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Rejection of Oil Emulsion using Tubular Surface Filters

(Penyingkiran Emulsi Minyak dengan Penuras Permukaan Tiub)

MUHAMMAD ABBAS AHMAD ZAINI*, IAIN W. CUMMING & RICHARD G. HOLDICH

ABSTRACT

This study was aimed to characterize the rejection of oil emulsion by two tubular surface filters, namely 13 μm slots and 4 μm circular pores. A 17 mm helix was incorporated inside the filters to increase the wall shear stress. Rejection of oil, throughputs and flux decay were measured at varying shear stresses (R_w) and transmembrane pressures (TMP). The results showed that the13 μm slots produced a fairly good oil rejection of 22 μm at R_w=0.26 Pa and TMP=6.9 kPa, while the circular pores gave the rejection of 14.4 μm at R_w=0.21 Pa and TMP=34.5 kPa. The results suggested that the circular pores filter could achieve a better oil retention under lower TMP, while the slots filter could be satisfactorily operated under moderate shear stress and low TMP. The increase of TMP offered poor oil rejection by both filters and this was also true for slots-helix configuration.

Keywords: Helical insert; microfiltration; oil emulsion; rejection; surface filter

ABSTRAK

Kajian ini bertujuan untuk mencirikan penyingkiran emulsi minyak oleh dua penuras permukaan tiub, iaitu 13 μm bukaan celah dan 4 μm bukaan lingkar. Tekanan ricih dinding ditingkatkan dengan menggabungkan 17 mm pilin ke dalam penuras. Penyingkiran minyak, hasil turasan dan kegagalan fluk telah diukur pada tegasan ricih (R_w) dan tekanan transmembran (TMP) yang berbeza. Hasil kajian menunjukkan bahawa 13 μm bukaan celah menghasilkan penyingkiran minyak yang agak baik iaitu 22 μm pada R_w=0.26 Pa dan TMP=6.9 kPa, manakala bukaan lingkar memberikan penyingkiran 14.4 μm pada R_w=0.21 Pa dan TMP=34.5 kPa. Hasil kajian mencadangkan bahawa penuras bukaan lingkar boleh mencapai penahanan minyak lebih baik di bawah TMP yang lebih rendah, manakala penuras bukaan celah boleh dikendalikan dengan sempurna di bawah tegasan ricih sederhana dan TMP yang rendah. Peningkatan TMP memberikan penyingkiran minyak yang rendah bagi kedua-dua penuras dan begitu juga dengan penurasan bukaan celah-pilin.

Kata kunci: Emulsi minyak; penurasan mikro; penuras permukaan; penyingkiran; pilin masukan

INTRODUCTION

There has been increasing concern over the presence of stable oil dispersed in water from a number of associated process industries, bilge water from ships and offshore oil production. This environmental problem not only creates hazardous conditions to aquatic creatures but also affect human health and sea activities. Scientists and engineers have now been searching for effective measures to abate this matter and at the same time complying with stringent allowable oil concentration of 30 mg/L (International Maritime Organization 1972).

For many years, hydrocyclone has been used to offset this problem, but its efficiency reduces with decreasing droplets size (Hargreaves & Silvester 1990; Wolbert et al. 1995). It has been reported that ultrafiltration may succeed while removing small oil drops lesser than 10 μm, yet it only works for low volume of effluents due to severe fouling of filter (Chakrabarty et al. 2008). Hence, larger surface area of filter is required to accommodate a larger volume of effluents and this of course will incur additional costs.

In order to achieve the desirable throughputs at industrial scale, microfiltration of bigger pore rating than that of ultrafiltration has been introduced and investigated over the past 15 years. Yet the problems of fouling and low throughputs still persist (Koltuniewicz et al. 1995; Mueller et al. 1997; Ohya et al. 1998; Wang et al. 2009). Both ultrafiltration and microfiltration commonly operates under significant depth filtration to which the oil drops are allowed to penetrate the tortuous flow network and become captured throughout the depth of the filter matrix. This has been the main reason behind the easily clogged filter that leads to internal fouling (Cumming et al. 1999a).

