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High frequency back-pulsing for fouling development control in ceramic microfiltration for treatment of produced water

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ABSTRACT

Produced water is a mixture of dispersed oil in water, dissolved organic, and inorganic particles. Membrane technology is a potential process to meet future discharge standards and regulations on the Norwegian Continental Shelf (NCS). Fouling minimization with back-pulsing technique was assessed in this study. The influence of produced water properties i.e., oil concentration, oil types, particles and membrane flux during the back-pulsing was investigated. α-Al_2O_3 MF membranes (nominal pore sizes; 0.1, 0.2 and 0.5 µm) were tested. Results show that control of fouling rates is better at low oil concentrations, low flux, larger oil droplet size and no fine oil/particles. Selection of membrane pore sizes depends on the feed properties. At low oil concentration (i.e., 50 ppm) and low flux (i.e., 250 $l.m^{-2}.hr^{-1}$ (LMH), a pore size of 0.5 µm resulted in the lowest fouling rates, whereas for higher oil concentrations and low flux and presences of fine particles the 0.2 µm pore size performed better. At high flux (i.e., 500 LMH), high oil concentrations and presences of particles the 0.1 µm membrane had the lowest fouling rates. A permeate quality of <5 ppm could be achieved even though the oil in the feed was increased from 50 to 350 ppm of oil in water.

Keywords: Produced water; Oil emulsion; Ceramic microfiltration; Fouling rate; Back-pulsing

1. Introduction

Produced water is a by-product from oil production, and consists of dispersed oil in water emulsions, dissolved organic compounds, and inorganic particles. The composition typically includes residual concentrations of chemical additives from the production line along with traces of heavy metals and inorganic compounds. The total amount of produced water discharged on the Norwegian Continental Shelf (NCS) in 2008 was about 149 million m^3, amounting to 1400 tonnes of oil for all discharge points on the NCS [1]. The discharges will increase in the years ahead, primarily resulting from increased water production from the major fields on the NCS, and increased use of chemicals for enhanced oil recovery (EOR) from mature reservoirs. Forecasts show that until 2012–2014 the discharge of produced water into the sea per year will increase to around 200 million m^3 [2]. In order to reduce the environmental impact of contaminants in produced water, current regulations for installations on the NCS have set a limit to 30 mg/l of oil in water [2].

The main strategy for reducing the pollution from produced water has traditionally been to reduce the content of dispersed oil. One of the reasons behind this strategy is

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that good removal of dispersed oil has also resulted in substantial removal of soluble hydrocarbons [3]. Therefore, technology improvement is needed to remove the oil in water to be as low as possible. Membrane filtration has the potential for a very effective separation of oil from water and thus is a potential alternative to meet future demands. Investigations on using micro and ultra-filtration to remove the dispersed oil from oily water streams have been reported [4–7]. Generally, for feed waters with 0.1–10% of oil content, either microfiltration (MF) or ultrafiltration (UF) has the capability of producing water with less than 5–100 ppm of oil. Both polymeric and inorganic membranes have been studied, however, in recent years there has been a growing interest in using ceramic membranes for this application. The use of ceramic membranes in oily wastewater effluent treatment has advantages over organic membrane because organic membranes are sensitive to both polar and chlorinated solvents, as well as to high oil fractions.

Membrane fouling is the major drawback in applying membrane technology, which in long-term operation will result in a progressive decrease of flux and induce loss of separation efficiency. Problems encountered during membrane filtration include; (1) accumulation of oil droplets on the membrane which may cause concentration polarization or cake layer depositions; (2) fouling from oil drops into some pore causing pore plugging or adsorption of either oil, surfactant, co-surfactant or organic material on the membrane surface. These are considered some of the dominant fouling phenomena.

Various strategies for fouling management may be applied in membrane processes, which either include physical or chemical methods. In crossflow mode of operation, membrane fouling may be reduced as a function of operating conditions, in particular the hydrodynamic conditions within the membrane module. The use of higher shear stress, turbulence promoters, and vortex promotion are some of the methods used to reduce fouling on the membrane surfaces. Back transport and removal of foulants are created by the instabilities generated by the fluid hydrodynamics which promotes good mixing and minimizes polarization effects [8].

The physical removal of a fouling layer utilizes mechanical forces and hydraulic cleaning to dislodge and remove foulants from the membrane surface. Hydraulic cleaning methods include forward flushing, vibrations, air sparging, CO_2 back permeation, back-flushing, back-shock/back-pulsing treatment, alternate pressurising and depressurising, and by reversing the flow direction at a given frequency [9].

High frequency back-pulsing is a promising technique for fouling control. The fundamental difference between back-pulse and backflushing/backwashing is the speed and force utilized to dislodge foulants from the membrane surface. In backflush, flow reversal occurs for 5–30 seconds once every 30 min to several hours, while flow reversal in back-pulsing occurs every few seconds at high pressure for a very short time [10]. The effectiveness of high frequency back-pulsing to minimize membrane fouling has been reported by several researchers [10–14]. All these studies demonstrated the effectiveness of high frequency back-pulsing in controlling membrane fouling and enhancing permeate flux using both polymeric and ceramic membranes.

Back-pulsing is an in situ membrane cleaning method to remove foulants from the membrane surface or pores. Back-pulsing is a cyclic process of forward filtration (J_f) followed by reverse filtration (J_b) at high frequencies, which can remove particle deposition that takes place within the first milliseconds. Back-pulsing can be conducted using permeate or compressed air. Several parameters are associated with back-pulsing, i.e., back-pulse duration (t_b) which is the amount of time operating under negative transmembrane pressure (ΔP_b) and backpulse interval defined as the duration of time in between two consecutive pulses. The mechanism of flux enhancement can be explained by diffusion of particles from the cake surface, erosion of the cake, decompression and subsequent washing away of the cake, and shock waves (due to the rapid valve closing) generated, knocking off part of the cake layer formed [14].

In this study tests were conducted to increase the understanding and to provide more information on the influence of back-pulsing techniques to improve the membrane performance, particularly in the treatment of produced water. Evaluation on the effects of different feed characteristics (i.e., crude oil types, oil concentration and solid particles) on the ceramic membrane performance using back-pulsing was also included.

The effectiveness of back-pulsing to reduce membrane fouling was done by comparing fouling rates between operation with and without back-pulsing. Membrane fouling rates were calculated using the expression;

$$dP / dt = \frac{TMP_f - TMP_i}{\Delta t} \tag{1}$$

where TMP_f is the final transmembrane pressure (TMP) at the end of filtration, TMP_i is the initial TMP at the beginning of filtration and Δt is the duration of filtration.

The effectiveness of the back-pulsing was measured as the degree of reduction of fouling rate observed with and without back-pulsing, expressed as;

$$dR(\%) = 100\,\frac{\left(dP / dt\right)_{NoBP} - \left(dP / dt\right)_{BP}}{\left(dP / dt\right)_{NoBP}} \tag{2}$$

where dR is the degree of fouling rate reduction by back-pulsing (BP), Subscripts BP, $NoBP$ denote value with and without back-pulsing.

The highest back-pulsing effectiveness will be 100%, meaning that back-pulsing could promote no fouling or give a fouling rate of zero. The higher the effectiveness the better the process performance is, with respect to eliminating fouling.

The oil removal efficiency (*R*) was calculated by the expression;

$$R(\%) = \frac{C_f - C_p}{C_f} \times 100 \qquad (3)$$

where C_f is the concentration of oil in the feed and C_p is the concentration of oil in the permeate

2. Materials and methods

2.1. Apparatus

α-Al_2O_3 ceramic membranes provided by ECO-CERAMICS, The Netherlands, with a nominal pore sizes of 0.1, 0.2 and 0.5 µm were mounted together in a tubular crossflow module. Each membrane has dimensions of 0.8 cm inner diameter, 1.1 cm outer diameter and 34 cm length. The effective membrane area was 85 cm^2. This experimental design enables direct comparison of the performance of different membrane pore sizes. A peristaltic pump was connected to the permeate stream to give a constant permeate flow. The TMP was monitored continuously using a pressure transducer, the temperature using a temperature transducer and the pH using a pH meter. A data acquisition unit was connected to all transducers to record the data. Permeate back-pulses were applied by connecting the outlet of membrane permeate side with a pressurised tank through a three way valve that is opened and closed in a minimum of 0.05 seconds. A 20 l pressurised tank was filled with deionized water for back-pulsing. An illustration of the membrane filtration setup is shown in Fig. 1.

