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Experimental Study of the Separation of Oil in Water Emulsions by Tangential Flow Microfiltration Process. Part 2: The Use of Ultrasound for In-Situ Controlling of the Membrane Fouling

Wai Lam Loh[1,2], Thiam Teik Wan[1,2*], Vivek Kolladikkal Premanadhan[1], Ko Ko Naing[1,2], Nguyen Dinh Tam[1,2], Valente Hernandez Perez[1,2], Yu Qiao[1], Zhao[1] and Zheng Wang[1]

[1] *Department of Mechanical Engineering, Faculty of Engineering, National University of Singapore, Singapore*
[2] *Centre for Offshore Research and Engineering (CORE), National University of Singapore, Singapore*

Abstract

Membrane has a major drawback in the form of fouling. For controlling the membrane fouling, a novel attempt of having in-situ cleaning using ultrasound cavitations allowing remediation of a polluted surface during filtration was investigated. Results of the experiments indicate significant recovery of filter permeability by the assistance of ultrasound. At a feed of 500 ppm (0.05%) oil concentration, 15.07% recovery in permeability were recorded with the mean filtration capacity to improve from 2749.6 L m^{-2} h^{-1} to 2389.4 L m^{-2} h^{-1}. Significant decline in resistance of 18.93% indicates reduced fouling and the energy consumption required for maintaining the filtration flux, which may be used to supply the energy required for ultrasound cleaning. Encouraging results shows it is indeed possible to conduct in-situ cleaning while the filtration is still in operation and reduce cost for membrane replacement.

Keywords: Microfiltration; Ultrasound cavitations; Fouling control; Permeability; Ceramic membrane

Introduction

In previous paper, the application of tangential flow microfiltration for the separation of oil-in-water emulsions has been investigated. The laboratory experiments proven that the ceramic membrane of 0.5 μm pores size has the ability of producing filtrate with permeate oil concentration meeting the standard required for the offshore produced water effluent. However, examination of the membranes after experiments showed very thin oil layers adsorbed to the membrane surfaces. Oil is viscous and sticky, once it has contaminated the surface that can adversely affect the membrane performance which can be difficult to remove from the membrane surface. The ceramic microfiltration filters used in the experiments are the type of complex internal pore structures which were assumed to be responsible for the internal deposition of oil. Oil drops will become trapped in a small pore and therefore, decline in permeation flux should be expected over time of operation. This limitation has certainly impeded the wide spread use of this technology, it causes severe flux decline and affect the quality of the water produced.

Severe membrane fouling may require interruption of production time for intense cleaning or replacement. The usual methods of membrane cleaning include physical and chemical cleaning methods. Forward flushing or backwashing need to drive the water through the membrane pores forwards or backwards by pressure. Chemicals such as detergent, alkalis and acids are often used to clean fouled membranes by chemical reaction to weaken the cohesion forces between foulants and the adhesion force between foulants and the membrane surface. However, these techniques interrupt the continuous filtration process leading to a waste in processing. Alternatively, ultrasound may be used as a solution for in-situ cleaning of fouled membrane. Ideas to apply ultrasound for membrane cleaning are found in patents and very limited numbers of scientific papers [1-5].

Muthukumaran et al. has attempted to clean a fouled polymeric ultra-filtration membrane by applying 50 kHz of ultrasound, the result indicate that the ultrasound irradiation is effective in the removal of protein foulants and so increases the permeate flux after fouling [1]. Pirkonen et al. applied two minutes ultrasonic pulse in every hour interval on the ceramic capillary filter and the results indicate a higher level of filtration rate as compared with the case without ultra sonification [3]. Takaomi Kobayashi et al. also applied the ultrasound to remove fouling of ultra-filtration (UF) and microfiltration (MF) membranes which were used to treat peptone and milk aqueous solutions. It was found that 28 kHz frequency US could enhance formation of the fouled layer in both filtration systems of peptone and milk solution [4]. The aim of this paper is to present the results of an experimental study on the use of ultrasound for in-situ controlling of membrane fouling.

Fouling Mechanism

Membrane fouling is defined as the process in which solute or particles deposit onto the membrane surface or into membrane pores such that membrane performance is deteriorated [7]. The predominant fouling mechanism observed with tangential flow microfiltration membranes are classified into three stages of development: the build-up of a cake layer on the membrane surface; blocking of membrane pores, and; adsorption of fouling material on the membrane surface or in the pore wall. As flow tries to move across the membrane, micron size particles or suspended droplets tends to form bridge-like structure within the void spaces, as illustrated in Figure 1, and trapped within the porous matrix.