To overcome this difficulty, the surface filters, having the micron size pores passing directly from one side to another have been introduced (Cumming et al. 1999b). They are relatively new and less common compared with depth filters but are more likely to hinder irreversible internal fouling that may lead to low throughputs (Cumming et al. 2000; Holdich et al. 2003). Its mechanism is similar to that of sieves but having a micron sizes pore rating. These filters were fabricated into tubular form to induce cross-flow, because such mode has been widely known to be more efficient in comparison with dead-end (Koltuniewicz et al. 1995). The objective of our present work was therefore to characterize the rejection of oil emulsion by employing two tubular surface filters, i.e. 13 µm slots and 4 µm circular pores. A 17 mm helix was introduced into the filter to exert shear stress to the filter wall. The effects of varying shear stresses (R_w) and transmembrane pressures (TMP) on oil rejection, throughputs and flux decay were evaluated and discussed.

MATERIALS AND METHODS

PREPARATION OF OIL EMULSION

Synthetic oil emulsion was prepared from sunflower oil and surfactant Tween20 (2.5 wt%). The mixture was then homogenized and subsequently diluted to 1000 ppm to imitate typical concentration of produced water at offshore oil platforms. Stability of oil emulsion was analyzed in four media, i.e. distilled water, tap water, 0.25 M saline and 0.5 M saline. Two saline solutions were used to mimic the concentration of seawater. The mixtures were stirred and gently shaken twice a day for four successive days to induce dispersion. Size distribution of droplets in each media for each day was analyzed using Coulter Counter™ (Model TAII, Coulter Electronics) employing a 100 µm orifice tube.

EXPERIMENTAL RIG AND PROCEDURES

Schematic diagram of experimental rig for this work is shown in Figure 1. It is equipped with a backflush unit on the top of module to alleviate surface fouling. The peristaltic pump was calibrated against flow rate, to which two different flow rates were employed in this work, i.e. 4 and 5 L/min.

To induce back pressure, the retentate valve was partially closed so that the pressure gauge in permeate line gives a measurable reading. The permeate valve was slowly opened to collect permeate and the volume was measured against time. Transmembrane pressure (TMP) and the driving force of flux across the surface filter was estimated by the following expression (Cumming et al. 1999a),

$$TMP = \frac{P_{feed} + P_{retentate}}{2} - P_{permeate}, \quad (1)$$

where, P_{feed}, $P_{retentate}$ and $P_{permeate}$ are respectively the pressure reading at feed, retentate and permeate lines.

SURFACE FILTERS AND HELICAL INSERT

SEM images and array of (a) 13 µm slots (width: 13 µm, length: 400 µm) and (b) 4 µm circular pores are given in Figure 2. These filters were made of metal (nickel) and prepared through nuclear bombardment followed by chemical etching. Details on the characterization of these filters have been described elsewhere (Cumming et al. 1999b, 2000; Holdich et al. 2003).

The characteristics of oil emulsion in microfiltration were also evaluated by inserting a 17 mm pitch helix. Schematic representation of helix within the surface filter is shown in Figure 3, and its geometries are summarized in Table 1.

The presence of helix inside the surface filter leads to a considerable change in the flow pattern of oil emulsion. Therefore, the comparison between configurations of with and without helix is better done through constant wall shear stress, R_w, which is a measure of forces exerted upon the surface of filter as a result of liquid flow across the channel. The wall shear stress for a liquid in contact with the surface of filter can be expressed using the theory of laminar boundary layer by incorporating dynamic viscosity, μ and channel velocity, v_c.

