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Mixed pharmaceutical wastewater treatment by integrated membrane-aerated biofilm reactor (MABR) system – A pilot-scale study

Xin Wei [a,b,c], Baoan Li [a,b,c,*], Song Zhao [a,b,c], Li Wang [a,b,c], Hongyu Zhang [a,b,c], Chang Li [a,b,c], Shichang Wang [a,c]

[a] Chemical Engineering Research Center, School of Chemical Engineering and Technology, Tianjin University, Tianjin 300072, PR China
[b] State Key Laboratory of Chemical Engineering, Tianjin University, Tianjin 300072, PR China
[c] Tianjin Key Laboratory of Membrane Science and Desalination Technology, Tianjin University, Tianjin 300072, PR China

HIGHLIGHTS

- ▶ The design of integrated membrane aerated biofilm reactor system.
- ▶ A pilot-scale study of mixed pharmaceutical wastewater treatment.
- ▶ The effects of aeration condition and circulation flow rate on performance.

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ABSTRACT

A pilot-scale integrated membrane-aerated biofilm reactor (MABR) system, consisted of hydrolysis/acidification pretreatment, MABR process and activated carbon adsorption post-processing, was designed to treat the high-loading mixed pharmaceutical wastewater. A study of MABR process was conducted to investigate the effect of aeration condition, circulation flow rate and water quality on performance over 260 days. The performances of these processes were evaluated by the removal efficiency of COD, BOD_5, turbidity, NH_4^+–N and TN. MABR process could effectively remove above 90% of COD and 98% of ammonia. The capacities per unit volume of MABR could reach up to 1311 gCOD/m^3d and 48.2 gNH_4^+–N/m^3d with single membrane aeration, and the oxygen utilization rate could be as high as 45%. After post-processing, the effluent of integrated treatment MABR system kept stable with COD below 200 mg/L and NH_4^+–N below 3 mg/L.

1. Introduction

In recent years, the treatment of industrial wastewater has become one of the greatest concerns of the Chinese Government, mainly due to the environmental degradation causing by the growth of industry and economy. The pharmaceutical raw materials or bio-pharmaceutical industry wastewater must be treated to satisfy the integrated wastewater discharge standard, which have specific requirements of chemical oxygen demand (COD), ammonium–N (NH_4^+–N), total nitrogen (TN), suspended solids (SS) and turbidity. However, it is always hard to treat pharmaceutical industrial wastewater to the desired discharge standards due to the high COD loading rate and the wide variety of the contaminations which are difficult to be biologically degraded (Enick and Moore, 2007; Oktem et al., 2007; Carballa et al., 2004).

The membrane biofilm reactors (MBfRs) are uniquely suited for numerous treatment applications, including the removal of carbon and nitrogen when oxygen is supplied, and reduction of oxidized contaminants when hydrogen is supplied, such as biofilm membrane bioreactor (BF-MBR) and H_2-based membrane biofilm reactor (MBfR) (Martin and Nerenberg, 2012; Phattaranawik and Leiknes, 2011; Van Ginkel et al., 2011).

The membrane-aerated biofilm reactor (MABR) represents a new technology for wastewater treatment, in which gas permeable hollow fiber membranes are used for bubbleless oxygen transfer and also as the carrier of the biofilm (Gong et al., 2007; Syron and Casey, 2008). A number of laboratory scale studies have investigated the potential of the MABR as a technology for high-efficiency biological degradation (Casey et al., 1999; Syron and Casey, 2008). In the MABR, the membrane lumen is pressurized with either oxygen or air. The oxygen diffuses through the membrane wall without the formation of bubble and is utilized

* Corresponding author at: School of Chemical Engineering and Technology Tianjin University Weijin Road 92#, Nankai District, Tianjin 300072, PR China. Tel.: +86 22 27407854; fax: +86 22 27404496.
E-mail addresses: weixin@tju.edu.cn, baoan.li@gmail.com (B. Li).

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by bacteria in the membrane attached biofilm. Thus, high oxygen-mass transfer efficiency can be achieved, which confers an economic advantage in terms of aeration energy requirement (Li et al., 2010; Brindle et al., 1998). The large specific surface area of hollow fiber membranes can supply high specific area for microbial attachment, and thus the potentially high volumetric reaction rate can be attained due to the high specific biomass concentration (Syron and Casey, 2008). In the membrane-aerated biofilm (MAB), since oxygen and wastewater constituents are supplied from opposite sides of the biofilm, the microorganisms with specific biodegradation could be retained by the acclimation and accumulation for the treatment of special wastewater during the almost infinite SRT (Cole et al., 2002). In addition, the secondary pollution causing by air stripping of volatile organic compounds in conventional bioreactor can be avoided due to the bubbleless aeration in the MABR (Li et al., 2008).

