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Coal chemical reverse osmosis concentrate treatment by membrane-aerated biofilm reactor system

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ABSTRACT

Coal chemical reverse osmosis concentrate (ROC), which is characterized by high salinity and high organics, remains as a serious environmental problem. In this study, a lab-scale three-stage membrane-aerated biofilm reactor (MABR) system was designed to treat such a ROC. The effects of influent salinity and operating parameters (pH, DO and HRT) on the treatment efficiency were discussed. The removal efficiencies of COD, NH_4-N and TN under the optimal operating parameters reached to 81.01%, 92.31% and 70.72%, respectively. Simultaneous nitrification and denitrification (SND) as well as shortcut nitrogen removal were achieved. The salinity less than 3% did not induce significant decrease in treatment efficiency and microbial communities. Moreover, the dominant phyla in biofilms were *Proteobacteria* and *Bacteroidetes*. This work demonstrated MABR had great potential in ROC treatment.

1. Introduction

Currently, reverse osmosis (RO) technology has been widely used in desalination for over 50% of the installed capacity (Shannon et al., 2008). However, the major drawback of RO is the production of high volume of the RO concentrate (ROC) (Subramani and Jacangelo, 2014) and ROC contains high levels of salt, organic and inorganic contaminants (Sanmartino et al., 2017). Many of the contaminants are toxic and bio-accumulative, which has adverse effects on the environment (Kim et al., 2018; Pradhan et al., 2016).

Various conventional treatment processes have been endeavored to remove the contaminants from ROC such as adsorption (Zhao et al., 2012), electro-oxidation (Wang et al., 2018), membrane distillation (Sanmartino et al., 2017) and integrated technologies (Fang and Han, 2018; Joo and Tansel, 2015). However, the physical and chemical techniques were limited by high operation costs and secondary pollution (Pramanik et al., 2017; Wang et al., 2018). Biological process are high effective and low cost in removing organic and inorganic contaminants, which makes it an attractive, viable and economical option to process ROC (Ersever et al., 2014).

It must be noted that high salinity in ROC could cause high osmotic stress and may reduce microbial metabolic activities, which inhibits the biodegradation of conventional biotechnologies (Maeng et al., 2018; She et al., 2016). However, some halotolerant functional bacteria such as *Proteobacteria* and *Bacteroidetes* have been detected in some adverse environmental conditions (Ou et al., 2018), including saline wastewater (Chen et al., 2017). Indeed, *Proteobacteria* and *Bacteroidetes* play a crucial role in the nitrogen removal and organics degradation (Cleveland et al., 2006; Lydell et al., 2004). Therefore, it is feasible to apply a novel salt-tolerant biotechnology to process ROC.

Membrane-aerated biofilm reactor (MABR) is a burgeoning wastewater treatment technology, in which gas permeable hollow fiber membranes are utilized for biofilm carrier and bubbleless oxygen transfer (Wei et al., 2012). The theoretical oxygen utilization rate can reach 100% (Casey et al., 2015), resulting in less operating costs and volatile pollutants (LaPara et al., 2006). In MABR, oxygen is transferred from the hollow fiber membrane to the biofilm from one side while soluble contaminants in wastewater are supplied from the other side of the biofilm, forming a unique nutrient profile, which allows the formation of a particular biofilm stratification (Syron and Casey, 2008). For example, the oxygen diffused from the hollow fiber membrane causes the one side of biofilm to become aerobic and oligotrophic while the other side of biofilm interfacing with the wastewater becomes anoxic and eutrophic. Consequently, nitrifying bacteria mainly grow near the membrane interface of the biofilm while denitrifying bacteria are predominant near the wastewater interface of the biofilm. Therefore,

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MABR can remove carbon and nitrogen contaminants simultaneously in a single reactor (Casey et al., 1999).

MABR has been applied in extensive wastewater fields, including oil-spilled seawater (Li et al., 2015) and saline domestic sewage (Tian et al., 2017), which proved that MABR is practical for the treatment of saline wastewater. Moreover, a recent study indicated that MABR could remove 79.2% TN from a ROC with low organics and low salinity (TDS 5.5 g/L) (Quan et al., 2018), which preliminarily proved the feasibility of MABR in ROC treatment. However, few investigations were available on the application of MABR for simultaneous carbon and nitrogen removal in highly organic and saline (TDS > 15 g/L) ROC treatment. Furthermore, the influence of water quality and operating parameters on the treatment performance and microbial communities also need more attention.

Generally, membrane fouling is one of the factors affecting treatment efficiency (Jefferson et al., 2000). Biofouling occurs as a result of colonization of membrane surfaces with microorganisms and their productions (Gu et al., 2018). Extracellular polymeric substances (EPS) (Shi et al., 2017a) and soluble microbial products (SMP) (Shi et al., 2017b) appear to be the major source of membrane foulants. As for MABR, membranes fouling could have unfavorable impact on oxygen transfer (Cole et al., 2002). In order to mitigate membrane fouling and improve treatment efficiency, the selection of suitable membrane is also necessary.