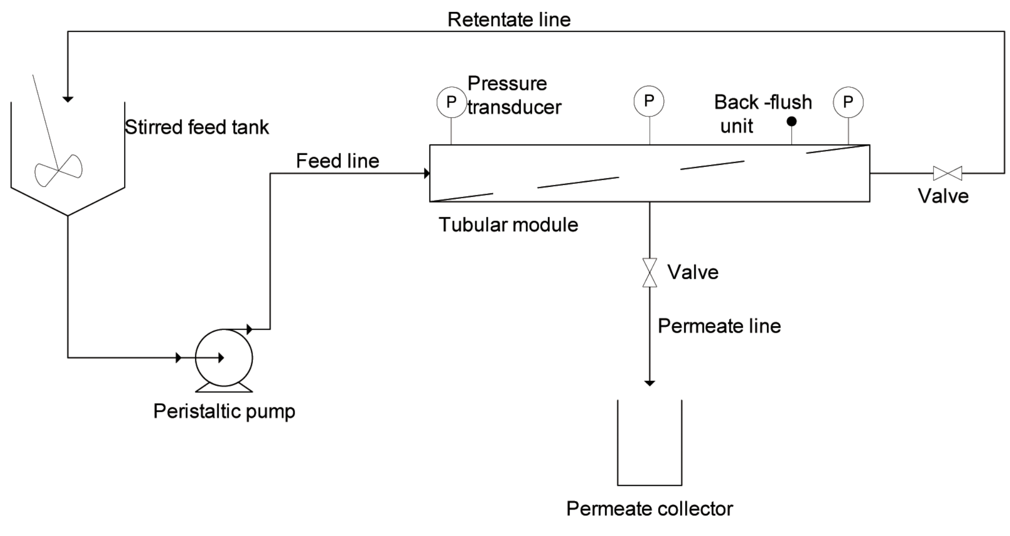


FIGURE 1. Cross-flow microfiltration rig

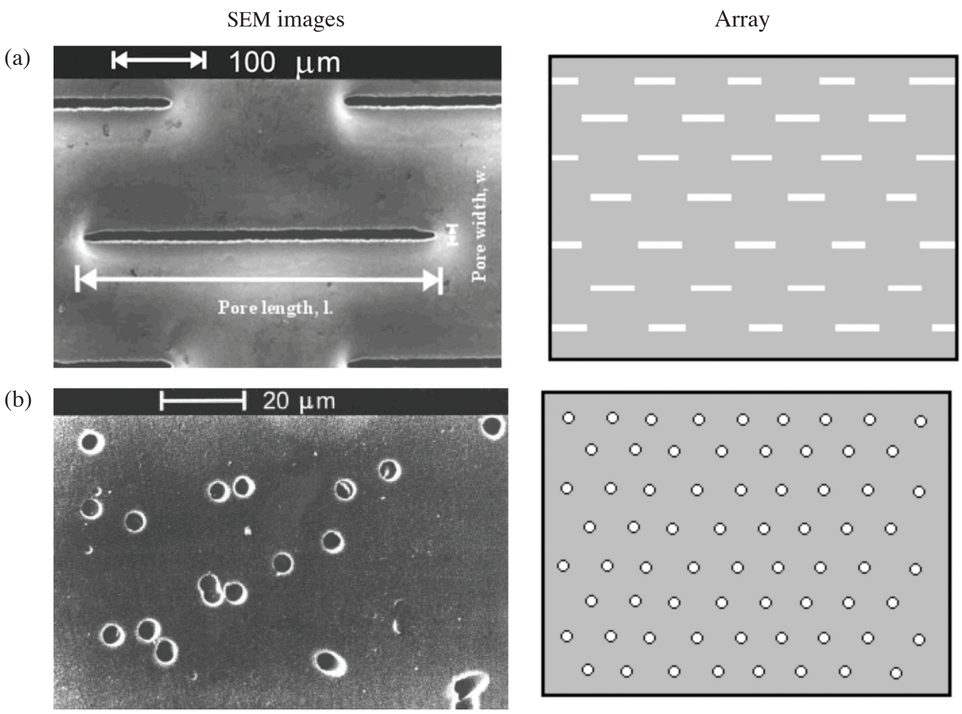


FIGURE 2. SEM images and array of surface filters (a) slots (Kosvintsev et al. 2007) and (b) circular pores (Cumming et al. 2000)

