Chemical Engineering Journal 295 (2016) 317–325

Contents lists available at ScienceDirect

Chemical Engineering Journal

journal homepage: www.elsevier.com/locate/cej

An innovative of aerobic bio-entrapped salt marsh sediment membrane reactor for the treatment of high-saline pharmaceutical wastewater

Kok Kwang Ng [a], Xueqing Shi [a], Say Leong Ong [a], Cheng-Fang Lin [b], How Yong Ng [a,*]

[a] Centre for Water Research, Department of Civil and Environmental Engineering, National University of Singapore, 1 Engineering Drive 2, Singapore 117576, Singapore
[b] Graduate Institute of Environmental Engineering, National Taiwan University, 71 Chou-Shan Rd., Taipei 106, Taiwan

HIGHLIGHTS

- Entrapped biomass MBRs and attached growth MBRs for pharmaceutical wastewater.
- Salt marsh sediment microorganisms in the MBRs achieved higher TCOD removal.
- Entrapped biomass MBRs could enhance the TN removal.
- Entrapped biomass MBRs had less fouling and produced less EPS/SMP values.
- Low TCOD removal, TMP rose faster and higher EPS/SMP values as HRT decreased.

ARTICLE INFO

Article history:
Received 6 January 2016
Received in revised form 5 March 2016
Accepted 8 March 2016
Available online 12 March 2016

Keywords:
Entrapped biomass
Membrane bioreactor
Membrane fouling
Pharmaceutical wastewater
Salt marsh sediment

ABSTRACT

A novel bio-entrapped salt marsh sediment membrane reactor (BESMSMR) was evaluated for treating high-salinity pharmaceutical wastewater, and membrane fouling behaviour was also assessed. The BESMSMR was operated in parallel with a conventional membrane bioreactor (CMBR), and a salt marsh sediment membrane bioreactor (SMSMBR) as well as an entrapped biomass MBR (bio-entrapped membrane reactor, BEMR) to facilitate a meaningful comparison of performance characteristics obtained in these reactor systems. Two hydraulic retention times (HRTs) of 60 and 40 h with organic loading rates (OLRs) varied from 7.0 to 11.9 kg COD/m^3 d were tested. The pharmaceutical wastewater used has an average total chemical oxygen demand (TCOD) of 17,931 ± 1851 mg/L and total dissolved solids (TDS) of 20,881 ± 2030 mg/L. The BESMSMR demonstrated the highest removal efficiencies of TCOD (78.4–81.3%), due to the ability of the marine sediment microorganisms seeded from coastal shores to thrive in the hyper-saline environment and degraded recalcitrant organic matter present in the pharmaceutical wastewater. The anoxic zone presented in the inner part of bio-carriers could have triggered the denitrification reaction, and thus improved the TN removal rate by 15–20%. Membrane fouling was reduced with lower concentrations of mixed liquor suspended solids (MLSS), extracellular polymeric substance (EPS), and soluble microbial products (SMP) in the entrapped biomass MBRs. Proteins rather than carbohydrates were the main component of EPS and SMP in the MBR systems. The novel BESMSMR possesses the benefits of salt marsh sediment and an entrapped biomass technique that offered effective organic removal for the high-salinity pharmaceutical wastewater and reduced membrane fouling.

1. Introduction

Pharmaceutical industries are characterised by a large number of complex medicine products, manufacturing processes, and plant sizes, as well as the magnitude and diverse quality of produced wastewater. The demand for drugs by people around the world has been increased sharply, principally due to the growth of human populations and the development of medical science for diseases. Therefore, a huge amount of hazardous and toxic pharmaceutical effluent is generated in the pharmaceutical industries, which needs to be treated prior to discharge [1]. As reported by Enick and Moore [2], up to half of the generated pharmaceutical wastewater is released into the aquatic environment without any proper treatment, especially in the developing countries. In addition, most of the pharmaceutical compounds are produced by chemical synthesis that involves several multifarious production processes; this categorizes pharmaceutical wastewater as one of

* Corresponding author. Tel.: +65 6516 4777; fax: +65 6774 4202.
E-mail address: howyongng@nus.edu.sg (H.Y. Ng).

http://dx.doi.org/10.1016/j.cej.2016.03.046
1385-8947/

Nomenclature

AnBEMR	anaerobic bio-entrapped membrane reactor
AnMBR	anaerobic membrane bioreactor
BEH-MBR	bio-entrapped hybrid membrane bioreactor
BEMR	bio-entrapped membrane reactor
BESMSMR	bio-entrapped salt marine sediment membrane reactor
BOD	biological oxygen demand (mg/L)
BSA	bovine serum albumin
CMBR	conventional membrane bioreactor
COD	chemical oxygen demand (mg/L)
DO	dissolved oxygen (mg/L)
EPS	extracellular polymeric substances (mg/L)
EPSc	extracellular polymeric substances (carbohydrate) (mg/L)
EPSp	extracellular polymeric substances (protein) (mg/L)
FBMBR	fixed bed membrane bioreactor
FISH	fluorescence-in-situ-hybridization
HRT	hydraulic retention time (h)
MBMBR	moving bed membrane bioreactor
MBR	membrane bioreactor
MLSS	mixed liquor suspended solids (mg/L)
NaOCl	sodium hypochlorite
OLR	organic loading rate (kg COD/m^3 d)
SMSMBR	salt marsh sediment membrane bioreactor
SMP	soluble microbial products (mg/L)
SMPc	soluble microbial products (carbohydrate) (mg/L)
SMPp	soluble microbial products (protein) (mg/L)
SRT	solids retention time (d)
TCOD	total chemical oxygen demand (mg/L)
TEA	triethylamine
TDS	total dissolved solids (mg/L)
TMP	trans-membrane pressure (kPa)
TN	total nitrogen (mg/L)
TOC	total organic carbon (mg/L)
TSS	total suspended solids (mg/L)
VSS	volatile suspended solids (mg/L)

