as a result of applying an electrical field. Subsequently floc size recession was associated with the electroosmotic extraction of the tightly bound water from the flocs. Furthermore, the sludge in MEBR with a higher percentage of inorganic solids was likely to have less electrostatic attraction forces with bound water. Moreover, ferric hydroxides flocs or flocs combined with other solids with smaller size shifted the floc size distribution toward lower values. The floc size was likely to stabilize at a certain value based on the average size of the hydroxide complexes and electrical density. The increase in floc size was widely known to hinder membrane fouling (Tafti et al., 2015). Small particles depositing on the membrane surface or inside the voids of the cake layer caused an increased membrane resistance. The bound water within the sludge flocs was one main sludge property affecting its fouling potential. The change in floc size increased floc PSD and declined bound water, thus reduced membrane fouling significantly. In addition, sludge with large floc size, high density was kept from being taken away with foam, guaranteeing the high biomass and treatment performance.

It was evident that a significant reduction of SMP was observed in MEBR compared to MBR (Fig. 3c). SMP in MEBR and MBR at 50th day were 29.24 and 51.37 mg/L respectively. SMP was removed as a result of electrokinetic effects through which the ferric cations neutralized the negatively charged particles. Proteins with high density of negative charge reacted electrostatically with the ferric cations to form stable flocs. SMP was considered as a major component causing membrane fouling (Tian, Chen et al., 2011). Electrokinetic effects in MEBR had considerable contribution to the removal of SMP, which made the sludge and the cake layer on membrane less viscous and cohesive. Thus, the filterability of sludge was improved and membrane fouling was reduced.

A substantial reduction of SRF in MEBR was observed compared to that in MBR (Fig.

3d). SRF of mixed liquor in MBR was around 3.52×10^{13} m/kg, while that in MEBR stabilized at 1.96×10^{13} m/kg. The reduction of SRF was mainly related to the changes of floc size, SMP, flocs bound water resulted from electrokinetic effects such as electrocoagulation, electrophoretic, electroosmosis, ions electromigration et al. SRF was in inverse proportion to the square of particle diameter according to Carman-Kozeny equation (Bani-Melhem and Elektorowicz, 2010). The increase in floc size caused by electrocoagulation in MEBR declined the SRF and minimized membrane fouling. In addition, the reduction of MLVSS/MLSS (from 0.8 to around 0.68) in MEBR also exerted positive effect on attenuating SRF. The reduction of SRF facilitated the following processes for excess sludge treatment. The reduction of MLVSS/MLSS was mainly related to electrocoagulation by the electrolysis on the iron anode. Iron content of the sludge in MEBR was around 5%, compared to that of 0.35% in MBR. The iron flocs changed the water distribution in the sludge, thus improved the dewaterability. The component of easily dewatered iron floc also facilitated the decline of SRF and promotion of dewaterability.

SOUR was a major indicator of microbial activity. Microbial activity expressed by SOUR of activated sludge in MEBR gone up to 12.53 $mgO_2/(g\ MLVSS\cdot h)$ compared to that of 10.68 $mgO_2/(g\ MLVSS\cdot h)$ (Fig. S1), showing an appreciable increase after applying intermittent DC. Though the sludge was a rather complex composition, SOUR could be considered as an indication of maintaining high microbial activity in MEBR. At an appreciate current density and exposure, the improved microbial activity in MEBR translated into amenability to application as a bioelectrical reactor where DC stimulated microbial metabolism and/or pure cultures (Thrash and Coates, 2008).