If the flock of particles is larger than pore diameter they would

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***Corresponding author:** Wan Thiam Teik, Research Associate, Faculty of Engineering, National University of Singapore, Singapore, Tel: 65-6516-8076; Fax: 65-6779-1936; E-mail: mpewtt@nus.edu.sg

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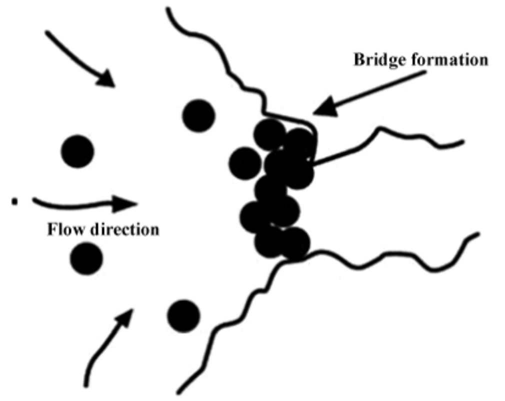

Figure 1: Formation of particle bridge at the neck of the membrane pores [6].

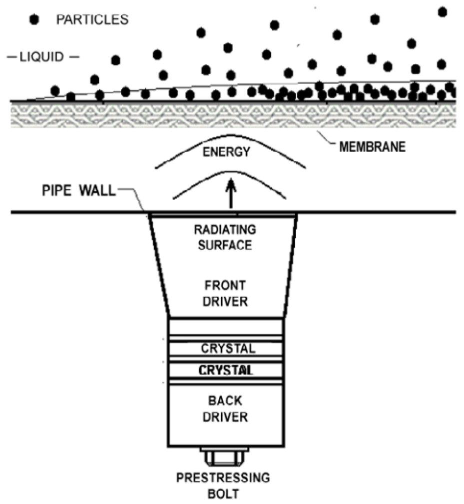

Figure 2: Ultrasonic cavitations within the liquid to prevent fouling materials from depositing at the membrane surface.

deposit on the wall or trapped within the neck of the pore, subsequently acts as collector of more droplets or fine particles released from porous medium [6]. The hydrodynamic drag due to ongoing flow of the host medium, may not be sufficient to entrain the foulants into the flow.

Fouling Control by Ultrasonic Cavitations

The main objective of this work is to prove the concept of in-situ cleaning of membrane filters using ultrasound cavitations. Ultrasonic cleaning offers several advantages over conventional methods. The ultrasonic waves generate an evenly distribute cavitations implosions in a liquid medium. The released energies reach and penetrate deep into pores of ceramic membranes which are normally inaccessible by other cleaning methods. The removal of contaminants is consistent and uniform regardless the complexity and geometry of the part being cleaned. Also, ultrasonic wave produces high frequency vibrations on the membrane filter. It creates an un-stationary flow field adjacent to the membrane surface to keep the particulate contaminants held in suspension, the resulting shear from ultrasonic vibration and cavitations will reduce the surface fouling, and the fouling materials dislodged from the membrane surface will be quickly swept away by the cross flow, as depicted in Figure 2.

Experimental Setup

A simple test rig is built separately from the test rig as shown in previous paper for the evaluation of the fouling control measures. The test rig consists of a flow loop equipped with a an emulsion feed tank, a clean water tank, a mixer, a centrifugal feed pump, a microfiltration filter module, flow meter, pressure gauges, valves, piping and an ultrasonic cleaning tank. The tangential flow microfiltration module is installed inside a stainless steel cartridge housing filled with clean water, and immersed totally beneath within an ultrasonic bath in an ultrasonic cleaning tank (4 x 50 w x 38 kHz ultrasonic transducers), as illustrated in the process and instrument diagram (P&ID) in Figure 3.

In this experiment, the effectiveness of fouling control measure using ultrasound for tangential flow microfiltration process was evaluated and compared with the case without the assistance of ultrasound. The experiments were conducted with an oily water feed of 500 ppm (0.05%) and 1000 ppm (0.1%) residual oil by volume respectively. Ultrasound of 38 kHz w was irradiated to the ceramic 0.5 μm microfiltration membrane as shown in Figure 2. The experimental objective is to investigate likely or not, the ultrasound cleaning would cure the membrane surface which has been fouled by such level of oil concentration in the feed stream. The oily water feed were prepared by emulsifying the desirable percentage of oil in clean water. The type of oil used in this work is Mobil Exxon DTE10 Excel 150 (Hydraulic Oil) with a heavy viscosity of 325.665 cP at 25˚C, 300 times much viscous than normal clean water. The oil-water mixture is agitated using a mixer for at least 5 minutes, further re- circulated to the emulsion feed tank by the centrifugal feed pump until a stable milky white mixture is formed.

