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Effect of cross-flow velocity, oil concentration and salinity on the critical flux of an oil-in-water emulsion in microfiltration

Henry J. Tanudjaja[a], Volodymyr V. Tarabara[b], Anthony G. Fane[c], Jia Wei Chew[a,c,*]

[a] School of Chemical and Biomedical Engineering, Nanyang Technological University, Singapore
[b] Department of Civil and Environmental Engineering, Michigan State University, East Lansing, MI 48824, USA
[c] Singapore Membrane Technology Centre, Nanyang Environment and Water Research Institute, Nanyang Technological University, Singapore

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ABSTRACT

Microfiltration is an attractive means for treating oily wastewater, especially when the size of the oil droplets are micrometer-sized since the conventional techniques become deficient. A systematic study on the critical flux of oil-in-water emulsion, which behaves differently from other colloidal foulants with regards to deformation, coalescence and splitting, has not been carried out to date. This was the goal of the current study, which employed the Direct Observation Through the Membrane (DOTM) technique to characterize the critical flux of oil-in-water emulsions of various concentrations, and at various cross-flow velocities (CFV) and salt concentrations. Five observations can be highlighted here. Firstly, the oil droplets with a mean droplet diameter of approximately 5 µm exhibited critical fluxes equal to or greater than latex particles of 10 µm. This is likely due to the twin effects of membrane oleophobicity promoting back-transport of the oil foulant from the membrane and the presence of a droplet size distribution with larger drops that can enhance the shear-induced diffusion of the average drops. Secondly, the critical flux values did not agree with the model that is valid for the size range the mean droplet diameter falls in, but instead agreed with the model adapted for smaller particulate foulants. Thirdly, the increase in the critical flux with CFV was more significant for the lower oil concentration. Fourthly, a striping phenomenon was observed at higher oil concentrations and lower CFV values. Striping was not observed for latex particles. Fifthly, the critical flux decreased with salt concentration. These findings highlight the unique fouling behavior of oil-in-water emulsions in microfiltration.

1. Introduction

Oil-water separations have widespread, economically significant applications, with industries spanning Wastewater, Oil and Gas, Food and Beverage (F&B), Shipping and Maritime, and Metal and Machining. Notably, produced water, which is the largest waste stream generated by oil and gas industries, represents the largest source of oily wastewater [1,2]. The oil-in-water emulsions treated have a wide range of concentrations that depend on the industry, such as low concentrations in the range of 50–1000 mg/l in produced water in the oil and gas industry, and high concentrations in the range of 3000–200,000 mg/l in the food and metal processing industries [3]. Conventional techniques employed, like gravity separation (which is the most common primary treatment of oily wastewater) and the hydrocyclone (which is a staple technology for oil companies to de-oil produced water), are unable to remove smaller micrometer-sized droplets [4,5]. However membrane-based oil-water separations are able to separate such oil droplets [6–9], and additionally confer many benefits relative to other separation methods, for example, small footprint, high oil removal efficiency, low investment and operation cost, operational simplicity, consistent quality of effluent, and amenability to be implemented in-process [4,5]. Unfortunately, the primary disadvantage of membranes is that fouling leads to ineffective separation and higher energy costs. Hence, a mechanistic understanding of the fouling caused by oil droplets is needed to ensure the technical and economic success of membrane applications in oil-water separations.

Several studies of membrane fouling by oil emulsions during ultrafiltration and microfiltration have been carried out. The permeate flux and rejection is well acknowledged to be correlated with operating parameters like trans-membrane pressure (TMP), feed concentration, temperature and cross-flow velocity (CFV) [10–15]. Arnot et al. [16] indicated that oil emulsion fouling was unique in terms of significantly lower initial fouling rate for higher cross-flow velocities (≥0.8 m/s), although this would be membrane-specific in terms of flux. Furthermore, fouling by oil emulsion was found to best correspond to the cake filtration model, rather than incomplete pore blocking or

* Corresponding author at: School of Chemical and Biomedical Engineering, Nanyang Technological University, Singapore.
E-mail address: JChew@ntu.edu.sg (J.W. Chew).

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complete pore blocking [15–20]. A model for calculating the critical pressure required to force the entry of an oil drop into a membrane pore was formulated based on the Young-Laplace equation and found to be a reasonable predictor of the operating pressure at which oil droplets deposit on the membrane [21,22]. Another study advanced the modeling for cross-flow applications for the behavior of an oil droplet pinned at the entrance of a micrometer-sized pore by solving the Navier-Stokes equation; the shear induced by cross-flow not only increases the critical pressure of permeation but may also cause the oil droplet to break into smaller droplets [23]. More recently, computational fluid dynamics (CFD) was employed to qualitatively study membrane pore blockage by oil emulsions, and found contact angle and interfacial tension to be dominant parameters [24]. Models, which are necessary for extracting the underlying physics, require complementary experimental data. Our recent fundamental study of microfiltration membrane fouling by emulsified oil was conducted using a combination of real-time visualization via the Direct Observation Through the Membrane (DOTM) technique, force balance on a droplet, and permeate flux analysis [25]. The results indicate that membrane fouling by an oil emulsion is mitigated by both cross-flow shear and droplet coalescence, the latter of which is more typical of oil emulsion foulants than solid particles. In the rich database of knowledge accumulated for membrane-based oil-water separations, a gap exists with regards to critical flux [26], which is a function of various parameters and provides important operational and design heuristics [27–29], of the oil emulsion. Correspondingly, the focus of this work was on a systematic analysis of the critical flux of oil emulsions and how it is impacted by practical operating parameters such as oil concentration, cross-flow velocity and salt concentration.