2.2. Feed preparation for experimental trials

The oil emulsion feed was made of different crude oils with characteristics shown in Table 1, and diluted with salt (NaCl) water. The dispersed oil was prepared by mixing oil and surfactant in a Ultraturrax homogenizer (Janke & Kunkel TP 18/10, Germany) at a mixing rate of 20000 rpm for 10 minutes. The surfactant used was a non-ionic surfactant, SERDOX (Champion Technology, The Netherland), at concentrations of 65 and

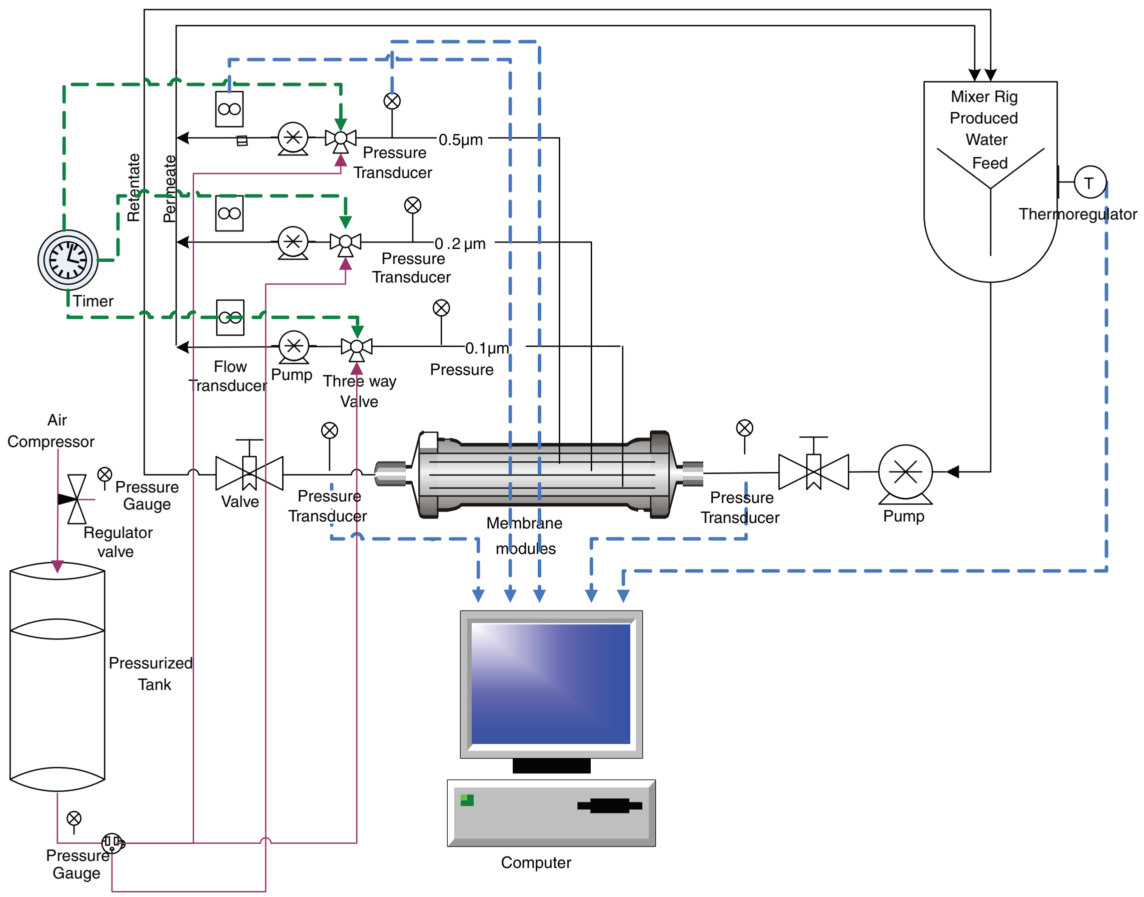

Fig. 1. Schematic diagram of experimental setup for back-pulsing-crossflow microfiltration.

Table 1
Oil characteristics for crude oils used at 60°C [15]

Crude oil type	Density (gr/cm^3)	Viscosity (cP)	Saturates (wt %)	Aromatic (wt %)	Resins (wt %)	Asph. (wt %)	IFT[a] (mN/m)	TAN[b] (mg KOH/g)
TB	0.84	4.50	63.69	28.63	6.29	0.35	12.92	0.49
Gli	0.86	12.70	51.29	37.46	9.83	1.22	17.89	0.00
Sei	0.94	505.00	35.76	41.35	18.18	4.58	17.20	3.36

[a]IFT: Interfacial Tension;
[b]TAN; Total Acid Number.

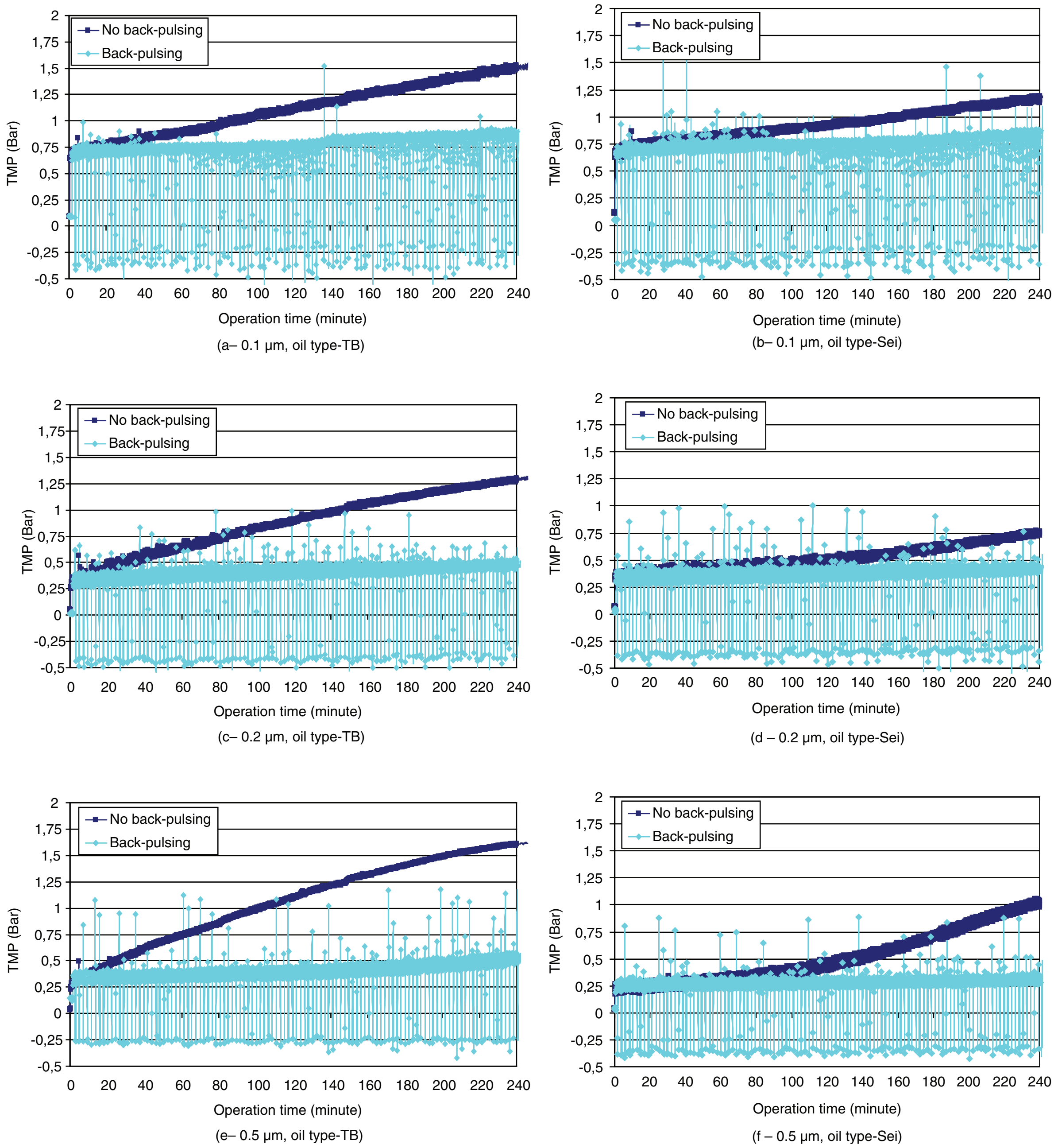

Fig. 2. *a,b,c,d,e,f* : Effect with and without back-pulsing on oil type-TB and oil type-Sei with concentration 50 ppm on different membrane pore sizes, with surfactant (450 ppm), without particles at flux 500 l.m^{-2}hr^{-1}.

