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Effects of air sparging, cross flow velocity and pressure on permeation flux enhancement in industrial oily wastewater treatment using microfiltration

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ABSTRACT

Air sparging was used as a means to solve the problem of fouling and decline permeation flux in oily wastewater microfiltration (MF). The main objective of this research was to investigate the fouling reduction/removal in cross flow MF of industrial oily wastewater by using an air sparging. In this research, the outlet industrial oily wastewater from the API (American Petroleum Institute) separator unit of Tehran refinery was tested in cross flow with flat sheet MF membranes. The membrane module was operated vertically. Air and industrial oily wastewater were injected in co-current flow. Compared to without air sparging, the result shows that permeation flux increase of up to 170% in air sparging. Furthermore, the effects of various cross flow velocity (CFV) and transemembrane pressure (TMP) with air sparging flow rate have been investigated. Increasing CFV, TMP and air sparging flow rate increase the permeation flux. The best results were found in the air sparging flow rate of 40 ml/s, TMP of 3 bar and CFV of 1 m/s. In this condition air sparging showed greater efficiency in permeation flux enhancement. The result shows that these techniques could be a promising approach in order to overcome the problem.

Keywords: Industrial oily wastewater; Microfiltration; Membrane fouling; Cross flow filtration; Air sparging; Fouling control

1. Introduction

Cross flow membrane processes such as MF, ultrafiltration (UF) and etc. are effective, efficient and energy saving methods for separating oil and grease from oil in water and oily wastewater in many petrochemical, refineries, environmental and materials processes. Although this filtration mode has many advantages, the permeation flux decline due to the membrane fouling becomes a severe barrier for its further developments and wide applications [1–3].

Reduction of concentration polarization and membrane fouling has been the focus of many studies. Several methods have been developed in various applications of UF, and nanofiltration (NF) and reverse osmosis (RO): the use of higher CFVs, pulsed flow, use of modified

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membranes, use of corrugated membranes, production of centrifugal instabilities, feed pH optimization, feed oil concentration optimization, use of static turbulence promoters, operation under uniform TMP, use of optimum salt concentration, feed temperature optimization, helical baffles, screw treaded inserts, vibrating membranes and more recently proposed high shear rotary UF [4–18]. Some of these methods have been investigated for improving permeation flux in UF of synthetic oily wastewater. Um et al. showed that nitrogen injection causes positive effect on promoting turbulence leading to permeation flux enhancement [19]. Viadero et al. showed that high shear rotary UF allows concentration of oil beyond the typical operating limitations of conventional UF modules [18]. Faibish and Cohen reported an increase by over 20% in rejection for a synthetic oily wastewater with polymeric UF membrane [20]. Cui and Wright reported successful air sparging in enhancing UF permeation flux in membrane processes [21]. Mercier et al. reported a high amount of 200% permeation flux enhancement for suspensions by sparging air in UF inorganic membranes [22,23]. Shi et al. and Genkin et al. used vibration membrane in their experiments to enhance permeation flux [17,24]. Derradji et al. and Xu et al. installed a turbulence promoter before the membrane module [25,26]. Pospi'sil et al. also found that the existence of air sparged flow increases permeation flux [27]. Implementation of two phase flow (air/wastewater) was reported to remarkably improve performance of some membrane processes [28].

Most of previous studies used experimental test methods to understand permeation flux enhancement by increasing turbulency. Although the operating factors could be correlated into some empirical equations, most factors had to be determined by performing a series of experiments, and the results were related to real hydrodynamic conditions hardly [3].

Major research efforts in recent decades have been aimed to overcome the drawbacks of membrane fouling and permeation flux decline. A variety of operation techniques are available in MF process such as: changes in CFV, implantation of turbulence promoters, back-flushing or back-pulsing, pulsatile flow, rotation of flat sheet membranes, and applications of electrical and ultrasonic fields. Generally, these technologies and processes cannot completely surmount upon this problem. Also, they are often costly and ineffective. In industrial and commercial membrane applications, chemical cleaning is used periodically to restore permeation flux. However, to reduce the frequency of chemical cleaning and consumption of chemical cleaning agents, it is useful to apply enhancement techniques such as air sparging [1,2].