Base Case Experiment

This first experiment is only meant to establish a base case (case 1) result for the comparison with the subsequent fouling control experiments (case 2-5). In a base case experiment, a new ceramic 0.5 μm membrane contaminated by allowing the clean water feed to flow along the tangential flow microfiltration module with a differential pressure of around 1.5 bar gauge and without switching 'ON' the ultrasound energy. The fouling process was continued for 10 runs of experiments, with 10 minutes process interval for each run of experiments, with a total a fouling time of 100 minutes. The experiment was then interrupted for a 10 minutes back-flush cleaning. The restored performance such as permeability and flux rate was recorded. The permeability and flow

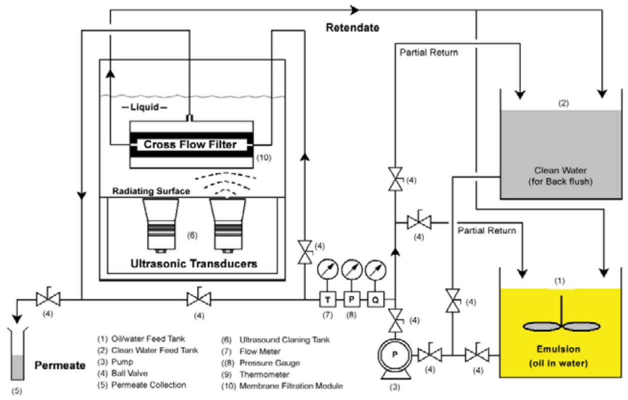

Figure 3: Schematic sketch of the experimental set-up.

resistance were calculated using equations available in permeability and flow resistance section.

Fouling Control Experiment

The main objective for the following experiments is to assess the performance of in-situ cleaning of membrane filters using ultrasound cleaning. It was found even with 10 minutes back-flush cleaning, only a 84.5% recovery from the initial permeability was recorded during the base case experiment after a 100 minutes filtration with clean water. It was impossible to restore back to the initial permeability once the filter was fouled. For practical reasons, a new ceramic filter was installed at each subsequent experiment.

In case 2 experiment, the experimental procedures are similar as in the base case, except it was replaced by a feed with lower residual content of 1000 ppm (0.1%) oil by volume. The ultrasound remains switched 'OFF' throughout the experiment.

In case 3 experiment, the experimental procedures are similar as in base case, except it was replaced by a feed with lower residual content of 500 ppm (0.05%) oil by volume. The ultrasound remains switched 'OFF' throughout the experiment.

In case 4 experiment, the experimental procedures are similar as in base case, except it was replaced by a feed with lower residual content of 1000 ppm (0.1%) oil by volume. However, this time the ultrasound energy switched "ON" throughout the course of experiment, even during the backwashing.

In case 5 experiment, the experimental procedures are similar as in base case, except it was replaced by a feed with lower residual content of 500 ppm (0.05%) oil by volume. However, this time the ultrasound energy switched "ON" throughout the course of experiment, even during the backwashing.

Permeability and Flow Resistance

The permeation flux across the membrane is calculated from J=V/(A*t) where J is the liquid flux across the membrane, A is the membrane surface area in contact with the liquid, t the run time of the experiment, and V is the volume of permeate collected during time t. The rate of permeate flux through membrane filter is dictated by the resistance of the filter to the flow of fluid and any resistance associated with the trapped particulate. The basic Darcy's Law is defined as:

$$\frac{dp}{dx} = \frac{\mu u}{K} \tag{3-1}$$

where dp=ΔP as the pressure drop across the filter, μ as the viscosity of the permeate fluid, dx is the thickness of the filter, K as the permeability, and u=J as permeation flux through the filter.

Equation (3-1) is rearranged to give a general equation as:

$$J = \frac{K\Delta P}{\mu \Delta x} \tag{3-2}$$

Permeability K refers to the ease with which water can flow through a filter. The resistance to permeation of a membrane is a function of the membrane pore size, feed stream components, and the degree to which gel layer formation and fouling layer formation occur. Increasing the feed stream circulation rate will, as a general rule, reduce gel layer thickness and increase flux.

