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Separation of tetracycline from wastewater using forward osmosis process with thin film composite membrane – Implications for antibiotics recovery

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ABSTRACT

To minimize the risk of antibiotic wastewater generated by the pharmaceutical industries, the potential separation efficacy of tetracycline (TC) from aqueous solution using forward osmosis (FO) process with thin film composite membrane was systematically studied. First, the microstructure and transport properties of TFC membrane were characterized. Then, the effects of membrane orientation, feed velocity and solution pH on the behavior of the FO process for TC separation were studied. Finally, the performance of TFC membrane for TC separation in a long-term FO mode operation was investigated. The results showed that the membrane performance in FO mode (active layer facing the feed solution) and PRO mode (active layer facing the draw solution) was highly affected by solute resistivity (K) value. The water flux and TC rejection achieved over 20 LMH and 99.0% in FO mode, respectively. High TC concentration factor (CF) of 2.6 was obtained in FO mode, indicating the concentrated TC solution could be harnessed to recover the TC by conventional crystallization. However, severe water flux decline accompanied with low tetracycline CF was found in PRO mode, which was mainly attributed to serious fouling and high K value occurred in the porous support. With the flow velocity rising, the shear stress and mass transfer coefficient (k) on the membrane surface increased, alleviating the membrane fouling. Acidic environment would favor the separation due to the change of TC speciation and TFC membrane properties. A long-term testing demonstrated that more than 97% TC rejection and 74% water flux recovery were well maintained with simple hydraulic cleaning after 5 cycles FO mode operation. This work implied that the FO based technology could be developed as an effective alternative for the treatment of tetracycline antibiotic wastewater as well as the recovery of antibiotics from the wastewater.

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1. Introduction

In the past decades, the extensive use of antibiotics for protecting human and animal health, as well as for improving the growth of livestock, has led to their excess accumulation in the environment [1]. In China, the annual production of antibiotics is about 210,000 tons [2]. Tetracycline (TC) antibiotics, including tetracycline, chlortetracycline and oxytetracycline, are the second class of antibiotics in production and usage worldwide, which are ranked first in China [3]. About 25–75% of tetracycline antibiotics are excreted and released in an unaltered form into the environment via urine and feces [4]. The emergence of tetracycline antibiotics in water has drawn a great attention due to the induced antibiotics resistance genes (ARGs), which have seriously jeopardized the human health and the ecological security [5,6].

Due to the large production of tetracycline, the wastewater from related pharmaceutical industries has become a serious pollutant source. The tetracycline antibiotics contaminated waste streams generated in manufacturing plants contains high level concentration of antibiotic from around 10 to 1000 mg/L [7–9]. Thus huge quantity of tetracycline antibiotics wasted in wastewater treatment process. If the tetracycline antibiotics could be reclaimed from wastewater, it would greatly reduce the amount of tetracycline antibiotics for disposal. Nevertheless, the traditional methods for wastewater treatment fail to remove tetracycline antibiotics effectively. To eliminate tetracycline antibiotics, advanced oxidation processes (AOPs) have been applied, including photochemical process, electrochemical process and photocatalytic process [1]. These processes are able to oxidize antibiotics

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by producing hydroxyl radicals from O_3/H_2O_2, UV/O_3 and $UV–TiO_2$. Though the AOPs could directly degrade tetracycline antibiotics, the methods could not recover the antibiotics from wastewater for reuse. Moreover, some AOPs need high operating cost due to the high energy consumption.

With the rapid development of membrane technology, membrane separation process has been gaining attention for antibiotic wastewater treatment. The reverse osmosis (RO) process, nanofiltration (NF) process and ultrafiltration (UF) process have been studied to remove tetracycline antibiotics from wastewater [8–10]. The rejection of examined antibiotics by some RO/NF membranes could achieve 98.5% [9]. More importantly, the tetracycline antibiotics in the RO or UF retentate can be recovered through conventional crystallization [8]. Nevertheless, RO, NF and UF are pressure-driven membrane processes, which are susceptible to membrane fouling [11]. Especially RO is still energy intensive process, in which 85% of energy consumption puts into the high pressure pumps [12]. Therefore, to explore other plausible membrane processes for tetracycline antibiotics separation with lower energy requirement and less membrane fouling is necessary.