3.4 Microbial community

Appropriate electric field exerted positive effect on treatment performance by improving microbial activity and promoting microbial community. High throughput sequencing was introduced to investigate the microbial community richness, diversity and structure shift in MBR and MEBR. Some differences of microbial community were observed between MBR and MEBR. Microbial community richness and microbial diversity were both promoted in MEBR as indicate by Chao 1 index and Shannon index (Fig. S2). It was assumed that the differences in microbial community richness and diversity were related closely to the addition of intermittent DC. The larger size and compact sludge floc in MEBR provided a new growth environment, which supplied a core carrier for the growth and propagation of microbes(Xin et al., 2016; Zhang et al., 2017). Thus, the microbial community richness and diversity increased, which enhanced community stability for maintaining high containments removal efficiency and resisting perturbations. A similar finding was observed in a previous study, which showed that the richness and diversity of anaerobic microbial communities were promoted by appropriate electric field stimulation(Zhang et al., 2012). The analysis on genus level was conducted to further identify the function succession of community. Abundance differences were observed, though most core bacterial genera were shared by the two sludge samples (Fig. 4). *Thiobacillus* had been detected as the primary and universal genus in coking wastewater treatment plants (Ma, Qu et al., 2015), and its relative abundance rose from 5.22% in MBR up to 6.32% in MEBR. *Comamonas* was considered as a versatile aromatic degrader for PAHs and NHCs as well as a promising denitrifier and nitrifier under various aeration conditions (Ibarbalz et al., 2013; Jia et al., 2016). Abundance of *Comamonas* in MBR and MEBR were 5.02% and 6.28% respectively. *Thauera*, as a degrader for phenol, methylphenol and indole, increase from 1.53% to 2.61%. *Truepera* was reported in close relation with the biodegradation of indole, and it abundance increased from 2.24% to 3.38%, which was consistent with the indole removal. The abundance increment of these microbes might be the primary reason in term of microbial community for the enhancement of pollutants removals in MEBR. Moreover, the relative abundance of some genera for nitrification and denitrification or participated in nitrogen removal (eg. *Nitrospira*, *Denitratisoma*, *Thermomonas*) rose in varying degrees, which was mainly attributed to the anoxic/anaerobic microenvironment created by the large and compact sludge flocs. This indicated that MEBR had enhanced potential for nitrogen removal at controlled conditions.

3.5 The relationships analyses

The treatment performance enhancement, membrane fouling reduction, changes in the properties of the mixed liquor and microbial community evolution were all resulted from the addition of intermittent DC. The relationships analyses were conducted with ordination method to discern the effects of operational parameters. The relationship between microbial community structure and main operational parameters (electrical exposure mode, current density and MLSS) was illustrated in Fig. 5a. It could be inferred that current density and electrical exposure mode affected the microbial community significantly and MLSS influenced some of the genera. An obtuse angle between current density and exposure mode indicated that they had an adversely interactive impact in regulating microbial community compositions, since current density and exposure mode affected electrical amount in opposite direction. *Thiobacillus*, as the primary genus in coking wastewater treatment plants was affected by electrical exposure mode dramatically. The dynamics of *Thauera* and *Comamonas* were highly associated with current density and exposure mode. Variation of exposure mode exhibited a positive impact on variations of *Thiobacillus*, *Nitrospira*, *Denitratisoma* and

Thermomonas. Negative correlations emerged between MLSS, current density and *Azoarcus*, *GP4* and *Isosphaera*. The fact that microbial community was positively or negatively affected by the operational parameters might be a primary reason for the community shifts. The correspondence analysis between mixed liquor properties and operational parameters suggested that the changes of properties in mixed liquor were highly related with current density and electrical exposure mode (Fig. 5b). Current density and exposure mode exerted different impact on the variations of mixed liquor properties. Zeta potential was associated with electrical exposure mode significantly, since exposure mode determined the amount of positive charge iron ion in the long period operation, thus exerting considerable effect on zeta potential. SMP was influenced by current density with a high significance. PSD and SRF were related with both current density and exposure mode. Membrane fouling depended on the properties of the mixed liquor(Kurita et al., 2016; Abass et al., 2018). A regression model was gained between membrane fouling rate expressed by TMP increment over time and properties of mixed liquor with a high correlation coefficient (Formula S2, R^2=0.95), suggesting that changes in properties of mixed liquor induced by operational parameters (current density and exposure mode et. al) led to fouling reduction or aggravation.