the complex industrial wastewaters with high chemical oxygen demand (COD), biological oxygen demand (BOD), salinity and toxicity, as well as a low BOD/COD ratio [3,4]. Moreover, pharmaceutical wastewater might contain various amounts of biological substances, solvents, catalysts, cleaning agents and disinfectants, which have severe negative effects on the aquatic environment [4–6].

Numerous studies have reported that pharmaceutical wastewater is mainly treated by physio-chemical and biological processes [4–7]. However, due to the cost-effectiveness of the latter treatment, biological processes such as anaerobic and aerobic systems are still the favoured option for the pharmaceutical wastewater treatment. Several studies have reported that anaerobic processes may not be capable of removing the hardly biodegradable, recalcitrant and refractory compounds present in pharmaceutical wastewater because these compounds could inhibit the methanogenesis and impede the ordinary metabolic function of anaerobes [5,8,9]. For example, Chelliapan et al. [6] reported that the anaerobic bioreactor could only effectively remove 45% of the COD in the pharmaceutical wastewater at an organic loading rate (OLR) of 3.7 kg COD/m^3 d. Ng et al. [9] also reported that the inhibition of methanogenesis occurred with the production of an approximately 164.9 mL CH_4/gCOD of methane, and that a very low COD removal efficiency was achieved by the anaerobic membrane bioreactor (AnMBR) when treating the pharmaceutical wastewater. Therefore, aerobic processes have become an attractive option for pharmaceutical wastewater abatement, as they can achieve more complete oxidation for organic compounds and have shorter start-up periods.

Comparing with other types of aerobic reactors, membrane bioreactor (MBR) has greater solid–liquid separation, higher volumetric loading, greater biomass retention and less sludge production [10–13]. Recently, a few studies have reported that the aerobic MBRs are effective for pharmaceutical wastewater treatment [7,14,15]; however, membrane fouling is always the major drawback of an MBR application. Therefore, many hybrid MBRs have been developed to eliminate the membrane fouling issue, such as the moving bed membrane bioreactor (MBMBR), the bio-entrapped membrane reactor (BEMR), the fixed bed membrane bioreactor (FBMBR) etc. This is because the carriers used in the hybrid MBR systems can not only function as mechanical agents for continuous cleaning of the membrane surface [16,17], but can also be used as a media support for immobilizing bacteria cells [18,19], allowing a high biomass concentration to be entrapped in the carrier with a more compact MBR system. Rafiei et al. [20] compared a bio-film MBR and a bio-entrapped hybrid MBR (BEH-MBR) with a conventional MBR (CMBR) for the treatment of synthesised phenolic wastewater, and found that the BEH-MBR could withstand phenol concentration shocks at 25–50% well and achieved the best phenol removal among these three MBRs. Ng et al. [12] also investigated the membrane fouling behaviour of a CMBR and BEMR, and reported that the BEMR produced less soluble organic products and had seven times less membrane fouling than the CMBR.

There are a number of studies reported on organic removal performance using conventional aerobic sludge for saline wastewater treatment, but the gradual adaption period is an obstacle and usually the treatment performance is limited [21,22]. High salt concentrations have adverse effects on biological treatment, resulting in low treatment performance because the high-saline could possibly cause unbalanced osmotic stress across the cell wall and loss of activity of cells [21,22], leading to plasmolysis and cell death. Consequently, the application of halophilic/halotolerant microorganisms usually remains the best way to enhance the biological treatment of high-saline wastewater. These halophilic/halotolerant microorganisms have a specified enzyme structure enabling them to remain active and stable [23], maintain osmotic balance [24], and extrude Na^+ out of their cells via an internal cell [25]. These characteristics could, in turn, enable the halophilic/halotolerant microorganisms to survive well in the saline environment. To date, there are a few studies reported the performance of halophilic biomasses in pure culture systems for the treatment of high-saline wastewater, which would not be economical and practical for full-scale applications. Our previous investigation by Ng et al. [26] reported that the mixed-culture microorganisms from a marine environment inoculated in an MBR could thrive, and were able to achieve biodegradation when treating the wastewater in high-saline environment. However, the membrane fouling behaviour of the mixed culture microorganisms from the marine environment in the MBR system at a saline concentration greater than 20 g NaCl/L was severe. Furthermore, no studies have investigated the entrapment of mixed marine culture in the MBR system with