



Permeability recovery of fouled forward osmosis membranes by chemical cleaning during a long-term operation of anaerobic osmotic membrane bioreactors treating low-strength wastewater



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ABSTRACT

Anaerobic osmotic membrane bioreactor (AnOMBR) has gained increasing interests in wastewater treatment owing to its simultaneous recovery of biogas and water. However, the forward osmosis (FO) membrane fouling was severe during a long-term operation of AnOMBRs. Here, we aim to recover the permeability of fouled FO membranes by chemical cleaning. Specifically speaking, an optimal chemical cleaning procedure was searched for fouled thin film composite polyamide FO (TFC-FO) membranes in a novel microfiltration (MF) assisted AnOMBR (AnMF-OMBR). The results indicated that citric acid, disodium ethylenediaminetetraacetate (EDTA-2Na), hydrochloric acid (HCl), sodium dodecyl sulfate (SDS) and sodium hydroxide (NaOH) had a low cleaning efficiency of less than 15%, while hydrogen peroxide (H₂O₂) could effectively remove foulants from the TFC-FO membrane surface (almost 100%) through oxidizing the functional group of the organic foulants and disintegrating the colloids and microbe flocs into fine particles. Nevertheless, the damage of H₂O₂ to the TFC-FO membrane was observed when a high cleaning concentration and a long duration were applied. In this case, the optimal cleaning conditions including cleaning concentration and time for fouled TFC-FO membranes were selected through confocal laser scanning microscope (CLSM) and scanning electron microscopy (SEM) images and the flux recovery rate. The results suggested that the optimal cleaning procedure for fouled TFC-FO membranes was use of 0.5% H₂O₂ at 25 °C for 6 h, and after that, the cleaned TFC-FO membrane had the same performance as a virgin one including water flux and rejection for organic matters and phosphorus during the operation of AnMF-OMBR.

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1. Introduction

Osmotic membrane bioreactors (OMBRs) have been recognized as a promising technology for wastewater treatment and reuse owing to their better water quality over traditional membrane bioreactors (MBRs) (Cornelissen et al., 2008; Achilli et al., 2009; Lay et al., 2011; Wang et al., 2014a,b,c; Luo et al., 2015, 2016; Bell et al., 2016; Hou et al., 2016, 2017; Qiu et al., 2016; Wang et al., 2016a). In OMBRs, the tendency of FO membrane fouling is lower, and the foulants are easily removed through physical cleaning even though

a little irreversible fouling still exists on FO membrane surface (Achilli et al., 2009; Wang et al., 2014a, 2014b; Holloway et al., 2015; Chen et al., 2014; Gu et al., 2015; Sun et al., 2016). However, it has been reported that the fouling of FO membrane after a long-term operation of anaerobic osmotic membrane bioreactor combined with microfiltration (AnMF-OMBR) is severer, and flux permeability of FO membrane can not be recovered just by physical cleaning (Hu et al., 2017; Wang et al., 2017). In the AnMF-OMBR, the salinity is effectively controlled at a low level owing to the MF membrane for solute discharge, which enables a longer continuous operation and a higher methane production rate (Hu et al., 2017; Wang et al., 2017). In this case, membrane fouling becomes a key factor causing the flux drop of FO membrane in the AnMF-OMBR. Thus, it is necessary to seek alternative methods for recovering the permeability of FO membrane in the AnMF-OMBR based on the fact that physical cleaning is invalid.

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Chemical cleaning is considered as an effective method for removing membrane foulants (Ang et al., 2011; Wang et al., 2014c, 2015; Zhou et al., 2017). Typically chemical agents including alkaline solutions, acids, oxidants and metal chelating agents are usually applied for membrane cleaning (Mohammadi et al., 2002; Ang et al., 2011; Regula et al., 2014). Alkaline solutions are effective to eliminate organic fouling by hydrolysis and solubilization (Ang et al., 2011; Wang et al., 2014c), while acids are usually applied for removing inorganic foulants by neutralization and double decomposition reactions (Wang et al., 2014c; Regula et al., 2014). Additionally, oxidants aim to mitigate both organic fouling and biofouling through direct oxidations (Wang et al., 2014c). Moreover, metal chelating agents can effectively remove the combined divalent cations from the biofoulants, thus breaking down the fouling layer structure (Wang et al., 2014c). Therefore, it is important to select appropriate agents for cleaning fouled FO membranes according to the type of foulants.

Due to the low fouling tendency of FO membrane, studies on chemical cleaning of FO membrane are limited. With regard to the fouled cellulose triacetate (CTA) FO membrane, Linares et al. (2012, 2013) have used 1% Alconox and 0.8% sodium ethylenediaminetetraacetic acid (EDTA) to remove the organic foulants, while Yoon et al. (2013) applied 100 mg/L NaClO for eliminating biofouling. Until now, there is only one literature on the chemical cleaning of thin film composite polyamide (TFC) FO membrane fouled in an FO-Cell with a short-term operation (Wang et al., 2015). Based on the fact that NaClO can damage the TFC-FO membrane (Soice et al., 2003; Kwon and Leckie, 2006; Kang et al., 2007), Wang et al. (2015) applied 0.1% NaOH/0.1% SDS followed by 2% citric acid or 0.5% HCl as chemical agents for cleaning fouled TFC-FO membrane, and approximately 100% of flux recovery rate was obtained.