450 ppm. Higher surfactant concentration was required to emulsify the high viscosity oil (Oil type-Sei) at high concentration (i.e., >50 ppm of oil in water) to maintain emulsion stability during the filtration tests. The stable emulsion was continuously stirred and thermoregulated in the storage tank (Fig. 1). The pH was selected at 4, 6.2 and 9, and adjusted using NaOH and HCl 1N at temperature of 27 ± 0.5°C. The oil concentrations tested were 50, 150, 250 and 350 ppm with salinity of 1-w/v%. Solid particles of kaolin (fine powder with particle size distribution (psd) of 0.1–4 µm, Aldrich, USA/Germany) were added at a concentration of 50 ppm. A scaling and corrosion inhibitor with concentration of 10 ppm (Champion Technology, The Netherlands), was added into the solution to make the final composition of the analogue produced water. Droplet size distribution of the resulting oil emulsion was measured by optical sensing and visual analyzes using a Jorin ViPA. The oil droplet characteristic was modelled to mimic typical produced water effluent from a hydrocyclone (<15 µm).

2.3. Experimental procedure

The membrane filtration was carried out on the analogue produced water for 3 or 4 h. A crossflow velocity of 4.5 m/s with constant flux mode operation was applied. The selected fluxes were 250 and 500 $l.m^{-2}.hr^{-1}$. The retentate and a part of the permeate were circulated back to the feed tank (volume 20 L) to maintain a constant oil concentration in the feedwater. The permeate TMP for each membrane pore size during the filtration was monitored to observe the progression of the fouling rate, whereas the total hydrocarbon in permeate was measured after 3 h operation.

The effectiveness of back-pulsing to reduce the fouling was assessed using equation (1) and the degree of reduction using this technique by equation (2).

Feed and permeate concentrations of treated analogue produced water were measured using Gas Chromatography, model Agilent 6890 following the standard modification of ISO 9377-2 [16]. The total hydrocarbon (THC) of the feed and permeate for each experimental trial were measured to evaluate the oil removal efficiency. The oil retention by the membrane (or removal efficiency) was measured and reported as the concentration of oil in the permeate.

The cleaning procedure after each experimental trial consisted of flushing the membrane module with distilled water followed by a chemical cleaning sequence. The fouled membranes were cleaned between successive experiments using 1%-v of Ultrasil 115 (Henkel-Ecolab Ltd.) and 1%-v of SurfactronCD50 (Champion Tech) for 1 hour at 80°C and TMP of 0.25 bar [17].

2.4. Modes of operation investigated

The structure of the results and discussion is based on membrane performance when back-pulsing was introduced. Furthermore, due to different feed characteristics usually present in the produced water, the influence of surfactant concentration, oil concentration, crude oil types, and solid particles were studied. Moreover, the effect of membrane operation, i.e., membrane flux, and the membrane properties, i.e., pore sizes, were investigated. At the end, the quality ofthe permeate with and without back-pulsing is presented.

3. Results and discussion

3.1. Influence of back-pulsing to control fouling development

The influence of high frequency back-pulsing was tested for different feed characteristics of analogue produced water. The first experiments were conducted to determine the possibility of high frequency back-pulsing as a means to reduce fouling of the membrane. The back-pulsing parameter used was a frequency of 0.2 Hz, back-pulse duration of 0.2 sec and reverse pressure -0.4 bar [18].

Results from testing two different types of oil and the three membrane pore sizes are presented in Fig. 2. All clearly show that high frequency back-pulsing could avoid and reduce further deposition of compounds

Table 2
Influence of back-pulsing to reduce the fouling rate for two oil types and three membrane pore sizes

NoBP/BP	Oil type	Surfactant concentration (mg/l)	Particles concentration (mg/l)	Flux (LMH)	Oil concentration (mg/l)	Fouling rate (mBar/min)			Backpulsing effectiveness (%)		
						0.1 µm	0.2 µm	0.5 µm	0.1 µm	0.2 µm	0.5 µm
NoBP	Sei	450	0	500	50	2.00	1.67	3.13	58.33	75.00	81.33
BP	Sei	450	0	500	50	0.83	0.42	0.58			
NoBP	TB	450	0	500	50	3.83	4.13	5.38	70.63	82.83	82.19
BP	TB	450	0	500	50	1.13	0.71	0.96			

on the membrane. The graphs show the time dependence of the TMP development when a 50 ppm concentration of oil type-TB and oil type-Sei based feedwater was first filtered with and without back-pulsing for 180 min for membrane pore sizes of 0.1, 0.2, and 0.5 µm respectively. Without back-pulsing, a marked increase

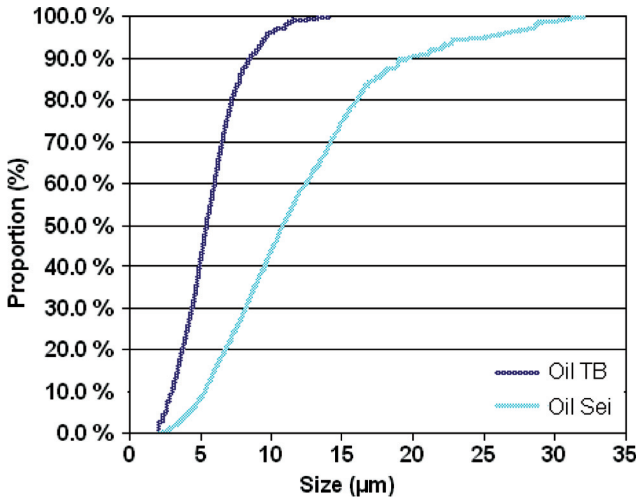

Fig. 3. Droplet size distribution (volume) of oil-TB and oil-Sei at 50 ppm oil in water with 450 ppm of surfactant concentration.

of TMP with time is seen, indicating that fouling develops due to the deposition of foulants on the membrane surface. Oil type-TB has a higher TMP development for all membrane pore sizes tested compared to the oil type-Sei [19]. This is due to the difference in droplet size distribution between oil type-TB and oil type-Sei, where oil type-TB has smaller droplet sizes. This promotes higher resistance (or higher TMP), which could be due to a more compact foulant deposition and also the fine droplet size distribution for oil type-TB possibly causing a pore restriction in addition to the cake layer deposition.

When back-pulsing was employed, a significant TMP reduction was observed for all oil types and all membrane pore sizes, indicating that back-pulsing has a positive effect on the microfiltration performance (i.e., less fouling). Table 2 shows the reduction of fouling rate measured for oil type-TB and type-Sei, and for all membrane pore sizes tested. Using back-pulsing, the fouling rate for each membrane pore size (0.1, 0.2, 0.5 µm) could be reduced by 58%, 75% and 81% for oil type-Sei and 71%, 78% and 82% for oil type-TB respectively. The largest membrane pore size (0.5 µm) obtained the highest improvement between with and without back-pulsing, however, the 0.2 µm membrane had the lowest fouling rate when operated with back-pulsing.