Based on the above results, it can be obtained exactly that an interactive relevance existed among microbial community structure, mixed liquor properties and operational parameters. A thorough and comprehensive understanding of the relationships would facilitate the optimizing control for fouling reduction as well as efficiency enhancement and be conducive to the rapid development of MEBR in domestic and industrial wastewater treatment.

3.6 Long-term performance of MEBR

According to the above results, microbial community evolution in a long period of operation played a vital role in the performance enhancement of MEBR. It assumed that the treatment efficiency of MEBR might be well even as the intermittent DC was cut off. The treatment performance of MEBR with cutting off DC was investigated (Fig. 6). Average COD and total phenols removal efficiencies of MEBR decreased from 83.38% to 78.33% and 93.97% to 87.97% respectively when intermittent DC was cut off. Treatment efficiency of MEBR when DC was cut off was still higher than the sum efficiencies of MBR and the electro-catalytic degradation (71.3%+2.38%=73.68 for COD, 82.47%+3.78%=86.25% for total phenols), which further confirmed the synergic effect between biodegradation and electrochemical effect in MEBR. The results suggested that the functional bacteria enriched by intermittent electric field still exerted dominating effect on pollutants removals, even though the DC was cut off. When the intermittent DC was switched on after a period of cutting off, COD and total phenols removal efficiencies recovered gradually in 4 days, verifying the comprehensive effects of mixed liquor properties and microbial community succession induced by intermittent DC with iron anode on the enhancement of treatment performance. The variations of membrane fouling were also investigated by monitoring the TMP. When intermittent DC was cut off, TMP increment rate of MEBR increased, indicating the exacerbation of membrane fouling without the assistance of electrokinetic effects. However, membrane fouling rate of MEBR (0.42 kPa/d) was still lower than MBR (0.56 kPa/d) when DC was cut off (Fig. S3), resulting from the residual electrokinetic effects. The exacerbation of membrane fouling of MEBR as cutting off DC was mainly associated with the changes in the mixed liquor. When the intermittent DC was switched on again, membrane fouling was hindered accordingly. The long term operation of MEBR with high efficiency also demonstrated the treatment stability and membrane fouling migration of MEBR with intermittent DC.

The study demonstrated that MEBR enhanced the treatment performance and substantially hindered membrane fouling with the employment of DC and electrodes. The analyses of the mechanism and relationships were beneficial to the operation and optimization of MEBR. The net cost increment of MEBR was mainly associated with the DC energy demand and sacrificial anode, which were related to the raw wastewater properties, operating conditions such as current density, exposure mode and HRT as well as the electrode price. The DC energy demand and anode consumption of MEBR were calculated to be 0.036 kWh/m^3 and 15 g/m^3 in the laboratory scale experiments. The precise economic analysis should be conducted in further pilot scale studies. Importantly, the improvement of treatment performance and reduction of membrane fouling slashed the frequency of necessity membrane cleaning and disassembling/assembling operations of membrane module as well as the according costs meaningfully. Thus MEBR exhibited considerable advantages compared to MBR and it could serve as a promising technology in wastewater treatment with potential application. The reactor design should be the main problem to be resolved for the pilot scale application of MEBR due to the complicated electro-biological system. The treatment and disposal of the excess sludge discharged from MEBR should be paid more attention, though it was reported that the dewaterability of the sludge electro-conditioned by membrane electro-bioreactor was improved significantly(Ibeid et al., 2015). In addition, electrode fouling due to the deposition of organic and inorganic sludge components was another aspect needed to be considered.