DOTM, which was developed to enable real-time visualization of particulate fouling phenomena at the feed-membrane interface [30], was used in this study to measure the critical flux of the oil-in-water emulsion. The specific objectives were to: (i) assess the effects of cross-flow velocity (CFV), oil concentration and salt concentration on critical flux; (ii) compare the critical flux of oil droplet foulants with latex particles of similar sizes; (iii) verify the agreement or lack thereof between the critical flux of oil droplets and shear-induced diffusion models developed for particulate foulants; and (iv) visualize the fouling phenomena at permeate fluxes just above the critical flux.

2. Experimental method

2.1. Oil-in-water emulsion

The dispersed phase was hexadecane (Sigma Aldrich), with a density of 773 mg/l (at 25 °C) and molecular weight of 226 g/mol, while the continuous phase was deionized water (MiliQpore) and Tween 20 (Sigma Aldrich). 800 ml of the oil emulsion stock solution was prepared, which consisted of 20 ml hexadecane, 4 ml Tween 20 and 776 ml deionized water, and was sufficient for all the experiments. The three components were mixed using a blender (Warring 8011 S) operated at 18,000 rpm for 10 s. Prior to every experiment, the feed solution was prepared by diluting the oil emulsion stock solution to the desired oil concentration, specifically, that typical in produced water of 50–500 ppm [31,32]. A new feed solution was prepared for each experiment, and the consistency of the feeds was verified by ensuring the oil emulsion droplet size distributions were similar.

The oil emulsion droplet size distribution was measured via the Focused Beam Reflectance Measurement system (FBRM; lasentec S400, PI-14/206). Fig. 1 displays the droplet size distribution of the stock solution with 25,000 ppm of oil in both the absence and presence of salt at various salt concentrations. The effect of increased salt concentration was such that the distributions became wider due to the greater percentage of oil droplets in the lower diameter range of smaller than 4 µm and in the upper range of larger than 7 µm. In both the absence and presence of salt, the number-based average oil droplet diameter was in the range of 4–6 µm, and 90% of the emulsion was in the diameter range of 1–20 µm throughout every experiment.

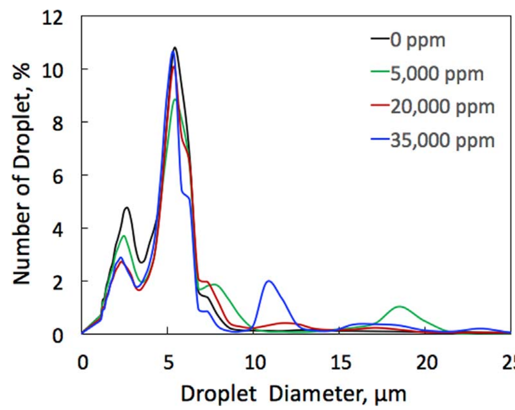

Fig. 1. Average oil droplet size distribution of 25,000 ppm of oil (stock solution) in the absence and presence of salt (NaCl concentrations between 5000–35,000 ppm).

2.2. Direct Observation Through Membrane (DOTM) setup

The Direct Observation Through Membrane (DOTM) setup employed was the same as that described in a previous study [33], which was based on the first report of this technique by Li et al. [30]. The schematic of the DOTM setup is depicted in Fig. 2, the key component of which is the light microscope (Zeiss Imager. A2 m) coupled with a camera (Axiocam 105 Color) to enable the observation of the feed-membrane interface during the cross-flow membrane filtration in real-time.

As the name DOTM implies, the membrane (Anopore, Whatman, Germany), which was a circular disk with a diameter of 47 mm and a nominal pore diameter of 200 nm, is transparent when wetted due to its relatively high porosity and straight through pores. Specifically, the membrane cell was such that the permeate channel was on top, and the focus was adjusted through the permeate channel and the membrane, such that the focal plane was the feed-membrane interface. The membrane was sandwiched using glue (Araldite) between two pieces of paper with dimensions of 55 mm by 135 mm and with a square cut-out in the center with dimensions of 27 mm by 27 mm. The cross-flow acrylic membrane cell had dimensions of 105 mm length by 35 mm width by 3 mm height, with 2 mm being the feed channel height and 1 mm being the permeate channel height.

The beaker containing the feed solution was placed on a magnetic stirrer plate (Heidolph, MR Hei-Mix S) to homogenize the oil emulsion suspension throughout the experiment, and a gear pump (Micropump Inc, GJ-N25. PF/S.A) enabled the circulation of the feed solution between the membrane cell and the feed solution beaker. For the permeate side of the membrane cell, a peristaltic pump (Cole Palmer; Masterflex L/S 7519-20/85) was employed to control the permeate flux

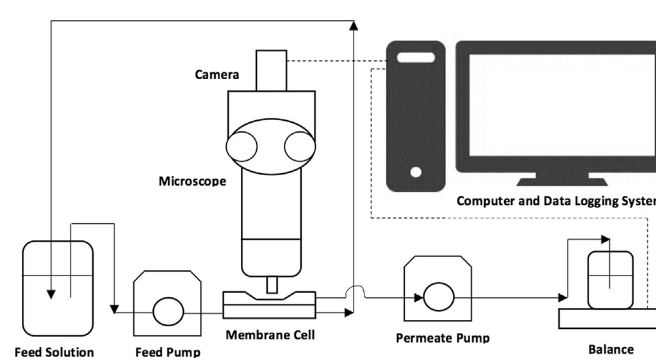

Fig. 2. Direct Observation Through the Membrane (DOTM) experimental setup.