The main aim of these methods is to produce turbulency which can prevent the oil droplets/particles suspension to deposit on the membrane surface. According to this concept, increasing shear stress on the membrane surface by air sparging may reduce the oil droplets/particles suspension deposition and enhance permeation flux. In this study, effects of operating conditions such as air sparging flow rate, TMP and CFV on the permeation flux and membrane fouling are investigated. The main objective of this research is to evaluate the effect of air sparging on fouling decline/remove in an industrial oily wastewater MF process.

2. Materials and methods

Experiments were carried out using a flat sheet Polysulfone (PS, 0.1 μm) membrane from Alfa Laval Co. (Denmark). Effective area of the membrane in the module was 64 cm^2. Membrane properties are reported in Table 1. Fig. 1 shows the structure of the employed membrane. The scanning electron microscopy (SEM) (Philips model XL30) was employed to analyze the samples. The PS membrane is porous and asymmetric. It consists of a top or skin layer with a thickness of about 20 to 50 μm supported by a porous sub layer with a thickness of about 150 to 200 μm.

Industrial oily wastewater disposal of API separator unit of Tehran refinery wastewater treatment unit was employed as feed. The oily wastewater treatment was operated in cross-flow batch concentration process method. The schematic representation of MF set-up is shown in Fig. 2a. In this method the feed crosses the membrane module adjacent to the membrane surface and the permeate passes through the membrane module vertical to the membrane surface. In other words, the feed was pumped to the cross-flow membrane module from the tank and permeate flow was taken out of the loop and collected in an Erlenmeyer flask and measured by using a digital balance (sensitivity of 0.01 g), and the retentate flow was completely returned to the tank. Computer data acquisition software was used to record the measured data in filtration time. The membrane module was vertically, with stirrer for uniform oil/particle

Table 1
Characteristics of the polymeric membrane

	Membrane			Recommended operating limits		
Series	Name	Material	Pore sizes	pH range	Pressure range (bar)	Temperature range (°C)
MF-GRM0.1pp (PS)	PS	Polysulfone	0.1 μm	1.0–13.0	1–10	0–75

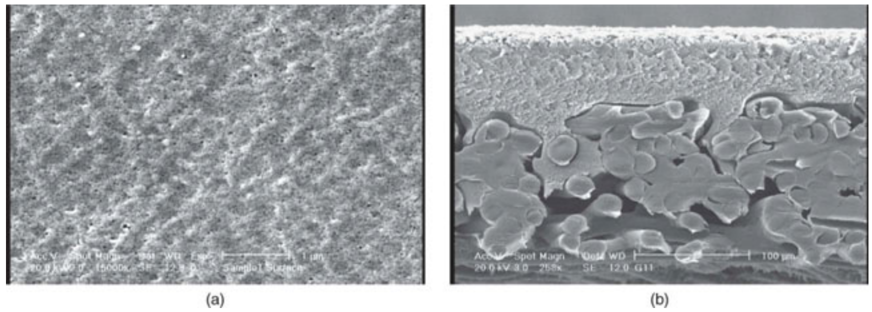

Fig. 1. (a) SEM of the PS membrane surface and (b) cross section.