In this work, a lab-scale MABR system was designed and constructed to remove carbon and nitrogen contaminants from a coal chemical ROC. Salt tolerance test and parameter optimization were performed to assess the salt tolerance and optimal treatment efficiency of the system. Biofilms were also analyzed to investigate characteristic of the microbial communities. This work was aimed at providing a new attempt and technical support for the application of MABR in ROC treatment.

2. Materials and methods

2.1. Design and configuration of MABR system

Considering the high contents of carbon and nitrogen contaminants as listed in Table 1, it is difficult to completely remove the contaminants in a single reactor. Consequently, a three-stage MABR system was designed and constructed to process the ROC. The system consisted of three reactors, namely, MABR-1, MABR-2 and MABR-3. The three reactors, characterized by different DO extents, were connected in series.

The polymer composite hollow-fiber membrane applied in this work (provided by Hydroking Sci. & Tech. Ltd., Tianjin, China) was dedicated to the MABR process. The high mechanical strength of membrane can effectively avoided rupture or distortion under higher hydraulic shock. Great biocompatibility has been detected, indicating excellent biofouling resistibility (Li et al., 2016). The membrane is validated as suitable for long-term operation with greater stability, antifouling property, oxygen transfer efficiency and contaminants removal performance. The main technical parameters of the membrane are specified in Table 2.

Table 1
General water quality indexes of the ROC.

Water quality index	Unit	Value
Chemical oxygen demand (COD)	mg/L	760–790
Total nitrogen (TN)	mg/L	268–279
Ammonia nitrogen (NH_4-N)	mg/L	65.1
Nitrate nitrogen (NO_3-N)	mg/L	25.9
Nitrite nitrogen (NO_2-N)	mg/L	6.36
Electrical conductivity (EC)	mS/cm	22.9
Total dissolved solid (TDS)	g/L	22.5
pH	–	8.18

Table 2
Parameters of the membrane and membrane module.

Item	Unit	Value
Effective length	m	1.3
Outer diameter	μm	700–780
Wall thickness	μm	70–90
Specific surface area	m^2	0.64
Tensile strength	MPa	49.9
Number	amount	200

As shown in Fig. 1, a hollow-fiber membrane module was installed and submerged in a rectangular container to constitute a bioreactor. The effective volume of each reactor was 6 L, which was designed to better fit the length and width of the membrane modules. The cross-flow velocity across the biofilm, which could improve the mass transfer efficiency, was controlled and regulated by circulating pumps and water distributors. Oxygen in the bioreactor was supplied by an air compressor. The inoculated activated sludge was provided by the secondary sedimentation tank of a wastewater treatment plant (Tianjin, China).

During the experiment, the ROC was pumped into MABR-1, in which the DO value was controlled approximately 0.5 mg/L, providing a favorable condition for the growth of anaerobic bacteria. The biodegradability of ROC could be improved due to the decomposition of macromolecular organics by anaerobic microorganisms, which was beneficial to the subsequent aerobic treatment process. Moreover, the NO_2-N and NO_3-N in ROC could be eliminated by denitrification. The effluent of MABR-1 was pumped into MABR-2. The DO value in MABR-2 maintained 2–4 mg/L to facilitate the growth of aerobic bacteria. The purpose of MABR-2 was to remove most NH_4-N in the effluent of MABR-1. Meanwhile, COD content decreased significantly due to the consumption of short-chain organics by aerobic bacteria. However, NO_2-N and NO_3-N were accumulated in the effluent of MABR-2 since excessive DO inhibited denitrification. Afterwards, the effluent of MABR-2 was pumped to MABR-3 to further remove the residual carbon and nitrogen contaminants. According to the performance of the first two reactors, The DO value in MABR-3 was set at 1–2 mg/L to achieve the simultaneous removal of COD, NH_4-N, NO_3-N NO_2-N and TN.

2.2. Determination of operating parameter extents

In term of operational parameters, pH, DO, HRT and temperature had significant impacts on system performance. It is reported that pH greater than or equal to 10 hindered the EPS biosynthesis due to cell morphological change (Shi et al., 2017a) while pH less than 7 had significant inhibition effect on nitrogen removal (Zou et al., 2016). Moreover, the pH of raw water was approximately 8.0, consequently, influent pH was set at 7.0, 8.0 and 9.0, respectively.

The reactor could operates neither at too low aeration pressure nor too high aeration pressure, because water could intruded inside the membrane at low pressure while the high pressure could hinder the denitrification and result in unnecessary energy consumption. According to previous experience (Li et al., 2016) and purpose of the system, aeration pressure was regulated from 0 MPa to 0.045 MPa and corresponding DO value maintain at 0–4 mg/L to satisfy the different DO demand of each reactor.

Extension of HRT in appropriate range could induce higher treatment efficiency due to sufficient time for the transfer of contaminants from the solution bulk to the biomass (Moussavi and Ghorbanian, 2015). However, excessive HRT could induce reduction of available substrate contents. According to the short-term tracking of effluent water quality, the investigated HRT of single reactor was determined as 18 h, 24 h and 30 h, respectively, which were acceptable in previous work.

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