4. Conclusions

MEBR with iron anode at appropriate exposure mode was found to be an efficient method for hindering membrane fouling and promoting treatment performance, primarily associating with the changes in properties of mixed liquor, such as the increase of floc size and reduction of SMP and SRF, caused by electrokinetic effects. Microbial activity was provoked to be enhanced by suitable current density and exposure. The microbial community evolution promoted by DC exposure also accounted for performance enhancement in MEBR. The understanding of relationships among microbial community structure, mixed liquor properties and operational parameters was beneficial to applying efficient strategies to MEBR.

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975-985.

Table 1 Results of GC-MC analysis for the influent and effluents of MBR and MEBR

Compounds	Raw	MBR	MEBR
Phelols			
Phenol	10.9[a]	0.8	0.5
Hydroquinone	0.5	0.7	0.9
Cathchol	1.3	0.9	0.9
P-Cresol	0.4	0.2	0.3
Phenol,3-ethylphenol	1.6	ND	ND[b]
Phenol,2,3-dimethyl	0.6	ND	ND
Phenol,4-tert-butyl	1.7	0.9	0.8
1,3-Benzenediol, 4,5-dimethyl-	1.6	ND	ND
Phenol,2,4-di-tert-butyl	0.6	ND	ND
2,6-Di-tert-butyl-4-methylphenol	1.5	0.9	0.6
1,3-Benzenediol, 4,5-dimethyl-	1.6	0.5	0.5
Phenol,4-methyl-2-nitro	1.2	0.6	0.5
Phenol,4-bis(1,1-dimethylethyl)-	0.7	0.2	ND
Phenol,2-[(trimethylsilyl)oxy]	ND	0.8	0.9
Phenol,2,6-dimethyl-4-nitro	ND	0.2	0.2
Nitrogenous heterocyclic compounds			
Quinoline	2.6	1.3	0.9
Isoquinoline	2.4	1.5	1.1

6-Amino-2-methyl quinoline	1.6	0.8	0.5
Pyridine	2.1	1.7	1.6
Pyridine,3-methyl-	1.9	1.3	1.1
2-Amino-4-methyl pyrimidine	1.2	0.8	0.7
Indole	1.4	0.2	ND
Indole, 2- methyl	1.1	ND	ND
2-Amino-3-picoline	1.6	0.7	0.6
2- methyl oxindole	1.4	0.3	ND
2,7-Diphenyl-1H-indole	1.2	ND	ND
2-Pyridine-carboxaldehyde	ND	0.6	0.6
3-Pyridine-carboxaldehyde	ND	0.8	0.5
1H-imidazole,1-methyl-2-nitro-	1.1	0.8	0.6
5-Methyl-2-nitro-1H-imidazole	1.2	0.3	ND
Long-chain hydrocarbons			
Undecane	2.0	1.3	0.9
Dodecane	1.2	ND	ND
Hexadecane	0.9	0.6	0.5
Heptadecane	1.2	0.8	0.6
Octadecane	1.1	0.3	ND
Eicosane	0.8	ND	ND
Heneicosanoic	0.6	ND	ND
Docosanoic	1.7	1.2	0.9

Pentacosane	0.8	0.5	0.3
Hexacosane	0.9	0.6	0.5
Octacosane	0.5	0.3	0.3
Nonacosane	ND	0.1	0.1
Polynuclear aromatic hydrocarbons			
Naphthalene	1.9	1.2	0.8
Naphthalene,1,3-dimethyl	1.5	1.1	0.9
Naphthalene, 2,6-Dimethyl	1.6	1.2	0.9
Naphthalene, 1,2,6-Trimethyl	0.2	ND	ND
Naphthalene, 1,2,6,7-Tetramethyl	0.2	0.2	0.3
Esters			
Dimethyl phthalate	1.2	0.7	0.6
Diisobutyl phthalate	0.8	0.6	0.4
Dibutyl phthalate	1.2	0.3	ND
Hexanedioic acid, bis(2-ethylhexyl) ester	0.5	ND	ND
Sum	65.9	28.8	22.3

a. Values represent the relative percentage of total peak area.

b. ND, not detected.