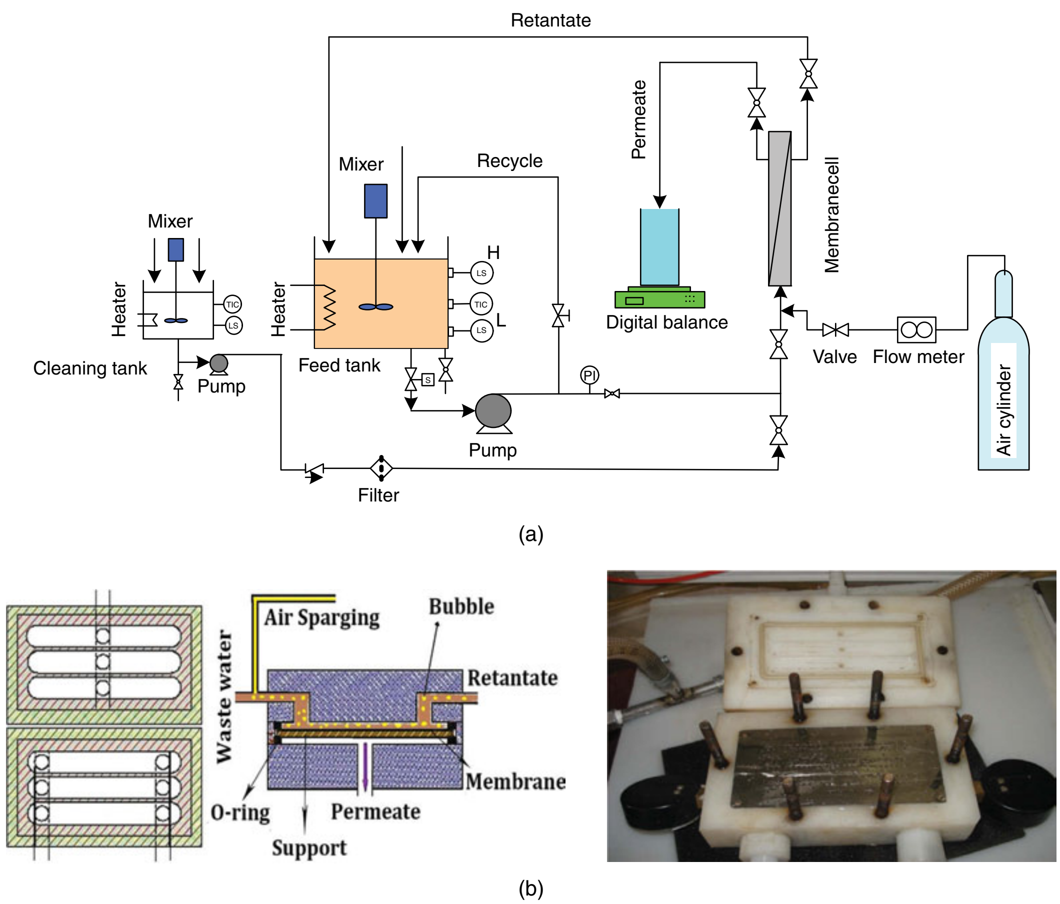

Fig. 2. (a) Schematic diagram of MF set-up (b) membrane module.

concentration along the system. The permeation flux through the membrane was obtained from the weight change of permeate in the Erlenmeyer flask divided by the membrane area and filtration time interval. The pressure drop between the feed and permeate side was measured by a pressure gage. The air used to generate bubbling was released from an air cylinder to the sparging header at the base end of the module, and the air flow rate was measured by a rotameter. This cycle has been repeated continuously. There was a by-pass before the feed inlet to recycle extra feed to the tank. There were two valves in the by-pass flow and retentate flow to adjust the main flow rate and desired operating TMP. The bypass flow had a significant influence on feed temperature. Because of the bypass flow, the pump heated the feed and it was needed to cool it to control the temperature, so the feed tank was equipped with cooling water coil and heat exchanger. A cross flow membrane module made from Teflon was used in the experiments (Fig. 2b). There were two ducts connected to two hoses. One of these hoses was the feed inlet and the other was retentate out let. The rectangular membrane was cut exactly to cover the whole area of the pool in one part of the membrane module. This part, had three holes at the bottom to conduct the permeate flow out of the membrane module. The membrane settled on a resistant compact foam layer, to protect it against deformation and displacement. Upper part and lower part of the membrane module were exactly symmetrical and have the same dimensions. An o-ring was placed between two parts of the membrane module. During the experiments, CFV, TMP, oil concentration, salt concentration, temperature and pH were carefully controlled. All of the adjustments and measurements for the MF experiments were the same.

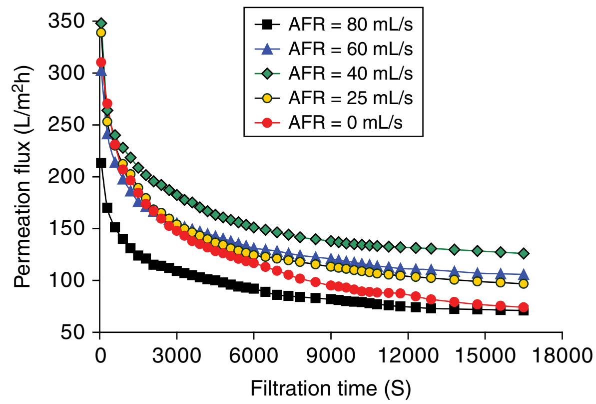

Fig. 3. Permeation flux of PS at different air flow rate (AFR) for industrial oily wastewater (TMP = 3 bar, flow rate = 1 m/s, temperature = 30°C).