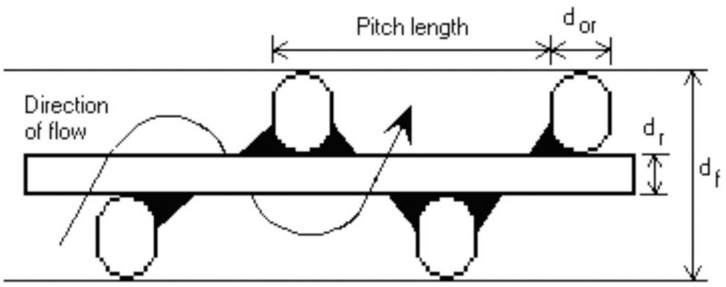


FIGURE 3. Visualization of helix inside the surface filter

TABLE 1. Geometry of helix

Diameter of tubular filter, d_f	1.4 cm
Rod diameter, d_r	0.5 cm
Diameter of o-ring, d_{or}	0.45 cm
Pitch length	1.7 cm
Helical path length	64.5 cm
Rod length	42.5 cm

$$R_w = \frac{6\mu v_c}{h}. \quad (2)$$

For non-insert configuration, the height of channel, h is simply the channel diameter, d_f, while h becomes $0.5(d_f - d_r)$ for configuration with helix inside the tubular surface filter. The flow properties of the two configurations used in this work is tabulated in Table 2.

PERFORMANCE OF FILTER

Filtration data for each surface filter at different configurations and flow rates were obtained from volume of permeate collected over time. These data were then translated into normalized flux rate against time to evaluate the throughputs and flux decay.

Rejection (or also known as grade efficiency) can be described as fractions (grades or size ranges) of oil

TABLE 2. Flow properties within tubular surface filter at different configurations

Filter configuration	Hydraulic diameter, d_h (cm)	Channel area, $A_c \times 10^4$ (m^2)	Feed flow rate (L/min)	Channel velocity, v_c (m/s)	Shear stress, R_w (Pa)
Without insert	1.4	1.54	4	0.43	0.21
			5	0.54	0.26
Inserted with 17 mm helix	0.7	0.726	4	0.92	1.40
			5	1.15	1.74

emulsion that could be retained from entering permeate. It can be mathematically expressed as follows:

$$rejection = \left(1 - \frac{permeate\ concentration\ in\ grade}{feed\ concentration\ in\ grade}\right) \times 100\%. \quad (3)$$

The concentrations of oil emulsion in feed and permeate were measured using total organic carbon analyzer (TOC 1200, ThermoFisher Scientific), while the droplets size distributions were obtained using coulter counter model TAII employing a 100 μm diameter orifice.

To assess the rejection of oil emulsion, two independent variables, i.e. shear stress and TMP were manipulated and correlated to the performance of surface filters while filtering the oil emulsion.

Results and Discussion

STABILITY OF OIL EMULSION

Figure 4 shows the particle size distribution of 1,000 ppm oil emulsion in four media after four days. There is no considerable difference could be observed on oil drops size at every volume undersize in different media.

It also shows that the biggest oil drops are ranging between 40 and 50 μm, while the oil emulsion is dominant (in volume; at 50% volume undersize) with 16 μm drops size. Thus, the synthetic oil emulsion prepared for this work is visibly stable and appropriate to be regarded as produced water. Oil emulsion diluted in distilled water was therefore utilized in the following investigations.

EFFECTS OF SHEAR STRESS AND FILTER CONFIGURATION

The practical purpose of adding the helix within the tubular surface filters is to enhance the shear stress exerted on the surface of filter. It is already known that, the higher the shear stress, the greater the forces sweeping or cleaning the surface of filter, thus theoreritically reducing the particulate deposition that can cause fouling.