Forward osmosis (FO), as a new membrane process, has been gaining popularity in the membrane separation area [11]. Unlike pressure-driven processes (RO and NF), FO is a natural process that utilized an osmotic pressure difference to drive water molecule across the membrane from a dilute feed solution into a concentrated draw solution [13]. Hence, FO possesses the advantages of low membrane fouling tendency due to the absence of hydraulic pressure [14,15]. Moreover, in FO system where recovery of draw solution is easy or unnecessary, FO will be energy-efficient [16–18]. Owing to these advantages, FO has been used for the treatment of municipal wastewater, oily wastewater and trace organic compounds (TOrCs) in water [19,20]. Furthermore, to produce fresh water and regenerate draw solution, FO could be combined with other membrane processes, such as RO and membrane distillation (MD) [21]. Especially, the FO can be utilized for the recovery of useful materials, such as nutrients and Na_2CO_3 [22,23]. The phosphorus in digested sludge centrate were extracted by FO process in the form of struvite ($MgNH_4PO_4{\cdot}6H_2O$) [22]. $Na_2CO_3{\cdot}10H_2O$ crystals were recovered from aqueous streams using FO process, and the purity of crystals was 99.98% [23]. Consequently, FO may be a promising technology for the recovery of tetracycline antibiotics from antibiotic wastewater.

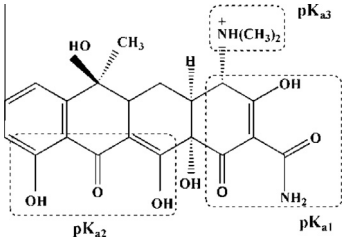
The application of FO process for the recoverable separation of TC from wastewater was proposed and studied in this study with commercial thin film composite (TFC) FO membrane. The effects of membrane orientation, feed velocity and the pH value of feed solution on TC separation were first investigated, followed by long-term studies of membrane cleaning and reuse for separation in FO process. To the best of our knowledge, this is the first time that FO process was studied to treat the TC wastewater, which may provide useful insights for the design of FO process for antibiotics separation from water during their production process.

2. Materials and methods

2.1. Solutions and FO membrane

Feed solution containing tetracycline for separation experiments was prepared from pure tetracycline hydrochloride powder (Beijing Solarbio Science & Technology, China). The main characteristics of TC hydrochloride are listed in Table 1. Solution of NaCl (Sinopharm Chemical Reagent, China) was used as draw solution. All the solutes were dissolved in deionized (DI) water, which has a conductivity of 5 μs/cm. The flat sheet TFC FO membrane obtained from Hydration Technology Hydration Innovations (HTI, USA) was recently commercialized. The main characteristics of this membrane are presented in Table 2. The parameters for *A* and *B* of TFC membrane are obtained from the Ref. [24].

Table 1
Characteristics of tetracycline molecule.

Structure	
Formula	$C_{22}H_{24}N_2O_8$
Molecular weight (g/mol)	444.44
pK_a[a]	3.3, 7.68, 9.3

[a] From Sassman and Lee [34].

2.2. Microscopic observation of TFC FO membrane

The micro-images of the membrane were obtained using a field emission scanning electron microscope (FESEM, S-4800, Hitachi, Japan) with at an accelerating voltage of 5 kV. Before imaging, samples were coated with a thin layer of gold by a sputter coater (EMS150T ES, EMS, USA). For the cross section observation, the FO membrane was freeze-fractured in liquid nitrogen to obtain a clean edge.

2.3. Forward osmosis system

A schematic diagram of the laboratory-scale FO system was illustrated in Fig. 1. A custom-made cross-flow membrane cell with two identical and symmetrical flow chambers was utilized. The flow chamber had a total effective membrane area of 40 cm^2 with length, width and height of 100, 40, and 2 mm, respectively. The feed and draw solutions were circulated with peristaltic pumps (Longer, China). The draw solution tank was positioned on a digital balance (SF6001F, Ohaus, USA) connected to a computer, and weight changes were recorded automatically every minute to determine the permeate water flux. In addition, the conductivity of feed solution was monitored by a conductivity meter (Eutech Instruments, Singapore) for the calculation of reverse salt flux. The test was conducted at room temperature (23 ± 1 °C).

2.4. Measurement of water flux and reverse salt flux of the FO membrane

In order to fully saturate the membrane porous support by water, the membrane was soaked in a 50% solution of ethanol for 5 min at the beginning of tests, then rinsed in deionized water [25]. All the tests were carried out in the membrane channel without any spacer under counter-current crossflow direction. TC solution with concentration of 1000 mg/L was used as feed solution for TC separation experiments, while DI water was used for baseline experiments. The pH of TC solution was kept at about 3.05. 2 M NaCl solution was employed as draw solution in all tests. The initial volumes of draw and feed solutions were fixed at 2 L and 1 L, respectively. Both the solutions were supplied at crossflow velocity of 12.5 cm/s (600 mL/min or Reynolds number (*Re*) of 531). The experiments were conducted for 9 h.

The water flux (J_W, $Lm^{-2}\,h^{-1}$, abbreviated as LMH) and reverse salt flux (J_S, $gm^{-2}\,h^{-1}$, abbreviated as gMH) were calculated as follows:

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