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CROSSFLOW MICROFILTRATION OF OIL IN WATER EMULSION VIA TUBULAR FILTERS: EVALUATION BY MATHEMATICAL MODELS ON DROPLET DEFORMATION AND FILTRATION

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Abstract. Three types of tubular filters, i.e. 13 μm slots, 4 μm circular pores and 0.45 μm ceramic membranes, and 13 μm flat sheet slots were used to challenge 1000 ppm oil in water emulsion, the test liquid. The rejection of oil drops by pore size was also evaluated by incorporating a 17 mm pitch helix within the tubular slots and circular pore filters. The experimental data were analysed using filtration models. The model of droplet deformation indicates that poor rejection was caused by a low interfacial tension of emulsion system and that the oil drops could easily penetrate the filters even at low transmembrane pressure. However, the results of filtration models suggested that the use of slots was more likely to reduce the flux decay and the incorporation of a helix can greatly reduced the resistance to filtration.

Keywords: Droplet deformation; filtration models; oil in water emulsion

Abstrak. Tiga jenis penuras tiub, iaitu 13 μm bukaan celah, 4 μm bukaan lingkar dan 0.45 μm membran seramik, serta penuras kepingan 13 μm bukaan celah telah digunakan untuk menapis 1000 ppm emulsi minyak dalam air. Penahanan zarah minyak berdasarkan saiz bukaan penuras juga diuji dengan memasukkan 17 mm pilin ke dalam penuras tiub bukaan celah dan bukaan lingkar. Data ujikaji telah dinilai dengan menggunakan model-model penurasan. Model ubah bentuk zarah menunjukkan bahawa kelemahan penyingkiran minyak adalah disebabkan oleh tegangan antara muka sistem emulsi yang rendah dan menyebabkan zarah minyak mudah melepasi bukaan penuras meskipun pada tekanan operasi membran yang rendah. Walau bagaimanapun, keputusan yang ditunjukkan oleh model-model penurasan mencadangkan bahawa penuras mikro bukaan celah dapat mengurangkan kegagalan fluk, serta penggunaan pilin boleh mengurangkan rintangan terhadap penurasan.

Kata kunci: Ubah bentuk zarah; model penurasan; emulsi minyak dalam air

1.0 INTRODUCTION

Pollution of oily wastes not only creates hazardous conditions to aquatic living creatures but also can influence human health. Settling tanks, hydrocyclones and centrifuges have

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been used to treat stable oil in water emulsions. However, they are expensive due to high investment requirement and only efficient when removing particles greater than 10 μm [1, 2]. A promising technique for the removal of stable water-soluble oily wastes is microfiltration. A number of published literatures indicate that microfiltration can remove oil drops dispersed in liquid in the range of 0.1 μm [2-5]. In other development, advanced ceramic and special hydrophilic polymeric with tight pore size distribution have been investigated, and shown effective retention of oil, but suffer in fouling and rapid reduction of flux [5-7].

In this work, we examined the effect of tubular (crossflow) setting together with helical insert configuration on the retention of oil in water emulsion. A 17 mm pitch helix was imposed into two different tubular filters, namely 13 μm slots and 4 μm circular pores. A 0.45 μm ceramic membrane and 13 μm flat sheet slots (dead-end setting) were employed for comparison purpose. The test liquid was oil in water emulsion having a concentration of 1000 ppm, which was prepared from sunflower oil and Tween 20 (surfactant). Filtration of oil in water emulsion was carried out at transmembrane pressure of 2000 to 70000 Pa, average flow rate of 5 L/min for duration up to 15 minutes. The data obtained from this work were evaluated using mathematical models on droplet deformation and filtration.

These two models are important to further explain and give better understanding of the phenomenon that takes place during the microfiltration of dispersed oil. The model of droplet deformation is described as a method to predict the oil rejection. This model combines the effect of interfacial tension and contact angle to determine the possibility of drops deforming sufficiently to pass through the filter pores. The filtration models, on the other hand, are used to describe the flux decay during microfiltration. There are four equations that need to be examined to figure out the best and appropriate models to represent the flux decay.

2.0 MATHEMATICAL MODELS

2.1 Method to Predict the Oil Rejection

The model of droplet deformation assumes that the rejection of an oil drop is determined by its ability to deform and flow through a pore at the same velocity as the permeate, and that there is no rejection due to hydrodynamic forces on the drop. This model applied the Young-Laplace equation across the leading and trailing interfaces to determine the minimum pressure required to enable a drop to enter and pass through the pore. It can be illustrated in Figure 1 as follows [8].