3. Theoretical background

In this technique a gaseous flow is established parallel to the feed stream by injecting air or some inert gases like nitrogen at the inlet of the membrane module. Different configurations such as vertical upward flow, vertical downward, horizontal and inclined flows have been applied.

Introducing a ε value as [29]:

$$\varepsilon = U_g / \left(U_g + U_l\right)$$

where U_g and U_l are superficial air and liquid velocities respectively. While at ε values smaller than 0.25 a bubble flow regime prevails, when ε stays between 0.25 and 0.9, hydrodynamics of the system appear to be slug flow and at greater values the flow pattern would change to churn and finally annular flow.

4. Results and discussion

4.1. Effect of air flow rate

In order to investigate the effect of air sparging on permeation flux, the configuration with air sparging was tested with the permeate recirculation to the feed tank according to following procedure: a liquid (industrial oily wastewater) flow rate was set at the beginning of the experiment and the permeation flux was measured with the air injection. Without air sparging and the air flow rate was increased and the permeation flux was measured. Fig. 3 shows the time courses of permeation flux in cross flow MF under various air sparged flow rate intensities. Also, this figure shows the comparison of permeate flux on function of filtration time when operated in the presence and absence of air sparging. The Reynolds numbers of liquid (oily wastewater) and air flow rate are both higher than 20,000 within the operating conditions of this study; consequently, the flow pattern in both phases can be considered to be turbulence. At the beginning of filtration, the permeation fluxes under different air-bubble velocities are almost all the same since no cake has been formed yet. The fouling resistance is only attributed to the filter membrane at that time. After a prolonged operation, the permeate flux of both operating condition with and non air sparging slightly decreased and reached a constant value. The curves shown in the figure clearly indicate that the permeation flux decline would be restrained by the air sparged. The more intense the air sparged flow rate is, the higher the permeation flux will be obtained. This result reveals the permeation flux can be efficiently enhanced by an air sparged. However, the final permeation flux when using air sparging was found to be 1.704 times higher than that of without air sparging. The final permeation flux operated with air sparging was about 126 l/m^2h, whereas the latter was about 74 l/m^2h. During the first 10 min of filtration, permeation flux decreases rapidly in spite of a regular air sparging. It could probably capture some of small oil droplets within its structure leading to the rapid decline in the permeation flux.

More air sparged flow rate may result in a higher permeation flux due to the greater shear stress acting on the membrane surface. However, the degree of permeation flux enhancement at the steady state becomes maximum as air sparged flow rate increases. Further increase of the air flow rate even lead to the increase of the permeation flux: at air flow rate of 40 ml/s), which corresponds to the slug flow pattern (ε = 0.415), the final permeation flux was around 40% higher than the final permeation flux obtained air sparging (flow rate of 25 ml/s). The final permeation flux can be increased 70% when the air flow rate is from 0 to 40 ml/s.

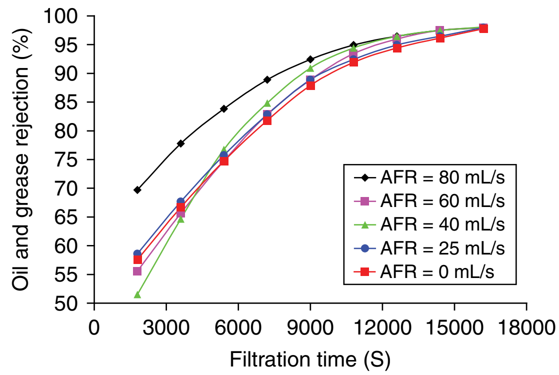

Fig. 4. Oil and grease rejection of PS at different air flow rate (AFR) for industrial oily wastewater (TMP = 3 bar, CFV = 1 m/s, T = 30°C).

