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Application of AC/TiO₂ Dynamic Membrane in Fouling Decline of Polymeric Membrane Support for Oily Wastewater Treatment

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Abstract

Oily wastewater is important environmental pollution which leads to irreparable damages to the environment. Ultrafiltration process is considered as a commonly used process is oily wastewater process. In the current study, two types of (AC/TiO₂) dynamic membranes (pre-coated and self-forming dynamic membrane) was used for oily wastewater treatment to decrease fouling of ultrafiltration membrane. The pre-coated dynamic membrane decreased fouling of the of ultrafiltration membrane while the self-forming membrane intensified the membrane fouling after three cycles of filtration. The COD rejection rate obtained for self-forming dynamic membrane was 45% that is more than the pre-coated dynamic membrane and polymeric membrane, this result shows high separation efficiency for this membrane. The obtained result showed that the pre-coated (AC/TiO₂) dynamic membrane can be considered as a good candidate to control the fouling effect of ultrafiltration membrane in oily wastewater treatment.

Keywords: Oily Wastewaters, Dynamic Membrane, Polymeric Support, Reducing Pre-coated Dynamic Membrane, Environmental Pollution.

Introduction

Oily wastewater produced in oil refining processes is the main environmental pollutant which affect the ecological balance [1]. The small size of oil droplets in the range of micron leads to difficult separation of oil from oily wastewater. Conventional methods to the treatment of oily wastewater due to use of chemical additives or high operation cost are restricted to meet the discharge standards. The technology of membrane separation has attracted a great attention to oily wastewater separation in recent year [2, 3]. Ultrafiltration and microfiltration are ranges of membrane process [4]. This technology has potential advantages of no chemical additives, high removing rate, no phase transition, which is more practical and economical. However, membrane separation technology application encounters several problems such as higher preparation cost and serious membrane fouling. It encourages us to focus on the practical and simple way of membrane modification in which the developed membrane is called dynamic membrane (DM). DM is formed in situ by deposition of a suspension containing fine particles either organic or inorganic materials on a porous support. The advantages of DM are simple preparation, ease of removal and regeneration. Pre-coated DM and self-forming DM are two basic types of dynamic membrane [5]. Pre-coated DM is formed before the filtration of feed solution on a porous support by deposition of dynamic membrane material, and self-forming DM is formed during the filtration of feed solution which contains dynamic membrane material. By use of the hydrophilic dynamic membrane materials can be reduced the fouling of support membrane through enhancing the

membrane surface hydrophilicity and decreasing exposures of support membrane to oil foulants [6]. In consequence by use of this technology can be acquired to efficient treatment of oily wastewater. In this study, Polyethersulfon (PES) and highly hydrophilic activated carbon (AC)/titanium dioxide (TiO_2) were applied as support polymeric membrane and material of dynamic membrane respectively. This DM was prepared by two methods to mitigate the fouling of support membrane. The antifouling performance of AC/ TiO_2 composite dynamic membranes (i.e., AC/ TiO_2 dynamic membrane and support polymeric membrane) and support polymeric membrane was compared together. In addition the treatment performance of these membranes through the COD¹ rejection rate was investigated.

Methodology

In this work, polymeric membrane used as support membrane was prepared by phase inversion method [7]. The casting solution was including, 16 Wt.% Polyethersulfon (PES), 2 Wt.% Poly (vinyl pyrrolidone) (PVP) and Dimethylformamide (DMF) as a solvent. To synthesis of AC/ TiO_2 as dynamic membrane material was used Sol-hydrothermal method [8]. The oily wastewater was taken from Sari Distribution Company of national oil products. The rate of COD for this wastewater was 7450 mg/l. For preparation of AC/ TiO_2 DM with an average particle size larger than the support polymeric membrane pore size, first this particles were passed from mesh of 120 to 325. To preparation of pre-coted dynamic membrane before the treatment of oily feed, AC/ TiO_2 particles were first added into deionized water