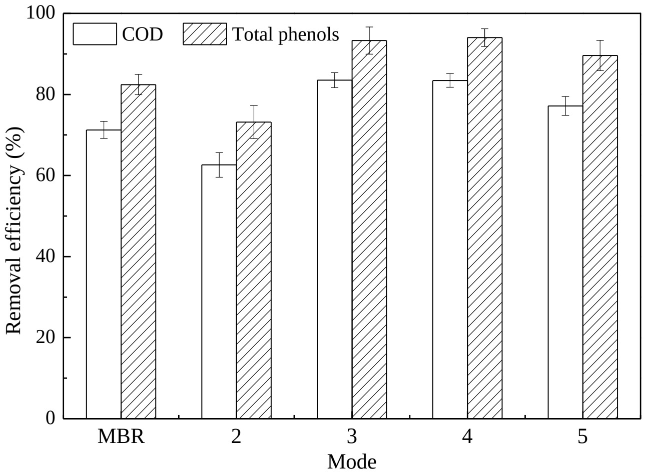


Fig. 1 Performances of MBR and MEBR at various electrical exposure modes (mode 2: 20’-OFF/10’-ON, mode 3: 21.5’-OFF/7.5’-ON, mode 4: 24’-OFF/6’-ON, mode 5: 25’-OFF/5’-ON)

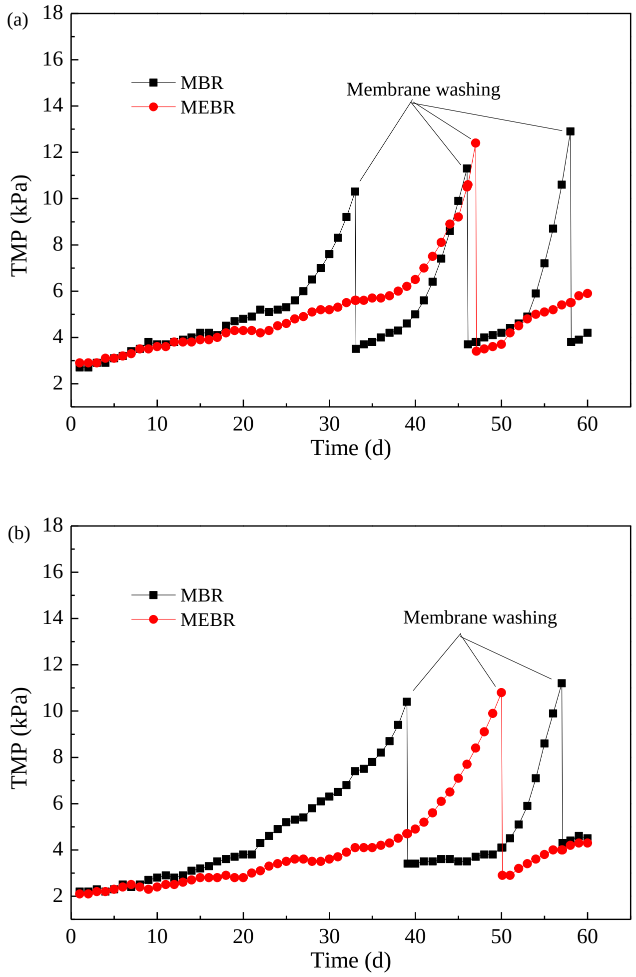


Fig. 2 Variation of TMP in MBR and MEBR (a. MLSS 4500 mg/L, b. MLSS 3000 mg/L)