Darcy 's Law states that the flux is directly proportional to the potential pressure drop and inversely proportional to the resistance, R:

$$R = \frac{\Delta P}{J} = \mu R_t \tag{3-3}$$

At a constant applied pressure for a given feed with constant composition and flow viscosity, the filtration flux is inversely proportional to the total resistance Rt. The total resistance can be expressed as follows [9]:

$$R_t = R_m + R_c + R_{cp} + R_g \tag{3-4}$$

The total resistance consists of resistance caused by the filter media (Rm) which can include the pore blocking resistance (R_p) and resistance by adsorption (Ra); resistance due to internal colloidal fouling (Rc); resistance due to formation of a highly concentrated layer adjacent to the membrane, concentration polarization (Rcp); and resistance caused by the formation of the gel layer (Rg), due to the increasing concentration of particles near the surface of the membrane.

Results and Discussions

The results of average permeability and flow resistance value of all experiments from case 1 (base case) to case 5 are tabulated in Table 1.

From the tabulated results, the fouling characteristics in terms of mean permeability from better to poor are sequenced in such an order : case 5 (9.967×10^{-11} cm^2), case 1 (9.290×10^{-11} cm^2), case 4 (8.802×10-^{11} cm^2), case 3 (8.661×10^{11} cm^2) and case 2 (7.92×10^{11} cm^2).

It seem that case 5 has the highest mean permeability (the least fouling) and case 2 has the lowest mean permeability, which indicates a worst fouling. The filter performance in term of mean flow resistance from better to poor has a similar sequence as above, where case 5 has the lowest mean flow resistance (least energy consumption) and case 2 with highest mean flow resistance, which indicates the highest energy consumption due to the worst fouling. The permeability history and flow resistance history of all experiments under comparable conditions from case 1 to case 5 are presented in the following Figures 4 and 5.

The effects of various factors toward the fouling (permeability decline) and energy consumption (flow resistance) would be discussed in the following sections.

Exp.	Oil Content of Feed (p)	Ultrasound Treatment (4x50Wx38 kHz) (on/off)	Mean Permeability K=Jμx/AP (cm²)	Mean Flow resistance R=ΔP/J (bars.s/m)	Mean Filtration Capacity (L, water/ m²hr)
1	<50	OFF	9.290E-11	7877.9	2563.0
2	1000	OFF	7.920E-11	9643.3	2185.1
3	500	OFF	8.661E-11	8764.4	2389.4
4	1000	ON	8.802E-11	8239.8	2428.3
5	500	ON	9.967E-11	7368.8	2749.6

Table 1: Mean permeability and flow resistance values (Case 1 to case 5: 0.5 μm 0.5 μm Doulton ceramic microfiltration membrane).

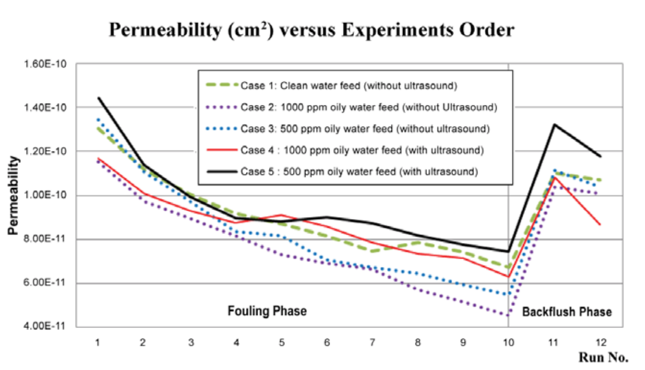

Figure 4: Fouling control experiment and flow permeability history (Case 1 to case 5: 0.5 μm Doulton ceramic microfiltration membrane).

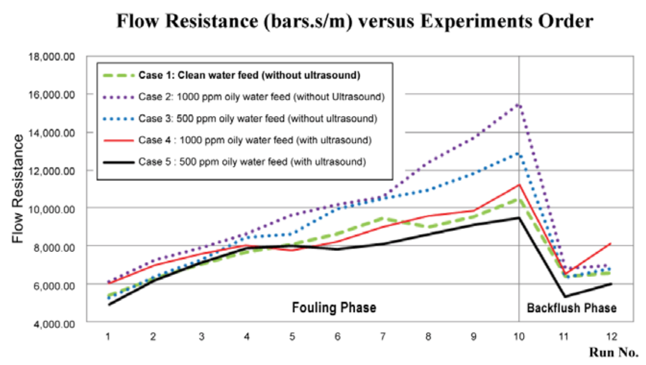

Figure 5: Fouling control experiment and flow resistance history (Case 1 to case 5 : 0.5 μm Doulton ceramic microfiltration membrane).