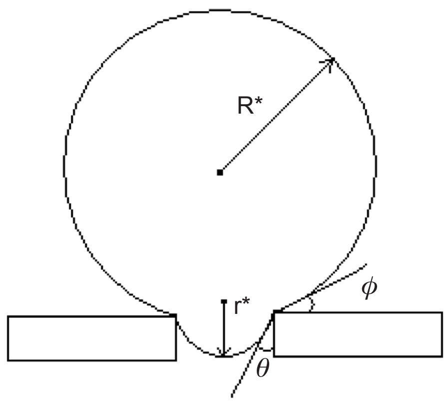

Figure 1 Illustration of an oil drop at a pore entrance

The leading edge of the drop is located within the internal pore surface at contact angle θ, between the filter material and the oil-water interface. The rear of the drop meets the outside surface of filter at contact angle ϕ. Correlation between the mass of the leading and trailing edges of the drop located in a pore, and the original drop mass gives the original drop radius, r_d as simplified in Equation (1) [8, 9].

$$r_d = \left[\frac{3}{4} \left\{ \begin{array}{l} \frac{r_p^3}{3\cos^3\theta}\left(2 - 3\sin\theta + \sin^3\theta\right) + \frac{2}{3}R^{*2}\sqrt{R^{*2} - r_p^2} \\ + \frac{1}{3}r_p^2\sqrt{R^{*2} - r_p^2} + \frac{2}{3}R^{*3} \end{array} \right\} \right]^{\frac{1}{3}} \tag{1}$$

Where r_p is the pore radius, θ is the drop contact angle of leading edge, and R^* and r^* are the drop radius of the trailing and leading curvatures, respectively. The radius of the trailing curvature, R^*, that would fail to pass through the pore is calculated using the Young-Laplace equation as represented in Equation (2) [8], where γ is the interfacial tension, and P_c is the transmembrane pressure.

$$R^* = \frac{2r_p\gamma}{2\gamma\cos\theta - r_pP_c} \tag{2}$$

The drop size (drop size equals to two times drop radius, $2r_d$) that would be retained by the filter pore can be calculated by embedding Equation (2) into Equation (1). In this study, the value of θ was set at 0, which indicates that the drops successfully penetrate the pore, wet on the surface of filter and deform from their original spherical geometry. Thus, the effect of transmembrane pressure, P_c on the drop size, r_d can be determined directly from this assumption.

2.2 Filtration Models

In basic filtration, the cake accumulates on the surface of filter medium. As the time passes by, the cake starts to act as the additional filter layer to which further particles continue to deposit. Such process takes place until the filtrate rate falls to an unacceptable level. Similar goes for crossflow microfiltration of dispersed oil, whereby the nature of flux decline is due to the deposition of material on the surface and within the filter pores. The internal fouling is relevant to situation in which the feed concentration is sufficiently high to form bridged cake on the membrane surface [10, 11].

The incompressible, dead-end filtration equation is used in the modelling of flux decay. This equation is developed from Darcy's Law, which is represented in Equation (3) as follows.

$$\frac{dt}{dV} = \frac{\mu\alpha c}{A^2 \Delta P} V + \frac{\mu R_m}{A\Delta P} \tag{3}$$

Where $\frac{dt}{dV}$ is the differential form of time over volume, V (m^3) is the volume of permeate, μ (kg/m.s) is the viscosity of fluid, α (m/kg) is the specific cake resistance, c (kg/m^3) is the concentration of dispersed oil, A (m^2) is the filtration area, R_m (m^{-1}) is the membrane resistance and ΔP (Pa) is the transmembrane pressure. The most crucial factor in filtration is the permeability of filter cake, which often expressed through specific cake resistance, as well as the membrane resistance [10].

Integrating Equation (3) gives,

$$\frac{t}{V} = \frac{1}{2}\frac{\mu\alpha c}{A^2 \Delta P} V + \frac{\mu R_m}{A\Delta P} \tag{4}$$

where the values of α and R_m can be calculated from the slope and intercept of t/V against V.