Figure 5 displays the effect of shear stress (R_w) on the performance of slots filter while filtering the oil emulsion. As shown in Figure 5(a), R_w of 0.26 Pa was effective to retain oil drops greater than 22.7 μm. However, this is 1.7 times higher than the pore rating of the slots due to the deformable nature of the oil drops. At a lower R_w of 0.21 Pa, the slots filter was only able to achieve 76% rejection efficiency of droplets greater than 28.7 μm. It indicates that the shear stress induced at low cross flow rate (4 L/min) and with non-helix configuration was unable to provide

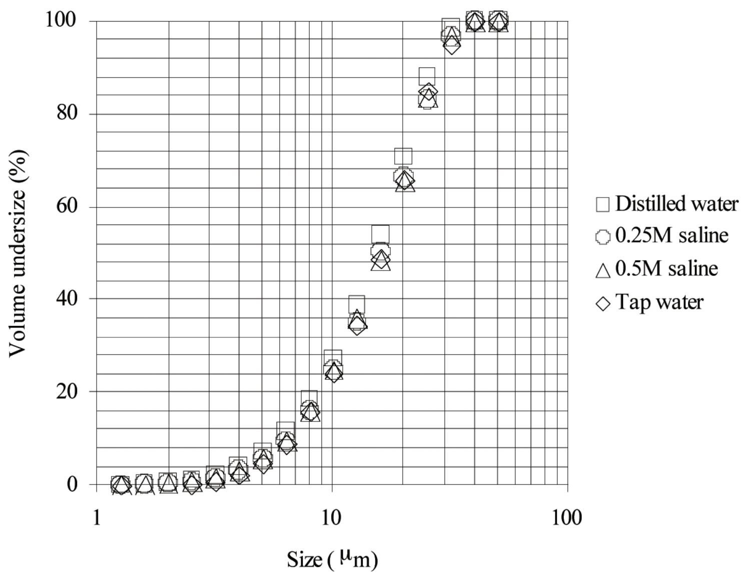


FIGURE 4. Oil emulsion stability in four different media

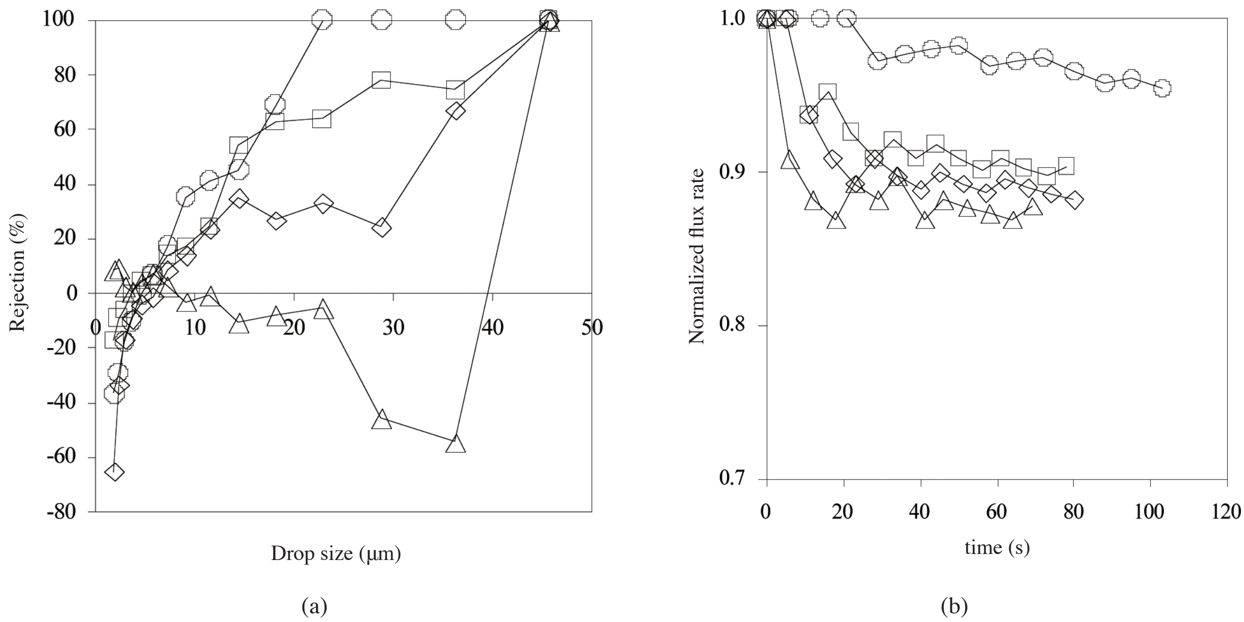
sufficient surface sweeping to impede greater oil drops to deform and so penetrate through the slots. The retention of oil drops, however became worst as the shear stress increased by inserting the helix inside the slots filter. It is suggested that the sweeping effect imposed by higher shear stress may also squeeze and assist the deformable oil drops to pass through the slots (Kosvintsev et al. 2007). It is also noted that the rejection of droplets at R_w of 1.74 Pa was better than that at 1.40 Pa. This could be explained through the possibility of the deformable droplets to partially block the throat of slots at higher shear stress, thus preventing bigger oil drops to pass through (Zaini et al. 2010). Generally, the ability to reduce resistance to filtration by using helix did not show any improvement on the filtration of oil emulsion by using slots filter, yet the retention of oil drops became much poorer as compared with the non-helix configuration.