1. Chemical Oxygen Demand

at concentration of 1 g/L [6] to prepare AC/TiO₂ suspension, then the suspension by support polymeric membrane at 5 bar was filtered. Self-forming dynamic membrane was formed through directly added AC/TiO₂ particles to oily feed at AC/TiO₂ concentration of 1 g/L [6]. Then, the oily feed and AC/TiO₂ suspension were simultaneously filtered by support polymeric membrane at 5 bar. In both methods, the permeate flux was recorded and the filtration was stopped at the permeate volume of 0.05 L, which the deposition dose of AC/TiO₂ on membrane surface was theoretically approximately 0.05 g. In this study, the morphologies of the support polymeric membrane and AC/TiO₂ composite DM were studied by a scanning electron microscopy (LEO1450VP model). The average size of AC/TiO₂ particles was characterized by a particle size analyzer (SHIMADZU SALD-2101). The hydrophilicity of these membranes was characterized with contact angle measurement (CA 500M bara contact). To investigate the fouling of AC/TiO₂ composite dynamic membranes in the treatment of oily wastewater, a dead-end filtration setup was applied the filtration process which was conducted for 3 cycles to the treatment of oily feed, which each cycle including preparation of dynamic membrane, filtration of oily feed and washing membrane. Flux recovery rate as the antifouling factor was calculated with the following equation (1) for each filtration cycle:

$$FR_n = J_n / J_0 \times 100 \quad (1)$$

J_n defines the initial permeate flux after each washing membrane ($n = 1, \dots, 3$), and J_0 is pure water permeate flux of polymeric membrane.

In addition, COD rejection ($R_c\%$) of the membranes for oil rejection rate efficiency was

used according to the following expression:

$$R_c = (1 - C_p / C_f) \times 100\% \quad (2)$$

where R_c is the COD rejection efficiency, and C_p and C_f are COD concentrations (mg/L) of the permeate and feed, respectively.

For cleaning of the fouled membranes by the O/W emulsion, the deionized water was used as cleaning agents. The cleaning process by continuous filtration system was performed for 30 min. The flux recovery rate was evaluated for cleaning efficiency of these membranes.

Discussion and Results

The average particle size of AC/TiO₂ was 20.225 μm that was larger than the average pore size of support polymeric membrane, that led to deposition of AC/TiO₂ particles on the membrane surface as a cake layer. AC/TiO₂ dynamic membrane (contact angle=12°) was more hydrophilic than polymeric membrane (contact angle=57.55°). Since higher hydrophilicity shows lower fouling potential [9], the antifouling performance of polymeric membrane would considerably improve through covering of AC/TiO₂ composite particle on surface of support membrane. Flux recovery rate was examined to evaluate the antifouling performance of these membranes in each filtration cycle. Flux recovery rate of support membrane, self-forming and pre-coated composite dynamic membranes after third filtration cycle were 60.7%, 75% and 66.5% respectively in the filtration of oily wastewater. According to obtained results the pre-coated DM enhanced the flux recovery rate by 8.5% and significantly reduced the membrane fouling. In contrast, self-forming DM decreased the flux recovery rate by 5.8% and aggravated the membrane fouling. The higher flux recovery rate

of the AC/TiO₂ pre-coated composite DM can be ascribed to the high hydrophilicity of AC/TiO₂ DM that increases the removal of oil foulants from its surface by washing membrane. In this case DM acts as a protective layer preventing deposition and adsorption of oil molecules on support membrane surface. In contrast, the self-forming DM showed lower fouling control ability compared to the pre-coated DM due to simultaneous deposition of oil foulants and AC/TiO₂ DM materials on surface of support membrane that led to absorption of some oil foulants on AC/TiO₂ particles and surface of support membrane before deposition of DM material on this surface that causes these contaminants are not readily washed out from the surface of support membrane. According to the obtained results, the COD rejection rate of polymeric membrane, self-forming composite DM and pre-coated composite DM were 42%, 45% and 39.5% respectively. Higher COD rejection rate for self-forming composite DM like the removal efficiency of oily feed for this membrane was due to formation of compact layer of DM material on the surface of support polymeric membrane due to simultaneous deposition of oil foulants and AC/TiO₂ DM materials on surface of support membrane.

Conclusions

Pre-coated AC/TiO₂ dynamic membrane had better performance to oily wastewater treatment than self-forming dynamic membrane in terms of fouling. In contrast self-forming DM had high COD rejection rate than two other membranes. According to the obtained results, pre-coated DM is an efficient strategy to reduce the fouling of support membrane in the treatment of oily wastewater.

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