Various forms of Darcy's Law have been applied to represent the flux decay phenomenon during the crossflow microfiltration [11]. They are,

(a) Cake filtration model

$$\frac{t}{V} = \alpha_1 V + B_1 \tag{5}$$

(b) Standard filtration model

$$\frac{t}{V} = \alpha_2 t + B_2 \tag{6}$$

(c) Intermediate blocking model

$$\frac{1}{J} = \alpha_3 t + B_3 \tag{7}$$

(d) Complete blocking model

$$-In\left(\frac{J}{J_o}\right) = \alpha_4 t + \beta_4 \tag{8}$$

where α and β subscripted by 1-4 refer to different constants related to specific cake resistance and membrane resistance, respectively, while J and J_o is the flux and initial flux, respectively. These models are used with an assumption that the pressure difference over the filtration area is constant. The basic fundamental theory of the constant pressure filtration is to treat the pores as a series of parallel non-connected cylindrical tubes, where a filter cake may form during filtration and the deposits can narrow the flow channel in a manner to partially or completely block the pores [11].

3.0 RESULTS AND DISCUSSION

Figure 2 shows the effect of transmembrane pressure on the size of drops that could successfully penetrate the pores of three different tubular filters at 0^o contact angle.

For slots, the effect of transmembrane pressure is notable due to the size of oil drops becomes larger even at a very low transmembrane pressure of about 1000 Pa. For the success of oil filtration using slots, it should be carried out at a much lower operating pressure to ensure that the size of drops to permeate is lesser than its pore rating. The critical operating pressure for circular pores is about 4500 Pa, which is 4.5 times higher than that of slots. While, the ceramic membrane could sustain a greater transmembrane pressure up to 40000 Pa without leaving the drops greater than its pore rating to permeate.

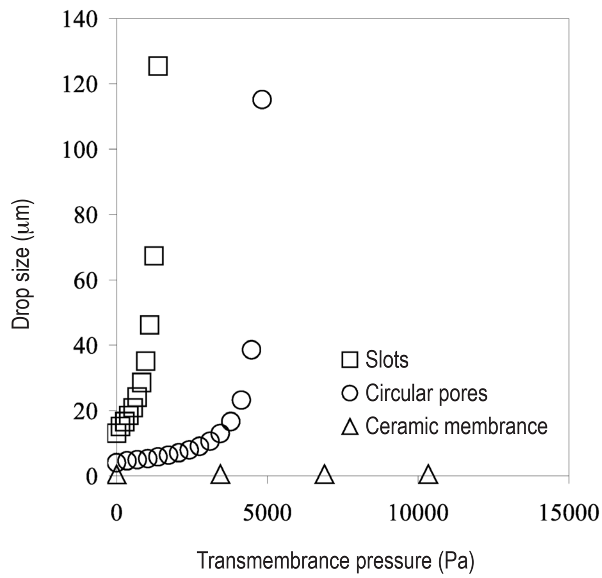

Figure 2 Effect of transmembrane pressure on drop size

Important factor that may influence the droplet deformation, as represented in Equation (2) is the interfacial tension. Its value for the test liquid was 0.005 N/m, which is much lesser than that of typical oily water (0.044 N/m). When an emulsion is formed, the surface contact area increases, thus increasing the total interfacial tension. Then, the molecules tend to migrant from the interface into the bulk phase and reducing the actual contact area between dissimilar molecules. This finally will lower the interfacial tension of the emulsion produced and results in the formation of small drops. This could be reason why the oil drops easily penetrate the pores of slots and circular pores even at low operating pressure. Thus, it was not surprising that the rejection for most of the filtration tests was not as good as expected while operating at a higher transmembrane pressure.

So, the model of droplet deformation in Figure 2 suggested that the size of drops could be shifted to a lesser degree if the interfacial tension could be increased, and that the filtration is carried out at a transmembrane pressure lower than the suggested critical value to prevent larger drops to penetrate through the pores throat.

The constants of filtration models are tabulated in Table 1. The cake filtration model represents a deposit increasing in mass and height on the surface of filter, while the standard filtration model illustrates a narrowing of the flow channel due to deposition of solids on the internal surface of the pore. The intermediate blocking

