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Q2 Performance and fouling mechanism of direct contact 2 membrane distillation (DCMD) treating fermentation 3 wastewater with high organic concentrations

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A B S T R A C T

In this study, direct contact membrane distillation (DCMD) was used for treating 16
 fermentation wastewater with high organic concentrations. DCMD performance character- 17
 istics including permeate flux, permeate water quality as well as membrane fouling were 18
 investigated systematically. Experimental results showed that, after 12 hr DCMD, the feed 19
 wastewater was concentrated by about a factor of 3.7 on a volumetric basis, with the 20
 permeate flux decreasing from the initial 8.7 L/m²/hr to the final 4.3 L/m²/hr due to 21
 membrane fouling; the protein concentration in the feed wastewater was increased by 22
 about 3.5 times and achieved a value of 6178 mg/L, which is suitable for reutilization. 23
 Although COD and TOC in permeate water increased continuously due to the transfer of 24
 volatile components from wastewater, organic rejection of over 95% was achieved in 25
 wastewater. GC–MS results suggested that the fermentation wastewater contained 128 26
 kinds of organics, in which 14 organics dominated. After 12 hr DCMD, not only volatile 27
 organics including trimethyl pyrazine, 2-acetyl pyrrole, phenethyl alcohol and phenylacetic 28
 acid, but also non-volatile dibutyl phthalate was detected in permeate water due to 29
 membrane wetting. FT-IR and SEM–EDS results indicated that the deposits formed on the 30
 membrane inner surface mainly consisted of Ca, Mg, and amine, carboxylic acid and 31
 aromatic groups. The fouled membrane could be recovered, as most of the deposits could be 32
 removed using a HCl/NaOH chemical cleaning method. 33

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46 Introduction

47 In recent years, an increasing amount of fermentation
 48 wastewater has been generated with the rapid industrial
 49 development in China. In general, fermentation wastewater is
 50 dark in color and has a high chemical oxygen demand (COD)
 51 value ranging from 1×10^5 to 6×10^5 mg/L (Zeng et al., 2009).

The highly-concentrated non-biodegradable organics, especially 52
 metabolites, make fermentation wastewater difficult to bio- 53
 degrade, so a treatment method is needed to prevent the 54
 environmental problems caused by its discharge. Multi-effect 55
 distillation (MED) and activated sludge treatment are the main 56
 methods developed to treat fermentation wastewater. However, 57
 MED requires too much energy, and activated sludge treatment 58

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wastes recyclable resource like protein in wastewater; thus, an appropriate treatment method for fermentation wastewater is urgently required to protect the environment and recover valuable resources in the meantime.

Membrane distillation (MD) is a membrane separation process driven thermally by the temperature difference between the feed side and the permeate side of the membrane. Theoretically, an MD system has the capability of producing pure water from natural water because only water vapor molecules can transfer through the porous hydrophobic membrane during the separation process (El-Bourawi et al., 2006). For MD systems there are four different configurations, namely: direct contact membrane distillation (DCMD), air gap membrane distillation (AGMD), sweeping gas membrane distillation (SGMD), and vacuum membrane distillation (VMD). Based on the characteristics of hydrophobic membranes, these MD configurations have been studied for seawater desalination, solute recovery and wastewater treatment (Hou et al., 2010; Khayet et al., 2004, 2005; Qu et al., 2009; Shirazi et al., 2014; Zarebska et al., 2014).

As the easiest and simplest configuration among them all, DCMD is the most widely studied (Drioli et al., 2015). To enhance water desalination, Cath et al. (2004) designed and investigated a new MD configuration and a new membrane module. Their research results showed that salt rejection in vacuum-enhanced DCMD could be greater than 99.9% in almost all cases. Al-Obaidani et al. (2008) developed an extensive analysis of DCMD performance and made an economic evaluation that the estimated water cost for DCMD with heat recovery was \$1.17/m³.