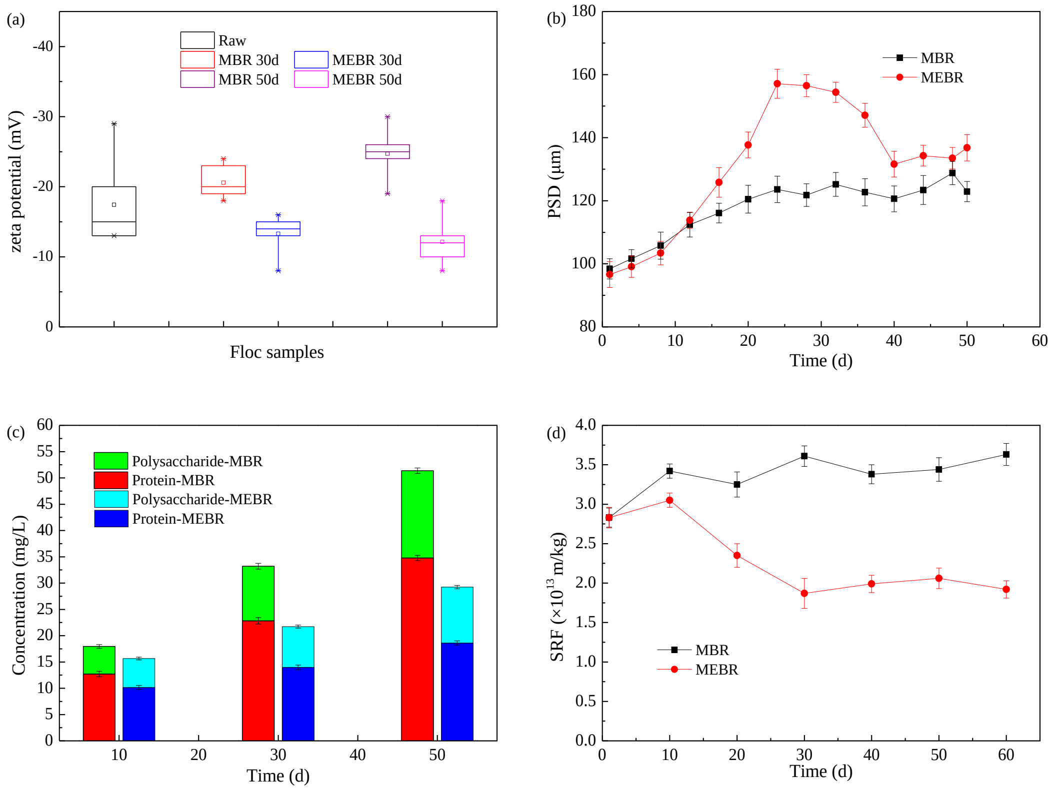


Fig. 3 Changes of mixed liquor properties in MBR and MEBR (a. zeta potential, b. floc PSD, c. SMP, d. SRF)