Recently, MABR process has been studied for the treatment of wastewater containing volatile organic pollutants such as acetonitrile, mineralize perchloroethylene and xylene (Li et al., 2008; Ohandja and Stuckey, 2006; Yang et al., 2006). Based on laboratory scale data, the MABR process has the potential to achieve high strength COD and nitrogen removal. However, commercial exploitation of this technology has not yet emerged and until the present there are very limited trials of this technology beyond laboratory scale. Several unresolved problems hampered the implementation of the full-scale MABR, which include (1) the possibility that membrane defects will cause significant process upset; (2) poor understanding of scale-up rules for membrane modules; and (3) inadequate procedure of the provision for enough oxidation capacity and liquid flow distribution (Syron and Casey, 2008; Stricker et al., 2011).

This research work has been carried out as follows to solve the above problems. Firstly, choosing suitable membrane is a critical factor for the long-term stable operation of MABR. Membranes used in MABR can be categorized as microporous hydrophobic membrane, dense membrane and composite membrane (Syron and Casey, 2008; McLamore et al., 2007; Ahmed et al., 2004). Microporous hydrophobic membranes are not applicable to the pilot-scale MABR due to the low operation pressure (below their bubble pressure) and the oxygen transfer deterioration along with the accumulation of water and the colonization of bacteria in membrane pores (Syron and Casey, 2008; Rothemund et al., 1994). The application of composite membranes is limited by their low strength, high cost and unknown long-term durability in the wastewater. Thus, in this work, hydrophobic polypropylene dense membrane with low cost and excellent stability was employed in the pilot-scale MABR process. The intramembrane gas pressure can increase to 0.5 MPa using these membranes. Secondly, curtain-like membrane modules specifically for MABR were designed to avoid short-circuiting and reinforce the load capacity of the membranes. The membrane modules could stably operate for a long time under 0.2 MPa pressure without the leakage. Thirdly, in order to overcome the mass transfer problem, sufficient oxygen supply was ensured by the increases of air flux and pressure, and the addition of fine bubble aided aeration equipments. Liquid flow distribution was improved through the reactor design, which could avoid back-mixing and uneven flow.

Based on our feasible laboratory scale experiment, a pilot-scale integrated MABR system was designed in this work to treat the wastewater from a high-tech pharmaceutical R&D building in Tianjin city of China. The membrane module and system construction were designed and built to solve several technical problems in scale-up of MABR and optimize the operation of integrated MABR system. The aeration condition and circulation flow velocity were adjusted to achieve the optimal performance of MABR.

2. Methods

2.1. Membrane and membrane module parameters

The membrane and membrane module parameters in this study are shown in Table 1. The used hollow fiber membrane was hydrophobic polypropylene dense membrane, which was prepared by melt spinning method from Hydroking Sci & Tech Ltd., (Tianjin, China). This polypropylene dense membrane has excellent aerated ability under high intramembrane air pressure and corrosion resistance with biofilm. The scale-up membrane module was designed as cross flow curtain-like module with the length of 1.6 m and the width of 0.6 m. In this work, the theoretical oxygen supply rate (T-OSR) was measured with varying aeration pressure, according to the method reported in the reference (Terada et al., 2003). T-OSR values at the different intramembrane air pressures are also shown in Table 1.

2.2. The configuration of integrated MABR system

The integrated MABR system was located in the basement of a high-tech pharmaceutical R&D building, and used to treat the mixed pharmaceutical wastewater. The characteristics of the wastewater samples are: COD 2000–3500 mg/L, BOD_5 480–1000 mg/L, BOD_5/COD 0.20–0.39, TN 80–164 mg/L, NH_4^+–N 74–116 mg/L, TP 18–47 mg/L, suspended solids 48–145 mg/L and turbidity 76–138 NTU. The influent temperature was varied from 12 to 26 °C and pH was between 7.2 and 8.5.

The schematic flow chart and assembly drawing of the integrated MABR system are shown in Fig. 1. The system is consisting of a hydrolysis/acidification pretreated pool, MABR, net pool, and activated carbon adsorption tank. Design characteristics of each part of system and hydraulic retention time (HRT) are listed in Table 2.

2.2.1. Hydrolysis/acidification process

Wastewater was collected into the hydrolysis/acidification pool from 15 pipes. A mechanical stirrer was equipped in order to achieve a hydraulic mixing of wastewater and sludge. Hydrolysis/acidification pool has two functions. (1) The fluctuation of COD loading and the concentration of toxic substances could be buffered or diluted. (2) The biodegradability of the wastewater could be improved, which provided a favorable condition for the subsequent aerobic treatment process.

2.2.2. MABR process

As shown in Fig. 1, the MABR reactor was divided into four serially connected pools with the width of 0.6 m, the length of 2.4 m and the height of 1.8 m (run-time water level: 1.5 m). The design

Table 1
Designed parameters of the membrane and membrane module.

Item	Unit	Value	
Hollow fiber length	m	1.4–1.5	
Hollow fiber outer diameter	μm	300–460	
Hollow fiber wall thickness	μm	40–60	
Membrane module	amount	32	
Module pattern		Curtain-like	
Total surface area	m^2	2660	
Specific surface area	m^2/m^3	287	
Packing density (clean membrane)	%	8.4	
Oxygen supply rate (theoretical value)	gO_2/m^2d	0.06 MPa	4.57
		0.12 MPa	7.82
		0.15 MPa	9.58
		0.18 MPa	10.77