In current study, we intend to investigate the chemical cleaning of the fouled TFC-FO membranes in the AnMF-OMBR for low-strength wastewater treatment. Until now, there are no studies on seeking chemical cleaning protocols for the fouled TFC-FO membrane in the AnOMBR with a long-term operation. Thus, the objectives of this study are 1) to find the appropriate chemical agents and correspondingly cleaning conditions for the fouled TFC-FO membrane in the AnMF-OMBR, and 2) to evaluate the performance of the cleaned TFC-FO membrane by the selected chemical agents and cleaning conditions in the AnMF-OMBR.

2. Materials and methods

2.1. Set-up and operating conditions

The AnMF-OMBR with an effective volume of 4.98 L was operated at the temperature of 25 ± 0.5 °C, and the schematic diagram has been shown in our previous publications (Hu et al., 2017; Wang et al., 2017). Both an FO and an MF membrane modules (each with an effective area of 0.025 m²) were immersed in the bioreactor. The TFC-FO membrane provided by Hydration Technologies Inc. was applied in this study with an orientation of active layer facing the mixed liquors. The MF membrane (polyvinylidene fluoride (PVDF), Zizheng Environment Inc., China) with a mean pore size of 0.20 μm was operated under the mode of constant flux controlling by a peristaltic pump. Produced biogas was recycled with a recirculation rate of 2 L/min for alleviating the membrane fouling and mixing the anaerobic biomass.

Synthetic wastewater with the COD, NH₄⁺-N, TN and TP concentrations of 372.6 ± 7.19 , 28.96 ± 0.37 , 35.17 ± 1.07 and 3.35 ± 0.16 mg/L respectively was used as the influent. Its chemical compositions can be found in our previous publications (Chen et al., 2011; Wang et al., 2012). The seed sludge with mixed liquor

volatile suspended solids (MLVSS) and mixed liquor suspended solids (MLSS) concentrations of 2.8 and 3.8 g/L respectively was cultivated in a fermentation flask for about 60 days after collecting from a local WWTP. The specific information on the local WWTP and the cultivating procedure followed Wang et al. (2017). NaCl solution with a concentration of 0.5 M was used as the draw solution whose salinity was kept stable through a conductivity controller connected to a concentrated NaCl solution of 5 M. The sludge retention time (SRT) was controlled at 90 days, while the hydraulic retention time (HRT) varied in a range of 12.5–90 h owing to the flux drop of FO membrane.

2.2. Chemical cleaning procedure

The chemical cleaning procedure for the fouled TFC-FO membrane is listed in Fig. 1. A series of fouled TFC-FO membrane samples for fouling observation and chemical cleaning were collected from the AnMF-OMBRs when the flux of TFC-FO membrane decreased to approximately 2 LMH. As for the chemical cleaning, the cleaning efficiencies of different cleaning agents were determined and then the optimal one was selected. According to previous studies (Jung et al., 2006; Madaeni and Samieirad, 2010; Regula et al., 2014; Wang et al., 2015), chemical cleaning agents including 0.2% sodium hydroxide (NaOH) as an alkaline solution, 0.5% hydrochloric (HCl) and 2% citric acid as acids, 0.2% disodium ethylenediaminetetraacetate (EDTA-2Na) as a metal chelating agent, 0.2% sodium dodecyl sulfate (SDS) as an anionic surfactant and 0.5% hydrogen peroxide (H₂O₂) as an oxidant were applied in this study. All chemical agents were provided by Sinopharm Chemical Reagent Co., Ltd (Shanghai, China). The chemical cleaning of each agent lasted for 9 h at 25 °C, and then the flux of cleaned TFC-FO membrane was determined by an FO-Cell system to calculate the flux recovery rate for choosing an optimal cleaning agent. After selecting the optimal one, its cleaning concentration (0.1%, 0.3%, 0.5% and 1%) and cleaning time (1, 3, 6 and 9 h) were evaluated for choosing its optimal cleaning conditions by two steps. Firstly, the scanning electron microscopy (SEM) and confocal laser scanning microscope (CLSM) were applied for assessing the cleaning effects of different cleaning groups by comparing the surface morphology and distributions of biofoulants. And then, those selected cleaning groups with good cleaning effects were further evaluated for choosing an optimal cleaning condition by determining the flux recovery rate

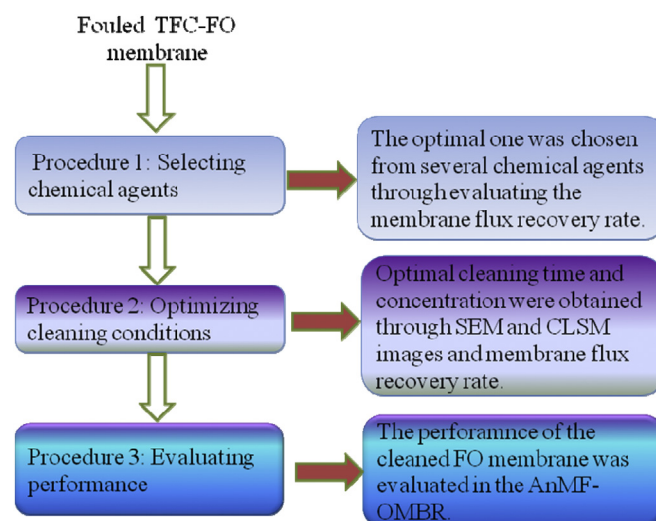


Fig. 1. Chemical cleaning procedure for the fouled TFC-FO membranes collected from the AnMF-OMBR.

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