The extent of flux decay when using slots at varying shear stresses is illustrated in Figure 5(b). At constant transmembrane pressure (TMP) of 6.9 kPa, the throughputs of slots range from 9100 to 10800 L/m^2.h, and was fairly stable with time. The filtration of oil emulsion at R_w of 0.26 Pa showed the lowest flux decay of about 5%, while the others remained high at 10-12%. This is consistent with the results shown in Figure 5(a). It can be concluded that 0.26 Pa was a suitable R_w to clean the surface of slots so as to reduce the resistance to flow, alleviate fouling and promote better droplets rejection.

Performance of 4 μm circular pores filter while filtering oil emulsion at a constant TMP of 34.5 kPa and different shear stresses is shown in Figure 6. Unlike the slots, the circular pores was effective to retain oil drops at a lower R_w of 0.21 Pa. At this condition, the droplets greater than 14.4 μm was able to be removed, yet it is 3.6 times bigger than the filter pore rating. As the R_w increases to 0.26 Pa, the cut-off reduces to 28.7 μm while the retention becomes worse at R_w=1.40 Pa. Notwithstanding that, the retention of oil drops greater than 28.7 μm was observed at R_w of 1.74 Pa, which is much better than that at 0.26 Pa. The positive effect of higher shear stress to provide sufficient cleaning on the surface of filter is also apparent in Figure 6(a), where the rejection of oil drops varying from 11.4 to 22.8 μm was found significantly improve from 45% at R_w=0.26 Pa to 80% at R_w=1.74 Pa. Moreover, the retention of oil drops between 4.5 and 11.3 μm at R_w of 1.74 Pa was far better than that at 0.21 Pa. Obviously, such trends shown by circular pores are somewhat identical to that of slots. Yet, it is believed that a much better oil rejection could be achieved at a higher shear stress under a lower TMP.

Figure 6(b) shows the reduction of flux against time for circular pores filter. The pressure across the filter was set at minimum of 34.5 kPa. Permeate was visibly unable to be collected at TMP below the aforesaid value because of small pore rating. Trends shown in Figure 6(b) are in agreement with the results obtained for rejection efficiency, at which the flux decay improved from 14 to 25% with increasing shear stress. As the shear stress increases, the speed of sweeping action induced by the flow assists the dismissal of bigger droplets that resting and accumulate on the pores surface. This mechanism is somehow opposite with that of slots (partial blocking), most likely due to the nature of smaller pore rating possessed by the circular pores filter. Throughputs from this filtration were two orders of magnitude lower in comparison with that of slots, i.e. between 32 and 74 L/m^2.h and also slightly decreased with increasing shear stress.