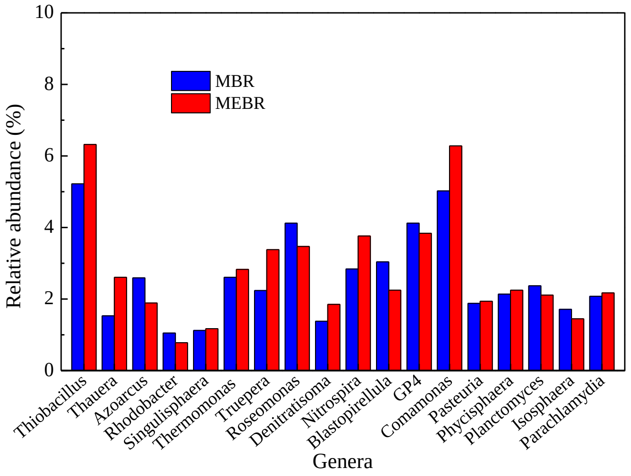


Fig. 4 Evolution of the primary genera in MBR and MEBR

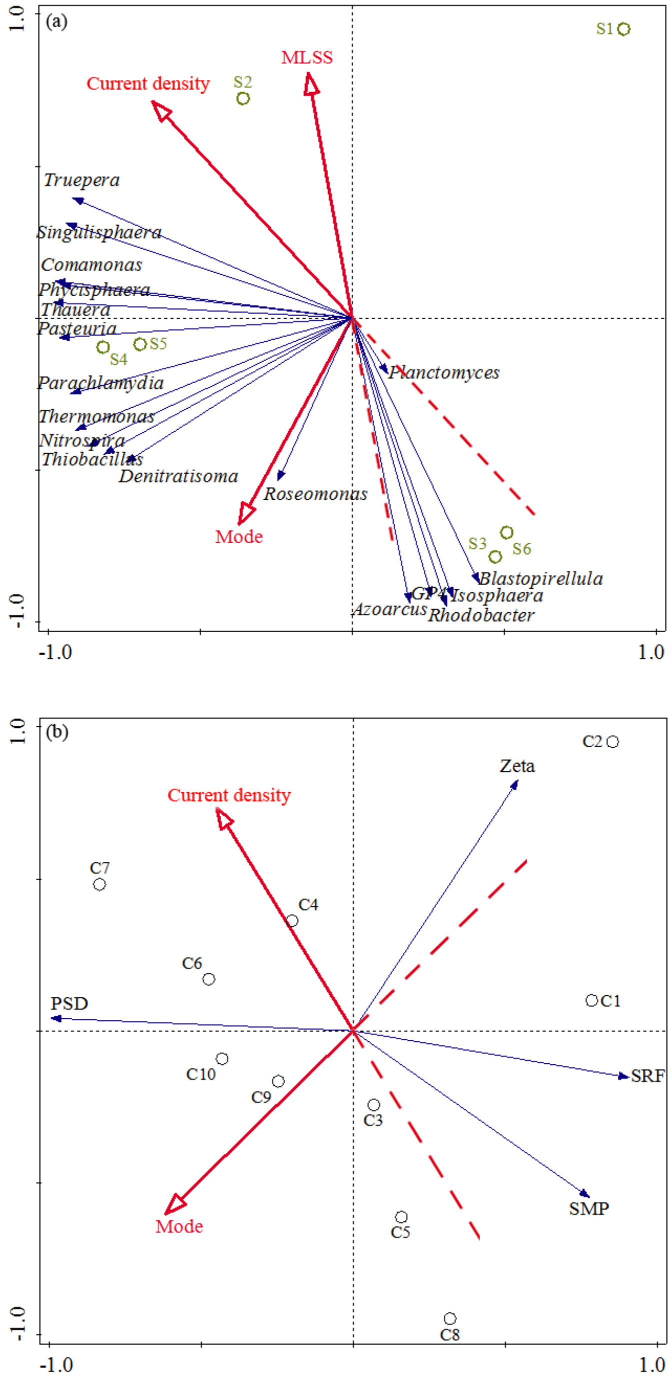


Fig. 5 Principal component analysis (PCA) of the correlations (a. microbial community composition vs operational parameters, b. mixed liquor properties vs operational parameters)