(*a* – 0.1 µm, oil type-TB) (*b* – 0.1 µm, oil type-Sei)

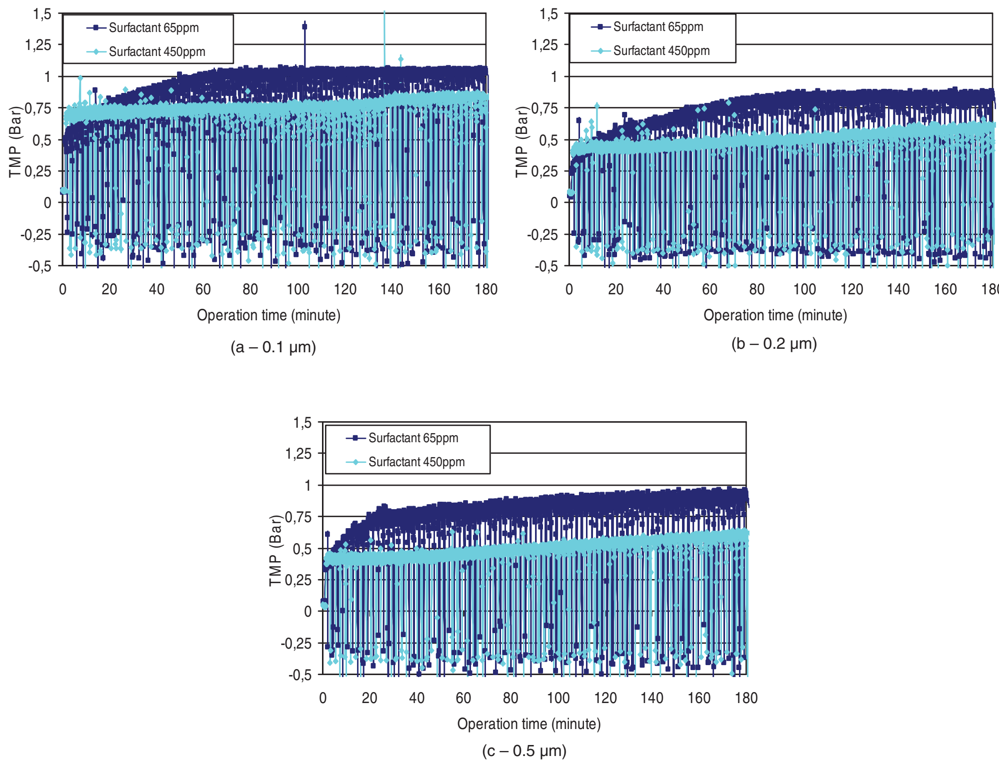

Fig. 4. *a,b,c*: Effect of surfactant concentration (65, 450 ppm) on different membrane pore sizes for oil type-TB (50 ppm), without particles, with back-pulsing at flux 500 l.m^{-2}.hr^{-1}.

(*c* – 0.2 µm, oil type-TB) (*d* – 0.2 µm, oil type-Sei)
(*e* – 0.5 µm, oil type-TB) (*f* – 0.5 µm, oil type-Sei)

3.2. Influence of surfactant concentration

The influence of surfactant concentration in the analogue produced water was studied for two different oil types when back-pulsing was employed. The concentration of surfactant will affect the oil size distribution. The surfactant could also increase the hydrophilization of the membrane, which thus reduces the total resistance during the membrane filtration. The surfactant concentration was tested at 65 ppm and 450 ppm. The higher surfactant concentration (450 ppm) was selected because it was also required further to generate and maintain stable emulsions for the high oil viscosity at high oil concentration (350 ppm). The droplet size distributions of oil type-Sei and TB with 450 ppm surfactant concentration were significantly different, as illustrated in Fig. 3.

The influence of varying surfactant concentration with oil type-TB at flux 500 l.m^{-2}.hr^{-1} and 50 ppm oil concentration, is shown in Fig. 4. Increasing surfactant

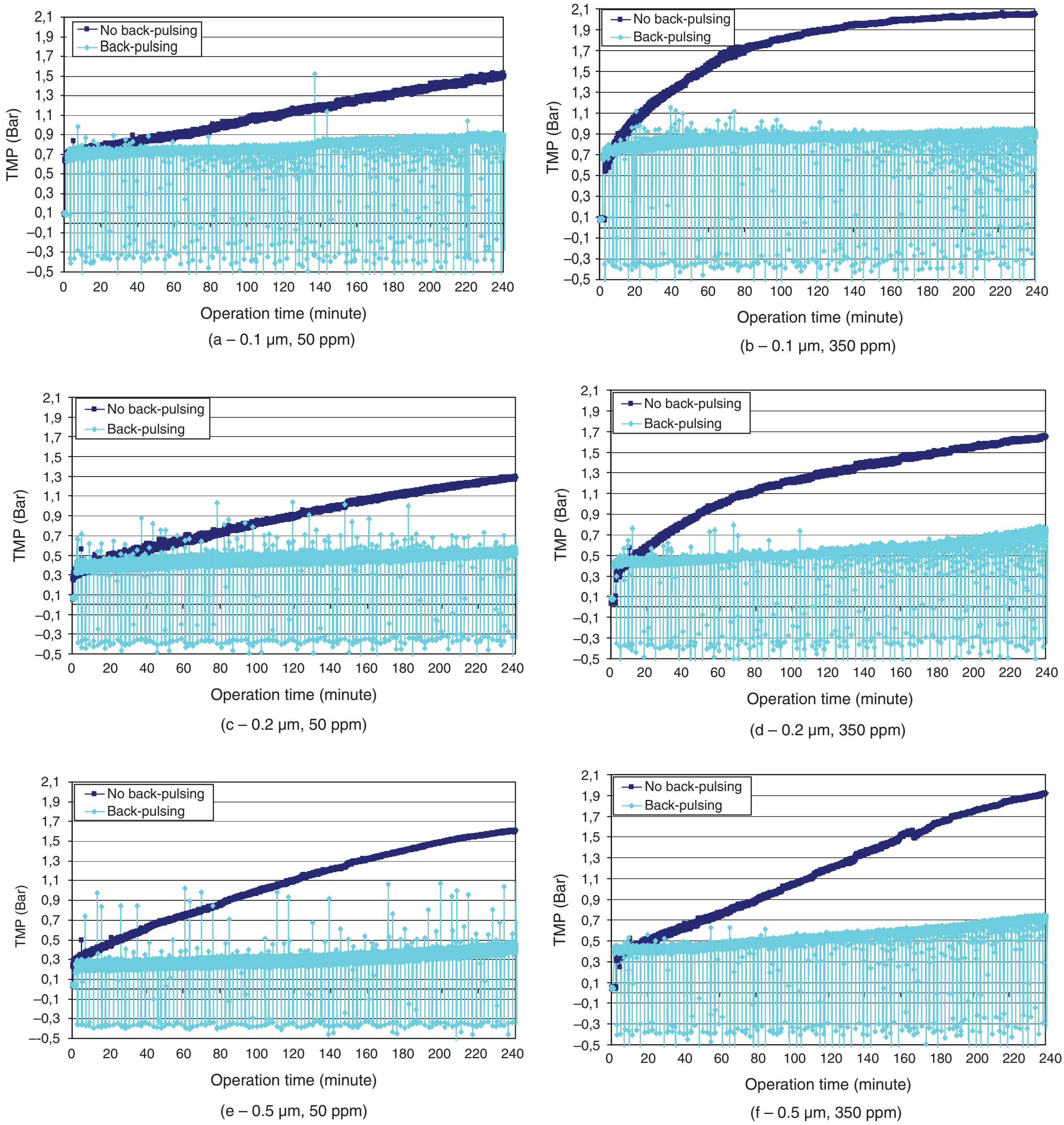

Fig. 5. *a,b,c,d,e,f*: Effect of oil concentration (50, 350 ppm) of oil type-TB on different membrane pore sizes with surfactant (450 ppm), without particles, with and without back-pulsing at flux 500 l.m^{-2}.hr^{-1}.

concentration reduces the TMP development over time for all membrane pore sizes. The reason for this behaviour is most likely due to the interaction between the hydrophobic chain of the non-ionic surfactant and the non-polar sites of the membrane, leading to a more hydrophilization of the membrane surface and pores. This interaction is previously reported in literature for non-ionic surfactants [20]. The interaction between the surfactant and membrane surface, which provides a hydrophilic barrier to the oil adsorption, is thus expected to reduce the compactness of the foulant cake layer on the membrane surface. The resulting fouling layer is possibly more permeable due to the higher surfactant concentration. Further, the attachment of the surfactant on the membrane surface could increase the hydrophilicity of the membrane surface that also reduces the resistance and subsequently the TMP.

(*a* – 0.1 μm) (*b* – 0.2 μm)
(*c* – 0.5 μm)

3.3. Influence of oil concentration

The effect of oil concentration on membrane filtration was studied for oil type-TB and oil type-Sei. The result of oil type-TB for operation with and without back-pulsing is presented in Fig. 5. Increasing concentrations from 50 to 350 ppm increases the TMP value and rate of change (or filtration resistance) for all membrane pore sizes without applying back-pulsing. This is due to the increase of oil droplet concentration on the membrane surface increasing cake-layer deposition, and thus the overall filtration resistance. An increase of oil concentration on the membrane surface also promotes pore narrowing or blockage due to the adsorption of oil droplets on the membrane surface or in the pore walls of the membrane, which further increases the overall resistance.