It is also interesting to observe that the rejection depicts negative values for drops size lesser than 6 μm for both



(a) (b)

FIGURE 5. Effect of shear stress on rejection of oil emulsion using 13 μm slots filter: (a) grade efficiency and (b) flux decay Symbols, (□): R_w=0.21 Pa, (○): R_w=0.26 Pa, (Δ): R_w=1.40 Pa, (Δ): R_w=1.74 Pa. TMP was set at minimum of 6.9 kPa

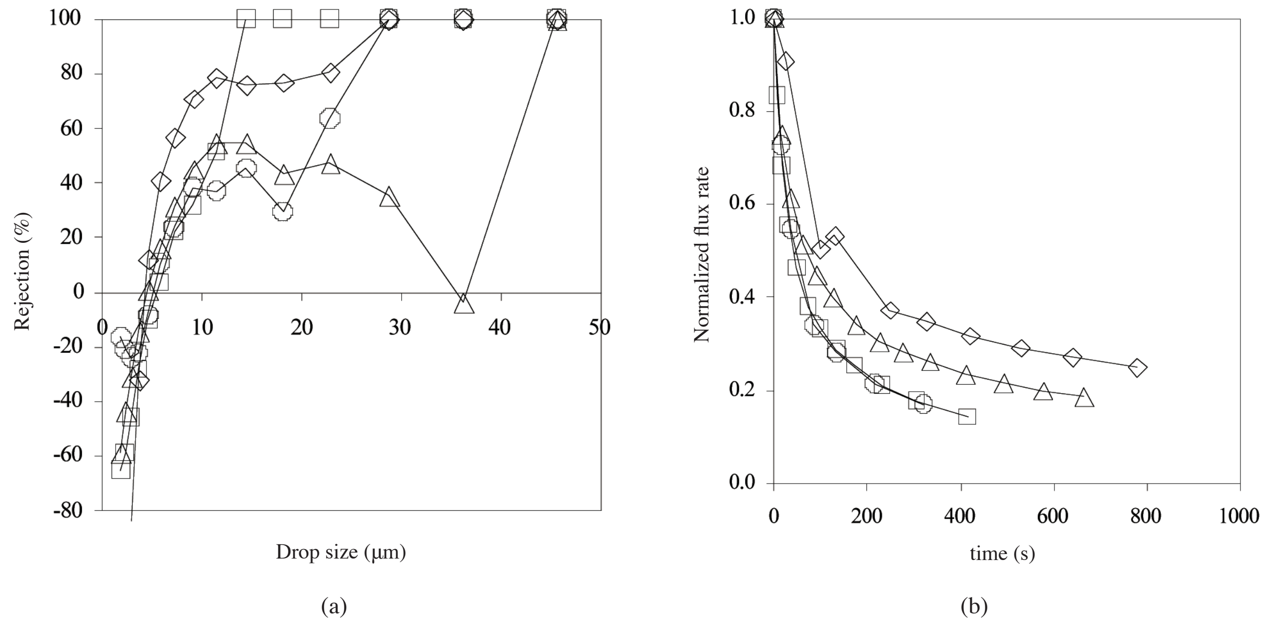


FIGURE 6. Effect of shear stress on rejection of oil emulsion using 4 μm circular pores filter: (a) grade efficiency and (b) flux decay Symbols, (□): R_w=0.21 Pa, (○): R_w=0.26 Pa, (Δ): R_w=1.40 Pa, (◊): R_w=1.74 Pa. TMP was set at minimum of 34.5 kPa

surface filters. This could be due to the breakage of such droplets to form fractions of smaller oil drops once leaving the filters (Desse et al. 2011; Gordon 1959; Varanasi et al. 1994). As a result, the mass or concentration of smaller oil drops in permeate becomes greater, thus decreasing the rejection efficiency. This is more prevalence as the R_w increases to 1.74 Pa. Such instability could be caused by lower surface tension of the emulsion and hydrodynamic forces exerted on the filter (Ullah et al. 2012; Varanasi et al.1994; Zaini et al. 2010).