With the advantage of a lower fouling tendency, DCMD has been applied in treating various kinds of complex wastewater in recent research. El-Abbassi et al. (2009, 2013) applied DMCD in treating olive mill wastewater (OMW); their study results showed that the OMW concentration factor for the membrane TF200 was 1.72 after 9 hr DCMD operation, and an integrated microfiltration/DCMD system could be used to obtain clean water and a phenolic-rich concentrate from OMW. In the studies of Jacob et al. (2015), DCMD showed a reasonable flux of 2.09 L/m²/hr and high rejections of ammonia and COD of up to 89.6%–96.3% and 97.8%–99.9%, respectively, when treating anaerobic effluent. Wijekoon et al. (2014) investigated the feasibility of DCMD for removing trace organic compounds (TrOCs) during water and wastewater treatment, and the results of their experiments suggested that DCMD could be used as a promising post-treatment in conjunction with thermophilic membrane bioreactor for TrOC removal. Khayet (2013) used surface-modified membranes to process low and intermediate radioactive liquid wastes by DCMD, and their experimental results indicated that DCMD with surface-modified membranes has potential for application in nuclear technology.

Although DCMD has been proved applicable for wastewater treatment in many studies, its feasibility and performance in treating fermentation wastewater have rarely been studied. For a yeast factory, the highly-concentrated fermentation wastewater generated from the centrifugal filter unit generally has poor biodegradability, owing to the high concentration of organics (10,000–90,000 mg/L COD) it contains, therefore an efficient pretreatment is needed to remove a large part of the organics before further advanced treatment. In this study, a

well-designed DCMD configuration with a self-made membrane module was used as a pretreatment for treating fermentation wastewater with high organic concentrations from a yeast factory. DCMD performance characteristics such as permeate flux, permeate water quality, as well as membrane fouling mechanism and recovery methods were investigated systematically.

1. Materials and methods

1.1. Characteristics of fermentation wastewater

The fermentation wastewater samples were obtained from a yeast factory in Harbin, China. Before DCMD treatment, all the samples were preserved in a refrigerator. COD, TOC and protein concentrations of the fermentation wastewater were characterized as 54,900, 20,900 and 1765 mg/L, respectively. For solutions without inorganics, the value of COD (mg O₂/L)/TOC (mg carbon/L) is generally 2.66 and varies with water quality (e.g., between 2.0 to 5.0 for municipal wastewater). Therefore, although COD was much higher than TOC in the studied wastewater, the value of COD/TOC (calculated as 2.63) is in the normal range. The pH value of the fermentation wastewater was 6.0–7.0. Analyzed by gas chromatography–mass spectrometry (GC–MS), fourteen major organic compounds (area percentage > 1%) including isoamyl, 2-methyl butyric acid, 2,3,5-trimethyl pyrazine, 2-acetyl pyrrole, 2-pyrrolidinone, phenethyl alcohol, benzoic acid, phenylacetic acid, 4-ethenyl-2-methoxyphenol, o-hydroxybenzoic acid, p-hydroxyphenyl ethanol, p-hydroxyphenylcyanide, 4-hydroxy-3-methoxyphenethyl alcohol and butyl phthalate were found in the fermentation wastewater.

1.2. DCMD set-up and running conditions

The schematic diagram of the DCMD set-up used in this study for fermentation wastewater treatment is shown in Fig. 1.

The membrane module in the DCMD set-up was a self-made polyester tube combined with two unplasticized polyvinyl chloride T-tubes. The outside diameter, inside diameter and effective length of the module were 20, 15 and 225 mm, respectively. Sixteen pieces of commercial hollow fiber polypropylene (PP) membranes (ACCUREL PP Q3/2, Membrana, Germany) with a total effective membrane area of 0.023 m² were packed in the module. The basic membrane properties are as following: pore average diameter 0.46 μm, outer diameter/inner diameter is 2.5 mm/2.0 mm, thickness 0.25 mm, porosity 80%, liquid entrance pressure (LEP) 400 kPa.

DCMD was operated to run for 12 hr to treat fermentation wastewater. With an initial volume of 1 L, the feed fermentation wastewater was pumped continuously into the tube side after being heated by a heater (DK-98-IIA, Tianjing Taisite Technology, China), and the permeate water with an initial volume of 1 L was pumped into the shell side after cooling by a cooler (SDC-6, Nanjing Xinzhi Biotechnology, China). To prevent the feed wastewater being overly concentrated, wastewater of the same properties was supplemented continuously to the feed tank at a rate of 115 mL/hr. Limited by the low power of the cooler, the permeate temperature was

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