Applying back-pulsing the TMP value and rate of change was reduced significantly. The difference of membrane fouling rate as a consequence of higher oil concentration is greater without back-pulsing compared to when back-pulsing is used. However, the trend showing an increase in fouling rate with increasing concentration for all membrane pore sizes tested is similar both with and without back-pulsing. With increasing concentrations there is a potential for greater fouling. Back-pulsing is efficient in removing deposits from the membrane surface but not adsorbed material within the membrane pores. Table 3 summarizes and illustrates the effectiveness of the back-pulsing technique when the oil concentration was increased from 50 to 350 ppm for different membrane pore sizes. Increasing oil concentrations will increase membrane fouling, where the effectiveness of back-pulsing tends to be reduced as the suspension concentration increases. The largest membrane pore sizes gain the least benefit from back-pulsing when oil concentration increases. This is most likely due to the fouling mechanisms by fine oil droplets causing pore plugging or internal fouling being significant. When internal fouling in the larger pores takes place back-pulsing is probably not effective to removing the foulants from the pores.

(*a* – 0.1 μm, 50 ppm) (*b* – 0.1 μm, 350 ppm)
(*c* – 0.2 μm, 50 ppm) (*d* – 0.2 μm, 350 ppm)
(*e* – 0.5 μm, 50 ppm) (*f* – 0.5 μm, 350 ppm)

Table 3
Influence of back-pulsing to reduce the fouling rate of non-back-pulsing operation with increasing oil concentration (50, 350 ppm)

NoBP/ BP	Oil type	Surfactant concentration (mg/l)	Particles concentration (mg/l)	Flux (LMH)	Oil concentration (mg/l)	Fouling rate (mBar/min)			Backpulsing effectiveness (%)		
						0.1 μm	0.2 μm	0.5 μm	0.1 μm	0.2 μm	0.5 μm
NoBP	Sei	450	0	500	50	2.00	1.67	3.13	58.53	75.00	81.33
BP	Sei	450	0	500	50	0.83	0.42	0.58			
NoBP	Sei	450	0	500	350	2.54	2.46	4.75	54.07	66.12	69.30
BP	Sei	450	0	500	350	1.17	0.83	1.46			
NoBP	TB	450	0	500	50	3.83	4.13	5.38	70.63	82.83	82.19
BP	TB	450	0	500	50	1.13	0.71	0.96			
NoBP	TB	450	0	500	350	6.17	5.33	6.92	78.39	74.20	74.71
BP	TB	450	0	500	350	1.33	1.38	1.75			

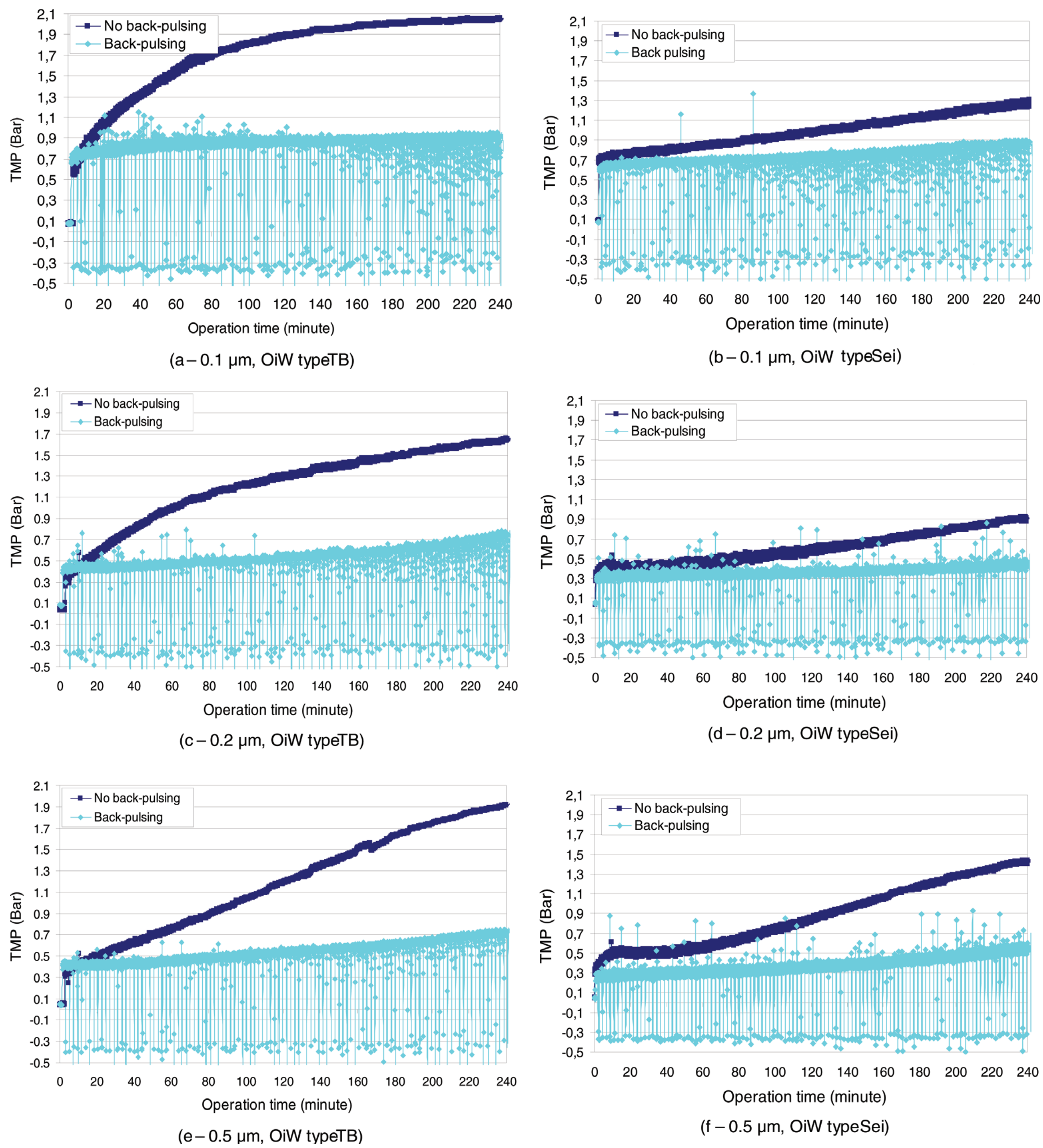

Fig. 6. *a,b,c,d,e,f*: Effect of different oil types (TB, Sei) with concentration 350 ppm on different membrane pore sizes with surfactant (450 ppm), without particles, with and without back-pulsing at flux 500 $l.m^{-2}.hr^{-1}$.

3.4. Influence of oil types / droplet size distribution

The membrane filtration performance is dependent on the oil types due to the different inherent properties i.e., viscosity, density. In preparing the analogue produced water used in this study, different oil types at similar mixing rates (i.e., mixing intensities to generate emulsions) and surfactant concentrations gave different oil droplet size distributions. Subsequently different membrane filtration performances were observed. Silalahi et al. [19] reported the influence of oil types and characteristics of the oil droplet size distribution which ultimately impacted membrane fouling. Analysis of oil droplet size distribution found for both oil types investigated in this study is presented in Fig. 3. Oil type-TB has the smaller oil droplet sizes and distribution of the two.

The effect of introducing back-pulsing during the filtration is shown in Fig. 6 *a,b,c,d,e,f* for oil concentration

Table 4
Influence of oil types (Gli, Sei, TB) to the fouling rate with back-pulsing

Oil type	Surfactant concentration (mg/l)	Particles concentration (mg/l)	Flux (LMH)	Oil concentration (mg/l)	Fouling rate (mBar/min)		
					0.1 µm	0.2 µm	0.5 µm
Gli	65	0	250	50	1.92	1.56	1.04
Sei	65	0	250	50	0.79	0.41	0.38
TB	65	0	250	50	1.67	1.29	0.96
Sei	450	0	250	50	0.54	0.25	0.17
TB	450	0	250	50	0.75	0.46	0.42
Sei	450	0	500	50	0.83	0.42	0.58
Gli	450	0	500	50	1.17	0.79	1.25
TB	450	0	500	50	1.13	0.71	0.96
Sei	450	0	250	350	0.92	0.63	0.69
TB	450	0	250	350	1.28	0.58	0.63
Sei	450	0	500	350	1.17	0.83	1.46
TB	450	0	500	350	1.33	1.38	1.75
Gli	450	0	500	350	1.67	1.96	2.92

of 350 ppm and membrane flux 500 $l.m^{-2}.hr^{-1}$. It is apparent that when back-pulsing was employed the fouling rates (i.e., TMP) were reduced significantly for both oil types (oil concentration 350 ppm). The fouling rate for oil type-Sei is less than oil type-TB when no back-pulsing used. This is attributable to the smaller droplet size distribution in oil-TB, which results in higher resistance or TMP. Table 4 shows that when back-pulsing is used the fouling rate was reduced to 54%, 68%, and 69% for the 0.1, 0.2, and 0.5 µm pore sizes respectively for oil type-Sei compared to without back-pulsing. For the oil type-TB the equivalent reduction was 73%, 71% and 74%.