At R_w of 1.40 Pa, both filters demonstrated a rapid decline in oil rejection for drops size above 22.8 μm. Such phenomenon has also been reported in a number of literatures (Aul & Olbricht 1991; Olbricht & Kung 1987). It occurs when the compositions of such droplets in permeate is significantly higher and each oil drop holds attractive forces due to the influence of shear stress. The oil drops then tend to coalesce to increase the concentration of much bigger oil drops in permeates thus decreasing the rejection efficiency.

EFFECT OF TRANSMEMBRANE PRESSURE (TMP)

The main purpose of increasing the pressure across the filter is to increase the throughputs. The effect of transmembrane pressure on the rejection and throughputs of the two surface filters is tabulated in Tables 3 and 4.

As can be seen in Table 3, the performance of slots at all shear stresses deteriorates as TMP increases to 10.3 kPa. Apart from that of shear stress, TMP also influences the oil drops to deform and penetrate through the pores. Throughputs were also consistently decreased with increasing TMP, while the flux decays remained fairly

TABLE 3. Effect of TMP on the performance of 13 μm slots filter

Shear stress, R_w (Pa)	TMP (kPa)	Cut-off (μm)	Flux rate (L/m^2 h)	Flux decay (%)
	6.9	-	10851	10
0.21	8.6	-	6989	12
	10.3	-	2866	10
	6.9	22.7	9760	5
0.26	8.6	22.7	6872	13
	10.3	-	2162	11
	6.9	-	9163	12
1.40	8.6	-	6651	37
	10.3	-	2766	13
	6.9	-	10573	12
1.74	8.6	-	7049	33
	10.3	-	2798	12

TABLE 4. Effect of TMP on the performance of 4 μm circular pores filter

Shear stress, R_w (Pa)	TMP (kPa)	Cut-off (μm)	Flux rate (L/m^2.h)	Flux decay (%)
	34.5	14.4	74	86
0.21	41.4	-	76	86
	55.2	-	85	67
	34.5	28.7	55	86
0.26	41.4	36.2	60	65
	55.2	-	72	72
	34.5	-	44	81
1.40	41.4	-	54	86
	55.2	-	54	88
	34.5	28.7	32	75
1.74	41.4	-	58	84
	55.2	-	64	85

stable. This indicates that the throats of slots became more clogged with oil drops at a higher TMP thus preventing a smooth flow of permeate flux, while at the same time the shear stress aid to exclude oil drops that lodged on the slots surface so as to decrease the oil rejection. At R_w= 0.26 Pa and TMP = 8.6 kPa, slots filter was able to maintain similar retention of oil drops greater than 22.7 μm, yet the percentage of flux decay was slightly increased while the throughputs was significantly decreased. It can be concluded that, the retention of oil drops using slots surface filter could be suitably operated under moderate shear stress and low TMP.

From Table 4, the effect of TMP on the performance of circular pores is somewhat identical to that of slots, at which the rejection efficiency decreases with increasing TMP. There was also no considerable improvement that could be observed in throughputs and flux decay of circular pores by increasing the TMP. Therefore, the filtration of oil emulsion using circular pores filter could be successfully achieved under low or high shear stress and low TMP.

Conclusion

The stability of synthetic oil emulsion prepared from sunflower oil and Tween20 was established on the basis of particle size distribution. The 13 μm slots filter was able to achieve two orders of magnitude higher throughputs than the 4 μm circular pores filter. The slots was effective to retain oil drops under moderate shear stress and low transmembrane pressure, while the filtration using circular pores was suitable to be carried out at low or high shear stress and low transmembrane pressure. It was found that an increase in transmembrane pressure decreased the rejection efficiency of oil drops by both filters. Furthermore, the inclusion of helix inside the slots filter was a fruitless effort because it introduced unnecessary resistance inside the channel which decreased the rejection. The surface filters are promising candidate to counter the problem arises from oily-water pollution, yet suitable operating conditions, i.e. flow rate, shear stress and transmembrane pressure have to be closely monitored in order to minimize the effect of droplets deformation, break-up and coalescence so as to gain better oil rejection.

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