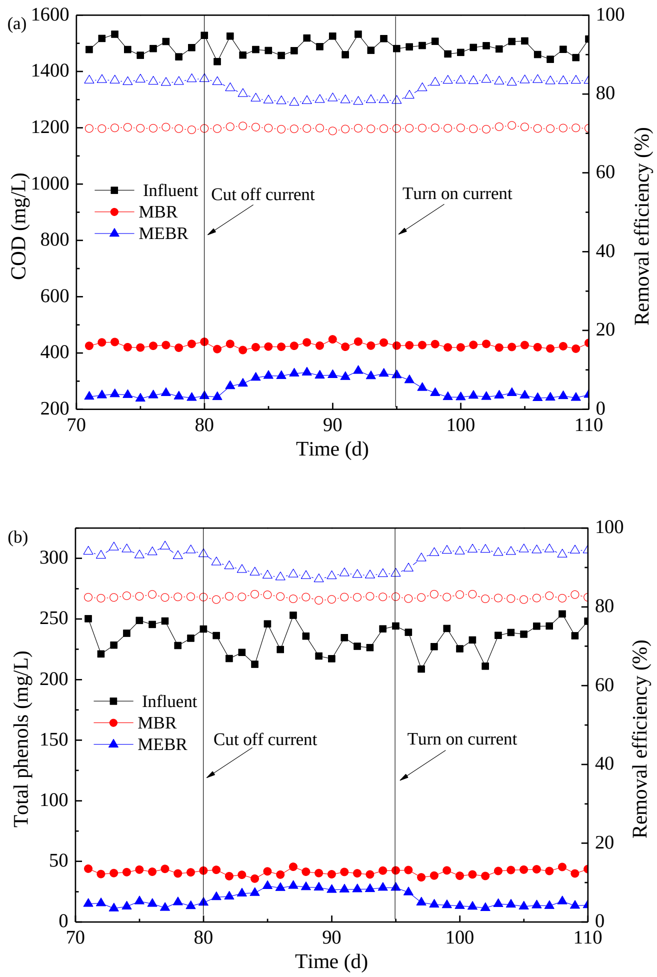
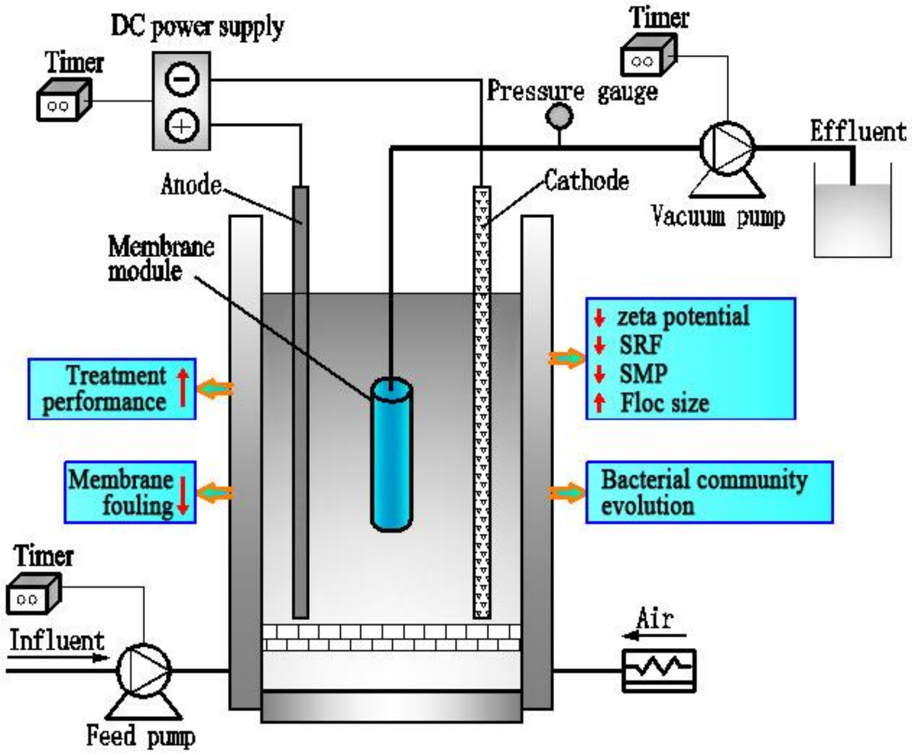


Fig. 6 Long-term treatment performance of MEBR (a. COD, b. total phenols)

Graphical abstract



Highlights

◆ MEBR coupling intermittent DC and MBR was established to treat coal chemical industry wastewater.

◆ MEBR exhibited significant efficiency improvement and membrane fouling reduction.

◆ Changes in properties of mixed liquor caused by electrokinetics accounted for fouling migration.

◆ MEBR with intermittent DC and iron anode promoted microbial community evolution.