When back-pulsing was not employed, the difference in fouling rate between the two oils is quite large, however, after back-pulsing is applied the difference between the two oil types is less. At the end of the filtration period tested, the TMP for both oil types with back-pulsing is close. Analysis and summary of fouling rates is presented in Table 4. Even though the difference in droplet sizes and distribution between the two oils is large, when back-pulsing is used the difference in fouling rate is not that different. Therefore, these results supports the findings that back-pulsing is sufficient to lift up the foulants which block or cover the membrane surface and pores but not efficient for extraction of foulants which have penetrated the pores and cause internal fouling.

(*a* – 0.1 µm, OiW type-TB) (*b* – 0.1 µm, OiW type-Sei)
(*c* – 0.2 µm, OiW type-TB) (*d* – 0.2 µm, OiW type-Sei)
(*e* – 0.5 µm, OiW type-TB) (*f* – 0.5 µm, OiW type-Sei)

3.5. Influence of particles

The presence of particles in the analogue produced water were also investigated (Fig. 7). Silalahi et al. [19] reported that the addition of particles tends to increase the membrane resistance (or TMP) compared to without particles. This result is contrary to the observations of others who have shown that particles may adsorb to the oil emulsion and in effect increase the average oil/particle size distribution and hence reduce the TMP. For example, Mueller et al. [6] reported that addition of 250 ppm diatomaceous earth to a 250 ppm crude oil emulsion solution reduced the resistance of 0.2 µm ceramic membranes. This was accounted to the adsorption of oil by the particles. Panpanit et al. [21] reported that the addition of bentonite in ultrafiltration enhanced the flux of an oil emulsion solution due to an increase of the average particle size. Turcaud et al. [22] reported that the addition of particles with sizes larger than 3 µm have little fouling effect on membrane filtration while particles near 0.2 µm produced rapid fouling. Kaolin with a particle size average of 3 µm and concentration of 30 ppm gave a permeate flux decrease of only 14% during 300 min of filtration time. In this study, it was not observed that addition of particles tend to increase the size distribution. Therefore, the addition of the particles could be expected mainly to increase the concentration of the suspension in the feed and therefore increase the TMP. Further, the fine particle size distribution of the kaolin particles promote faster fouling and results in higher resistance due to the possibility of plugging the membrane pores.

Without back-pulsing, the presence of colloidal particles increased membrane fouling for all membrane pore

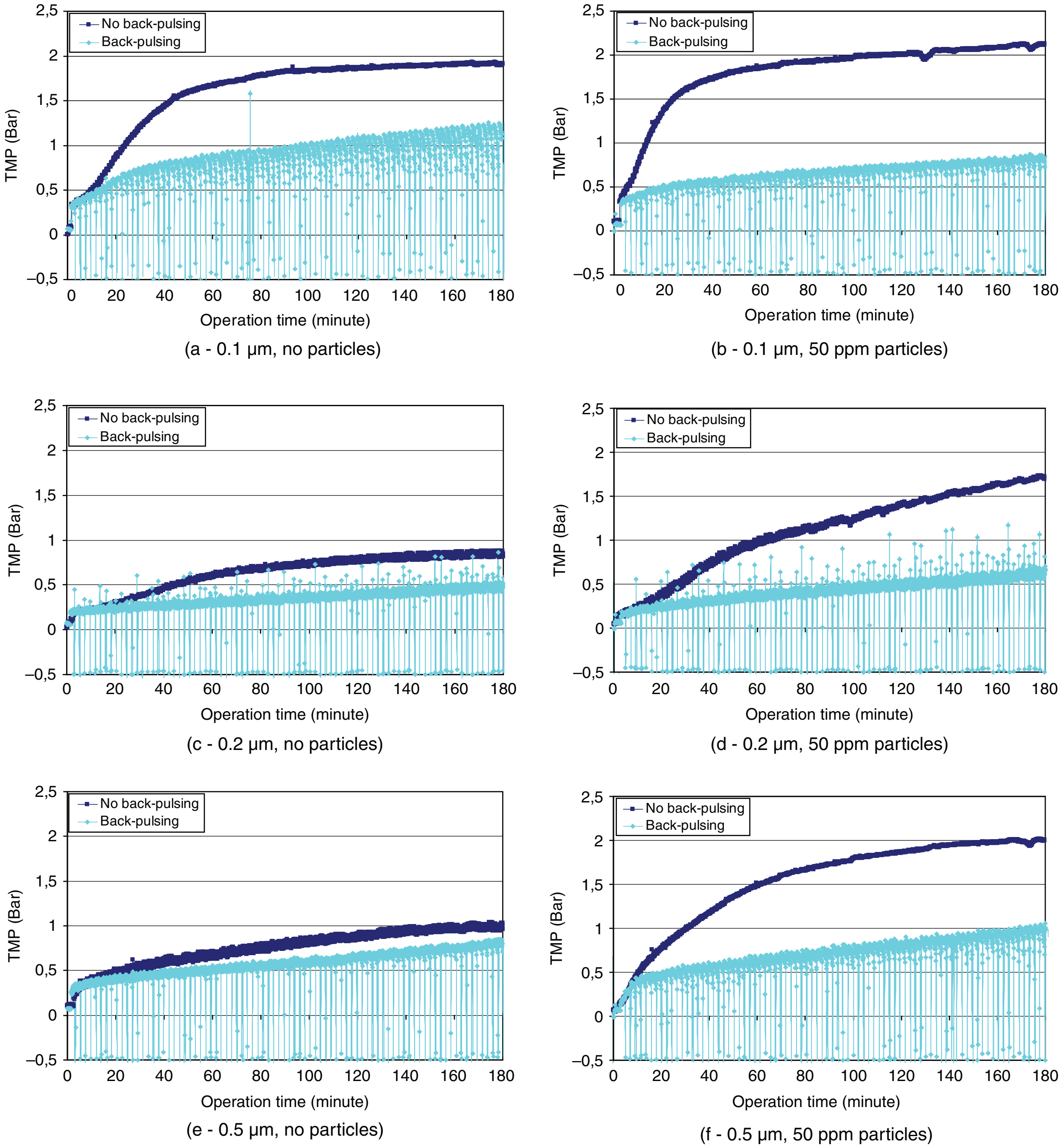

Fig. 7. *a,b,c,d,e,f*: Effect of particles (0, 50 ppm) of oil type-TB (150 ppm) on different membrane pore sizes with surfactant (65 ppm), with and without back-pulsing at flux 250 $l.m^{-2}.hr^{-1}$.

sizes tested. The impact of the fine particles on membrane filtration performance was most clearly seen on the membranes with the larger pore sizes. The 0.5 µm membrane pore size showed the highest TMP value and rate of change when the colloidal particles were added to the analogue produced water. This was to be expected since the kaolin particles had a size ranging from 0.1 to 4 µm. The main fouling mechanism caused by this fraction of particles is pore plugging which creates a higher resistance as the porosity of the membrane is reduced compared to membranes with smaller pore sizes. The smaller membrane pore sizes are less prone to pore plugging and fouling by cake-layer formation dominates as the main fouling mechanism.