Table 1 Prediction of flux decay through filtration models

Filter tube configuration	Cake filtration			Standard filtration			Intermediate blocking			Complete blocking		
	α_1 (s/m^6)	β_1 (s/m^3)	r^2	α_2 (m^{-3})	β_2 (s/m^3)	r^2	α_3 (m^{-1})	β_3 (s/m)	r^2	α_4 (s^{-1})	β_4	r^2
Slots	8.0×10^6	7.9×10^4	0.723	122	7.9×10^4	0.728	1.38	900	0.728	2.1×10^{-3}	-0.0154	0.711
Slots with insert	1.1×10^7	5.2×10^4	0.605	164	5.2×10^4	0.611	1.86	592	0.611	3.8×10^{-3}	0.117	0.586
Circular pores	3.0×10^{10}	7.6×10^5	0.957	6853	1.3×10^6	0.991	95	16986	0.991	3.1×10^{-3}	0.143	0.938
Circular pores with insert	3.8×10^{10}	-3.4×10^5	0.841	7851	6.9×10^5	0.992	109	9597	0.992	3.7×10^{-3}	0.252	0.913
Ceramic membrane	3.0×10^8	5.6×10^5	0.923	365	5.7×10^5	0.909	18	28463	0.909	5.8×10^{-4}	0.049	0.878

model represents the internal pores of filter that are partially blocked due to the physical effects (such as transmembrane pressure, and drag and hydrodynamic forces), particulates deposition and wettability phenomenon.

Selection of the best model to adequately represent the filtration data and predict the flux decay was based on the correlation of the filtration data that linearly fit to the models. For slots, either with or without insert, there are no models that could sufficiently describe the filtration data, as suggested by the poor correlation of determination, r^2. This could be due to its high throughput as compared to the other two filters, as indicated by a lower α_1 in the model of cake filtration. Filtration by circular pores and ceramic membrane, on the other hand, could be well described by cake filtration. However, the inclusion of 17 mm helix inside the circular pores has changed the trend from the cake filtration model to the standard filtration and intermediate blocking models.

Table 2 shows the average values of membrane resistance and specific cake resistance of filters used in this study.

Table 2 Membrane resistance and specific cake resistance of filters

Filter	**Filtration area, A (m^2)**	**Membrane resistance, R_m (m^{-1})**	**Specific cake resistance, α (m/kg)**
Slots	0.0113	7.1×10^9	3.5×10^{10}
Slots with insert	0.0113	6.7×10^9	3.6×10^{10}
Circular pores	0.0139	4.4×10^{11}	8.8×10^{14}
Circular pores with insert	0.0139	-1.9×10^{11}	6.8×10^{14}
Ceramic membrane	0.0495	9.2×10^{11}	5.5×10^{13}
Flat sheet slots	0.0061	1.4×10^{10}	5.1×10^{11}

The membrane resistance of slots is about two orders of magnitude lower than that of circular pores and ceramic membrane. This signifies that the circular pores and ceramic membrane give a higher resistance to flow than the slots. Therefore, the throughput of slots would be much greater than the other two filters. The circular pores and ceramic membrane also give higher values of specific cake resistance than the slots. It infers that the slots configuration would be more likely to hinder from decays. As expected, the values of membrane resistance and specific cake resistance of flat sheet slots (dead-end configuration) are greater than the slots (crossflow), but they are much lower than those of circular pores and ceramic membrane. This proves the advantages of using slots configuration above the conventional circular pores in providing a higher throughput and a lower flux decay.

As shown in Table 2, the use of helical insert reduces the resistance to flow by decreasing the membrane resistance. This is true only for circular pores where the aforesaid value is negligible (negative). It is suggested that the helical insert configuration provides a fluid rotation and additional forces exerted on the filter so as sweeping the surface and preventing oil deposition on the pores throat. As for the slots, the similar values of membrane resistance even after the helix was inserted, suggested that the drops could be squeezed and so penetrate through the pores as a result of the hydrodynamic forces exerted by the helix. The use of helix, however, is insignificant towards the decrease of specific cake resistance for both slots and circular pores.

4.0 CONCLUSIONS

While filtering the oil in water emulsion using slots and circular pores, the drops can still pass through the filter even at low transmembrane pressure if the interfacial tension of an emulsion is low. The model of droplet deformation shows that a small increase in transmembrane pressure can allow passage of significantly larger drops. A suitable surfactant should then be used to increase the interfacial tension so that the oil drops could behave more like a solids particle for a better rejection. Generally, the imposed fluid rotation exerted by the helix could reduce the resistance to flow by sweeping the surface of filter so that no oil drops would deposit on the pores throat. The advantages of using slots above circular pores are high throughput, low flux decay and small specific cake resistance. However, the inclusion of helix into slots could give the unsuccessful rejection of oil.

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