The effect of filtration time on oil and grease rejection of the PS membrane under the different air flow rate operational conditions is presented in Fig. 4. Samples for measurements of the feed and the permeate oil and grease content was taken as necessary and analyzed by the procedure outlined in standard method (APHA, 2001). Oil and grease rejections of the all runs vary similarly with filtration time, at various air flow rate increases a little more sharply and this can also be attributed to the same morphologies and material of the membrane at all runs.

The permeation flux increased with air flow rate ranged of 0–80 ml/s. But when the air flow rate exceeded 40 ml/s, the trend reversed. As shown in Fig. 5, while air flow rate reached 80 ml/s, the permeation flux was even lower than that of 40 ml/s. It is due to high volume fraction air in fluid and also had a negative effect of decreasing the effective membrane area due to partial occupation of membrane pores by bubbles. Effect of air sparging on decreasing membrane fouling and increasing permeation flux can be stated in two reasons: (i) two phase flow is resulted from air sparging which increases shear stress (force) and elimination of fouling from membrane (Fig. 6). (ii) Air sparging increases turbulence and permeability of membrane wall which decreases concentration polarization on membrane surface and it prevent oil droplets and solid particles sedimentation. Although, this process consumes air and increases operating cost but it also increases membrane efficiency and life time.

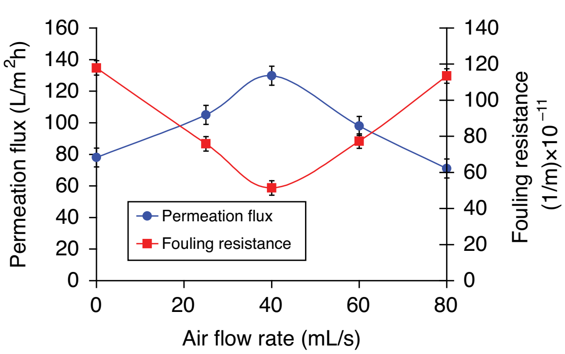

Fig. 5. Effect of air flow rate on permeation flux and fouling resistance.

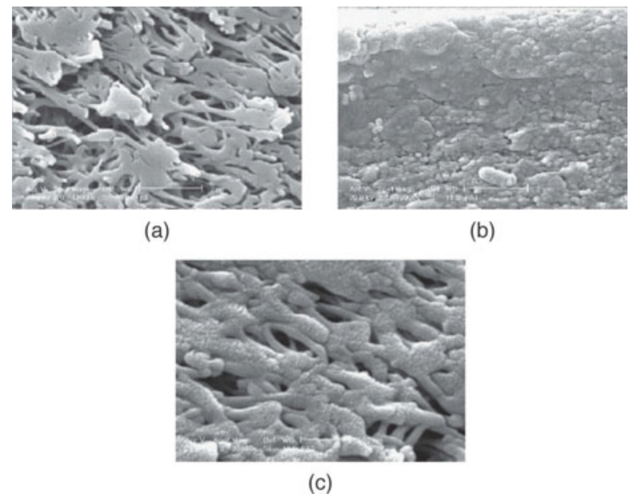

Fig. 6. Cross sectional SEM of the membranes (a) before filtration, (b) after filtration without air sparging and (c) after filtration with air sparging.

Fig. 6 shows SEM cross section images of membrane before filtration (Fig. 6a), after filtration without air sparging (Fig. 6b) and after filtration with air sparging (Fig. 6c). Apparently, the membrane surface has been fully fouled with oil particles and other impurities when without air sparged into the membrane module, however, less fouling is observed when air is sparged into the membrane module. As a matter of interest, less fouling in the presence of air sparging pertains to the more turbulence of cross flow close to the membrane surface which avoids precipitating the oils and other impurities.

These results are in agreement with the results obtained during air sparging in membrane processes involving oil-in-water emulsions. Um et al. investigated the effect of nitrogen injection during UF of a 5% wt. synthetic oily wastewater [19]. Permeation flux improvements of up to 200% were achieved under operation conditions which correspond to a bubble flow. On the other hand, Ducom et al. observed the decrease of permeation flux at low air velocities, with permeation flux enhancements when the air velocity was increased during NF of a 10% vol. synthetic oily wastewater [30].