When the back-pulsing technique was used during the membrane filtration the fouling rate was reduced

Table 5
Influence of particles in feed to the fouling rate with back-pulsing

Oil Type	Surfactant concentration (mg/l)	Particles concentration (mg/l)	Flux (LMH)	Oil concentration (mg/l)	Fouling rate (mBar/min)		
					0.1 µm	0.2 µm	0.5 µm
TB	65	0	250	50	1.67	1.29	0.96
TB	65	50	250	50	1.33	2.67	3.09
TB	65	0	250	50	2.83	3.46	3.46
TB	65	50	250	50	1.79	3.33	5.21
TB	65	0	250	150	4.94	2.11	3.44
TB	65	50	250	150	2.83	2.56	4.72
TB	65	0	250	250	5.56	3.89	4.89
TB	65	50	250	250	3.83	3.67	6.22
TB	65	0	250	350	5.83	4.78	5.17
TB	65	50	250	350	4.33	3.56	6.50
TB	450	0	500	50	1.13	0.71	0.96
TB	450	50	500	50	1.04	1.71	2.17
TB	450	0	500	350	1.33	1.38	1.75
TB	450	50	500	350	1.29	1.96	2.38
Sei	450	0	500	350	1.17	0.83	1.46
Sei	450	50	500	350	1.08	1.88	2.21
Sei	450	0	500	50	0.83	0.42	0.58
Sei	450	50	500	50	0.75	1.46	1.58
Gli	450	0	500	50	1.17	0.79	1.25
Gi	450	50	500	50	1.08	1.75	2.50

significantly. This is shown with the TMP measurements being much lower compared to results without back-pulsing for all membrane pore sizes tested, Fig. 7.

Table 5 shows the comparison of fouling rates observed for the feed water without and with kaolin particles for different oil concentrations, oil types, and surfactant concentration. When the oil concentration is varied from 50, 150, 250 to 350 ppm, the fouling rate increases for feed water without particles for all membrane pore sizes. A similar trend is observed for the analogue produced water with the addition of particles. When particles are present in the feed water during operation with back-pulsing, the smallest impact of back-pulsing is seen on the membranes with the larger pore sizes. The back-pulsing effectiveness was reduced with increasing membrane pore size. For the tightest membrane pore size of 0.1 µm the most positive effect was observed where the addition of particles resulted in lower or similar fouling rates as without particles. On the other hand, applying back-pulsing for membrane pore sizes 0.2 and 0.5 µm the fouling rate (or TMP measured) increased with addition of particles. This suggests that fouling is dominated by pore plugging and internal fouling by pore restriction in the presences of colloidal particles with a size less than the nominal membrane pore size. This type of fouling is difficult to reduce by high frequency back-pulsing. This observation also shows that back-pulsing is mainly very effective for removing fouling caused by external fouling on the membrane surface, particularly cake-layer formation. Therefore, understanding of feed oil droplet / particle sizes and distribution is necessary to enable a proper selection the most appropriate membrane pore size for a given application.

(*a* – 0.1 µm, no particles) (*b* – 0.1 µm, 50 ppm particles)
(*c* – 0.2 µm, no particles) (*d* – 0.2 µm, 50 ppm particles)
(*e* – 0.5 µm, no particles) (*f* – 0.5 µm, 50 ppm particles)

3.6. Influence of flux

The selected membrane flux influences the drag forces generated at the membrane surface by permeate going through the membrane. The higher the operating flux the higher the amount of suspension that is potentially brought to the membrane surface will be and thus increasing the overall membrane resistance. The influence of flux on the fouling rate was therefore studied in order to obtain information regarding the flux ranges, which would give a low fouling rate and thus give the basis for long-term sustainable membrane filtration operation.

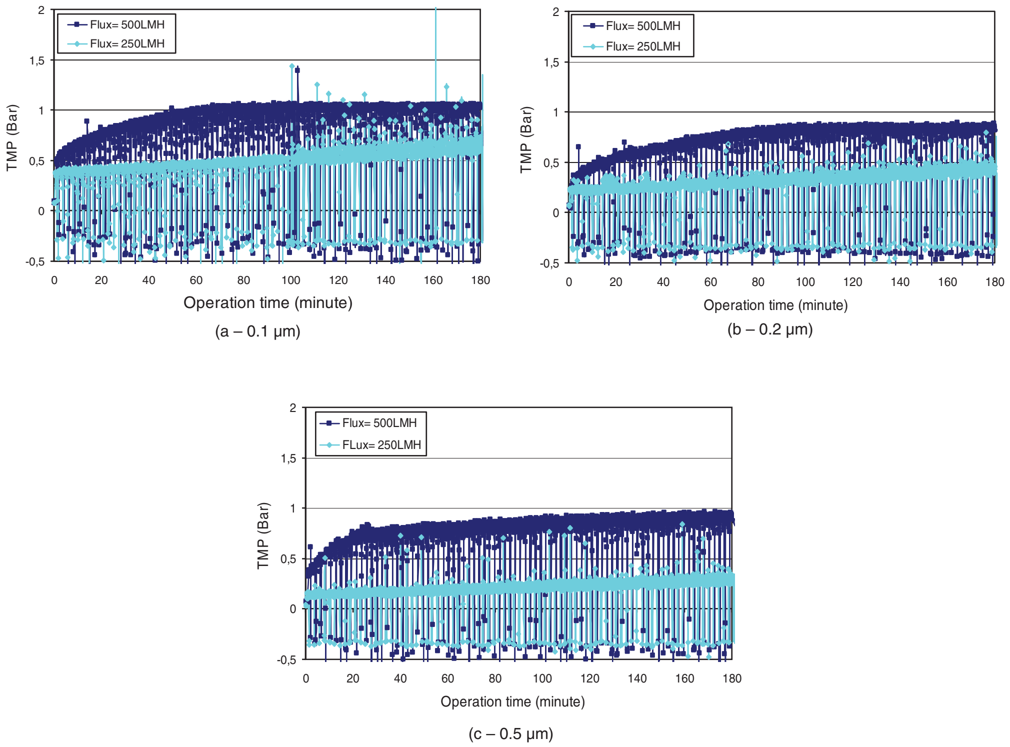

Fig. 8. *a,b,c*: Effect of different fluxes (250, 500 $l.m^{-2}.hr^{-1}$) on different membrane pore sizes for oil type-TB (50 ppm), with surfactant (65 ppm), without particles, with back-pulsing.

Table 6
Influence of increasing of flux to the fouling rate with back-pulsing

Oil type	Surfactant concentration (mg/l)	Particles concentration (mg/l)	Flux (LMH)	Oil concentration (mg/l)	Fouling rate (mBar/min)			Ratio fouling rate when flux is doubled		
					0.1 µm	0.2 µm	0.5 µm	0.1 µm	0.2 µm	0.5 µm
TB	65	0	250	50	1.67	1.29	0.96	1.70	2.68	3.61
TB	65	0	500	50	2.83	3.46	3.46			
Sei	450	0	250	50	0.54	0.25	0.17	1.54	1.67	3.50
Sei	450	0	500	50	0.83	0.42	0.58			
TB	450	0	250	50	0.75	0.46	0.42	1.50	1.55	2.30
TB	450	0	500	50	1.13	0.71	0.96			
Sei	450	0	250	350	0.92	0.63	0.69	1.27	1.33	2.12
Sei	450	0	500	350	1.17	0.83	1.46			

Fig. 8 shows the membrane TMP measured when using back-pulsing for 50 ppm oil concentrations for two fluxes, 250 and 500 $l.m^{-2}.hr^{-1}$. It is clearly shown that when the flux is doubled the fouling rate also increased. The influence of the flux was assessed by comparing the fouling rates between the low and the high flux modes of operation. Table 6 shows the effect of increasing the flux when applying back-pulsing for different feed water characteristics, i.e., surfactant concentration, oil concentration and oil types. The ratio fouling rate (i.e, ratio between fouling rates of 500 and 250 $l.m^{-2}.hr^{-1}$) depends on the membrane pore sizes. From all the difference membrane pore sizes tested, the impact of doubling the flux on the fouling rate has greater effect when increasing the membrane pore size. This corresponds to the feed water properties where the small oil droplet/particle sizes in the range of the membrane pore size promote internal fouling. Furthermore, the more open the membrane is the more oil droplets and particles will accumulate at the membrane surface due to the higher flux. This data also suggests that the dominant fouling mechanism is the internal fouling also for back-pulsing operation