The effects of oil contents on permeability and flow resistance

In figure 4, the comparison of results indicates both the experiments with oil-water feed at 500 ppm and 1000 ppm residual oil and without the assistance of ultrasound, have the lowest permeability value (case 2 and 3). Both the mean permeability value in case 2 and 3 as revealed in Table 1, are found lower as compared with the base case experiment with clean water feed under comparable conditions. That means the presence of oil even in a small concentration, escalated the fouling, hence reduce the permeability and the filter processing capacity. Similar trends were observed in the flow resistance curve in Figure 5. The comparison of results indicate both the experiments with oily water feed (case 2 and 3) have higher energy consumption (higher resistance) as compared with the case with clean water feed (case 1). That means the presence of oil even in a small concentration, escalated the fouling and increase the energy required for maintaining the filtration flux.

The effects of ultrasound cleaning on permeability and flow resistance

In Figure 4, the comparison of results indicates both the experiments with ultrasound treatment (case 4 and 5), have higher permeability value as compared to the experiments without ultrasound treatment (case 2 and 3). In case 5 experiment with reduced feed concentration to 500 ppm of residual oil by volume, the mean permeability is 9.967×10^{-11} cm^2 is even better that case 1 with only clean water feed (without ultrasound). It shows a 15.1% improvement in permeability when compared the case without the assistance of ultrasound cleaning (case 3, mean permeability of 8.661×10^{-11} cm^2), and 7.30% improvement from the case with only clean water feed and without ultrasound cleaning (case 1, mean permeability of 9.290×10^{-11} cm^2). That means with the assistance of ultrasound cleaning, it reduces the degree of fouling, hence improve permeability and the filter performance.

Similar trends were observed in the flow resistance curve in Figure 5. The comparison of results indicates both the experiments with ultrasound treatment (Case 4 and 5) have lower energy consumption (lowest resistance) as compared with the case without ultrasound cleaning (Case 2 and 3). The fouling control experiments also indicate significant recovery of filter permeability by the assistance of ultrasound. At 500 ppm (0.05%) oil concentration, 15.07% recovery in permeability were recorded with mean filtration capacity to improve from 2749.6 L m^{-2} h^{-1} to 2389.4 L m^{-2} h^{-1}. Significant decline in resistance of 18.93% also indicate reduced fouling and the energy consumption required for maintaining the filtration flux, which may be used to supply the energy required for ultrasound cleaning. Encouraging results shows it is indeed possible to conduct in-situ cleaning while the filtration is still in operation.

Conclusion

The effects of ultrasonic cavitations on the permeability performance of tangential flow microfiltration using ceramic 0.5 μm membranes were investigated. The presence of heavy oil even in a small concentration has escalated the fouling, reduce permeability, increase resistance and the energy required for maintaining the filtration flux. However, the experimental results indicate significant recovery in flow permeability when the tangential flow microfiltration is assisted by the ultrasound cleaning. At 500 ppm (0.05%) feed concentration, 15.07% recovery in permeability was recorded with a mean filtration capacity to improve from 2749.6 L m^{-2} h^{-1} to 2389.4 L m^{-2} h^{-1}. This suggests that the ultrasound assisted tangential flow microfiltration process is a promising technique for preventing membrane fouling during oily water treatment at feed concentration below 1000 ppm residual oil. For ultrasonic cleaning of microfiltration membranes to be a viable option, it must be energy-effective [8]. The results of experiment at 500 ppm (0.05%) feed concentration indicates a significant decline in resistance of 18.93% indicates reduction in the energy consumption required for maintaining the filtration flux, suggests the possibility of re-utilizing the energy saved for the supply to the ultrasonic application. However, it is unclear at this stage, whether by using ultrasound is sufficient to substitute the back-flush cleaning for continuous operation without frequent filter replacement. The results indicate necessity for optimization of the cleaning procedures to enable the best cleaning for possible substitution to the conventional back-flush operation. More works is required to decide the relevant criteria (such as the power, frequency, suitable membrane materials, etc) for better assessment of this application.

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