Table 7
Summary back-pulsing on different membrane pore sizes

No	Oil type	Surfactant concentration (mg/l)	Particles concentration (mg/l)	Flux (LMH)	Oil concentration (mg/l)	Fouling rate			Optimum membrane pore size (μm)
						0.1 μm	0.2 μm	0.5 μm	
1	Gli	65	0	250	50	1.92	1.56	1.04	0.5
2	Sei	65	0	250	50	0.79	0.41	0.38	0.5
3	TB	65	0	250	50	1.67	1.29	0.96	0.5
4	Sei	450	0	250	50	0.54	0.25	0.17	0.5
5	TB	450	0	250	50	0.75	0.46	0.42	0.5
6	Sei	450	0	250	350	0.92	0.63	0.69	0.2
7	TB	450	0	250	350	1.28	0.58	0.63	0.2
8	Sei	450	0	500	50	0.83	0.42	0.58	0.2
9	Sei	450	0	500	350	1.17	0.83	1.46	0.2
10	TB	65	0	250	150	4.94	2.11	3.44	0.2
11	TB	65	50	250	150	2.83	2.56	4.72	0.2
12	TB	65	50	250	250	3.83	3.67	6.22	0.2
13	TB	65	0	250	350	5.83	4.78	5.17	0.2
14	TB	65	50	250	350	4.33	3.56	6.50	0.2
15	TB	450	0	500	50	1.13	0.71	0.96	0.2
16	TB	450	0	500	350	1.33	1.38	1.75	0.1
17	Gli	450	0	500	350	1.67	1.96	2.92	0.1
18	TB	450	50	500	350	1.29	1.96	2.38	0.1
19	Sei	450	50	500	350	1.08	1.88	2.21	0.1
20	TB	450	50	500	50	1.04	1.71	2.17	0.1
21	Sei	450	50	500	50	0.75	1.46	1.58	0.1
22	Gli	450	50	500	50	1.08	1.75	2.50	0.1
23	TB	65	0	500	50	2.83	3.46	3.46	0.1
24	TB	65	50	500	50	1.79	3.33	5.21	0.1

Table 8
Permeate oil concentration at different nominal membrane pore size

Oil Condition	Flux (LMH)	BP/NoBP	Permeate quality (ppm)			Oil removal efficiency (R)%		
			0.1 μm	0.2 μm	0.5 μm	0.1 μm	0.2 μm	0.5 μm
TB50P0	250	NoBP	2.27	2.87	4.77	95.5	94.3	90.5
TB50P0	250	BP	0.77	0.80	0.87	98.5	98.4	98.3
TB50P50	250	NoBP	3.54	4.29	5.74	92.9	91.4	88.5
TB50P50	250	BP	1.25	1.40	2.00	97.5	97.2	96.0
TB50P50	500	BP	1.85	2.15	3.67	96.3	95.7	92.7
TB200P0	250	NoBP	5.35	6.90	7.70	97.3	96.6	96.2
TB200P0	250	BP	1.72	2.05	3.95	99.1	99.0	98.0
TB350P0	250	NoBP	7.23	8.43	10.38	97.9	97.6	97.0
TB350P0	250	BP	1.92	2.25	4.85	99.5	99.4	98.6

TB (x): Concentration of oil type-TB at x mg/l; P(mg/l): Concentration of particles (mg/l); BP: Back-pulsing; LMH: l.m–2.hr–1

in that higher flux potentially increases transport of oil droplets/particles into the membrane for the larger pore sizes. The back-pulsing works sufficiently to remove the oil droplets that are larger than the membrane pore size and make a cake layer or external fouling.

(*a* – 0.1 μm) (***b*** – 0.2 μm)
(*c* – 0.5 μm)

3.7. Assessment of back-pulsing effect on different membrane pore sizes

The treatment of the analogue produced water was assessed for the nominal membrane pore sizes of 0.1, 0.2 and 0.5 μm. Different feed characteristics (i.e., oil concentrations, oil types and presences of particles) were used to assess the effect of back-pulsing for the different membrane pore sizes. A summary of the results is presented in Table 7.

Table 7 shows that the choice of optimum membrane pore size is dependent on the feed characteristics and the membrane operation conditions. This is due to: (1) high oil concentrations will increase the concentration of oil droplets on the membrane surface, (2) high fluxes will increase the drag force of permeate and transport of an increased amount of emulsions or suspension to the membrane surface, (3) the presences of colloidal particles which can cause pore plugging / blockage for the larger membrane pore sizes.

The 0.5 μm membrane pore size is better to use when there are minimal colloidal particles present in the feed water and operated with low oil concentrations (i.e., 50 ppm) and at lower fluxes (i.e., 250 $l.m^{-2}.hr^{-1}$). Presences of particles and higher oil concentrations (up to 350 ppm) could be preferably separated using a 0.2 μm membrane pore size at lower fluxes (i.e., 250 $l.m^{-2}.hr^{-1}$). Increasing the flux to 500 $l.m^{-2}.hr^{-1}$., with or without particles and for both low and high oil concentrations is preferably treated by selecting a 0.1 μm membrane pore size.

3.8. Oil retention

The oil retention of the membrane was measured and reported as the concentration of oil in the permeate from different membrane operations at different membrane pore sizes, shown in Table 8. The removal efficiencies were determined using equation (3).

The oil content in the permeate for filtration with and without back-pulsing increases with increasing membrane pore size [6] and thus reduces the oil removal efficiency (R). The removal efficiency (R) without back-pulsing at different membrane operation at flux of 250 $l.m^{-2}.hr^{-1}$ was in the range between 88 to 98% as a function of testing conditions. The use of back-pulsing increases the oil removal efficiency up to the range between 96 to 99.5%.

The higher oil passage for the larger membrane pore size is due to the fine oil droplets more easily passing through the membrane structure. Besides, the critical capillary pressure is lower at bigger membrane pore sizes. Therefore, bigger membrane pore sizes only require lower operating pressure for the oil to wet and pass the membrane. The fine oil droplets or particles also increase the operating pressures due to the higher fouling tendency, and as a consequence, the fine oil droplets and the larger membrane pore size contribute to more oil passing through to the permeate.

In addition to reducing the fouling rate (or TMP), the use of back-pulsing also reduced the concentration of oil in the permeate significantly, thus increasing the removal efficiency. The removal efficiency (R) for different operating conditions at flux of 250 $l.m^{-2}.hr^{-1}$. was in the range between 96 to 99.5%. The removal efficiency increased with increasing oil concentration, i.e., 98.5% at 50 ppm vs. 99.5% at 350 ppm (Table 8).

Although the oil concentration in the permeate increased with increasing concentration in the feed water, the oil concentration in the permeate did not increase significantly for the feed water oil concentration range tested (i.e., 50–350 ppm). This is probably due to a more efficient removal of the oil layer on the membrane surface by back-pulsing and thus reducing the amount of oil that could pass through the membrane. Furthermore, the back-pulsing results show that the target of very low oil concentration in the permeate (i.e., <5 ppm) could be achieved in all of the operating conditions tested. The use of back-pulsing shows the possibility to achieve the target oil concentration in the permeate even with a larger membrane pore size.

4. Conclusions

The influence of feed properties (i.e., oil concentration, oil types, and particles) and membrane flux on the effectiveness of back-pulsing on membrane filtration performance were investigated in this study. Without back-pulsing to control and minimize membrane fouling, the fouling behaviour strongly depends on the feed properties, oil types/oil droplet size, oil concentration, and colloidal particle concentrations. However, when back-pulsing is employed the effect of feed water characteristics is less. Membrane fouling rates were significantly reduced when back-pulsing is employed compared to a more conventional backwashing procedure. Fouling rate assessment showed that lower oil concentrations, larger oil droplet sizes and size distribution, and minimal amount of colloidal particles are expected to give a better and more sustainable long-term membrane operation. From the feed properties assessment, back-pulsing showed a high efficiency in improving membrane performance when the fouling is dominated by external fouling (i.e., cake-layer formation) which is determined by the particulate sizes larger than the nominal membrane pore size, and is less effective for internal fouling (i.e., pore plugging and pore blockage). In general, lower fluxes are expected to give a longer membrane filtration operation between required chemical enhanced cleaning due to irreversible fouling. Further, a membrane pore size of 0.1 μm was more effective to use when colloidal particles are present in the feed water as this fraction is mainly responsible for internal membrane fouling. The 0.2 μm pore size membrane was found to be a good option for feed waters with high oil concentrations or high flux operation, while the 0.5 μm membrane pore size is viewed as good only for the lowest oil concentration conditions. The permeate quality was also improved when the back-pulsing technique was employed. Varying oil concentrations between 50 to 350 ppm of oil did not result in a significant increase of oil in the permeate compared to operation without back-pulsing. The results when using the back-pulsing technique showed that the target oil concentration in the permeate of <5 ppm could be achieved for all the operating conditions tested in this study.

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