The permeation flux increase at high air flow rates was explained with increased turbulence of the fluid (industrial oily wastewater) by air sparging. Due to increased turbulence, oil droplets come back bulk fluid which can lead to decreased membrane fouling resistance and increase of permeation flux. By injecting air, the viscosity of an air/liquid (air/wastewater) mixture decreased and so the turbulence in the module was increased, which could lead to an increase of permeation flux. From the results presented in Fig. 3, it is obvious that air sparging does to be an effective technique for permeation flux enhancement in MF of industrial oily wastewater.

4.2. Effect of TMP on the permeation flux at different air flow rate

According to Darcy's law, increasing TMP increases permeation flux, however, fouling restricts this fundamental law. Increasing TMP makes the oil droplets/particles suspension more compact on the membrane surface, and as a result, they block the membrane pores [31–33]. Thus, at an optimum TMP, permeation is high, while tendency to cake/gel layer formation is low. Effects of TMP on permeation of the MF membranes during treatment of industrial oily wastewaters with air and without air sparging are presented in Fig. 7. The results indicated that permeation flux increases with increasing TMP. Fig. 7 shows the cake/gel layer formed on the membrane surface under various air flow rate and TMP. Air sparged can significantly reduce the cake formation on the membrane surface, i.e., the cake/gel layer decreases with the increase of air flow rate. This indicates that air sparging is an efficient way in reducing cake/gel layer formation. This phenomenon can reasonably explain why the permeation flux can be enhanced by air sparged. On the other hand, the cake/gel layer also increases with increasing TMP. This is because the oil droplets staying on the membrane are more stable, or in other words, are more easily to deposit, under a high TMP. Although the fouling resistance may be increased due to the increase of cake/gel layer, the higher TMP is still enough to drive a higher permeation flux.

An increase in TMP leads to a higher permeation flux due to the higher filtration driving force. A 500% permeation flux increase can be obtained as the TMP increases from 0.5 to 4.5 bars. It can be noticed that the permeation flux does not linearly increase with TMP since the permeation fluxes are all higher than the without air sparging.

On the other hand, air sparging technique can be used for maintaining a given permeation flux with a relatively low TMP drop across the membrane, and therefore low energy consumption. Air sparging could be an interesting solution for improving performance. If air sparging could reduce a high TMP drop across the module could still provide considerable permeation flux enhancement, the performance of the process would be significantly improved.

The result shows that, TMP of 3 bars can be considered as the best operating TMP because at higher TMP, the cake/gel layer becomes denser and permeation do not increase any more.

4.3. Effect of CFV on the permeation flux at different air flow rate

Increasing CFV increases mass transfer coefficient in the concentration boundary layer and also increases the extent of mixing over the membrane surface [34–36]. In Fig. 8, effects of CFV at different air flow rate on permeation flux of the MF membranes are presented. Also, the influence of different air flow rate on permeation flux was compared. It can be observed that permeation flux increases with increasing CFV. At air flow rate equals to 0 ml/s, there is low turbulency so the cake/gel layer can be formed easily. Therefore, maximum fouling and minimum permeation flux is observed.

The permeation flux increased with CFV ranged of 0–2 m/s. These phenomena were on account of two competing effects on the formation of the cake/gel layer.

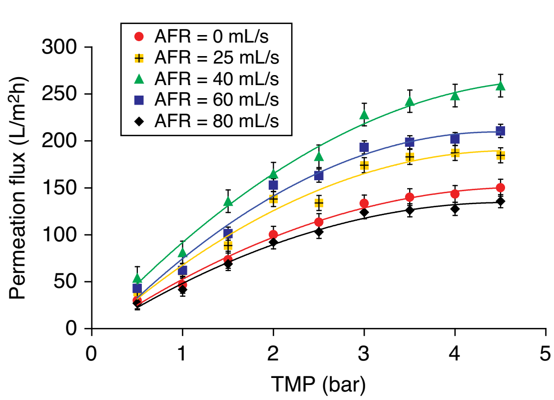

Fig. 7. Effect of TMP on permeation flux at different air flow rate (AFR) (CFV = 1 m/s, T = 30°C).

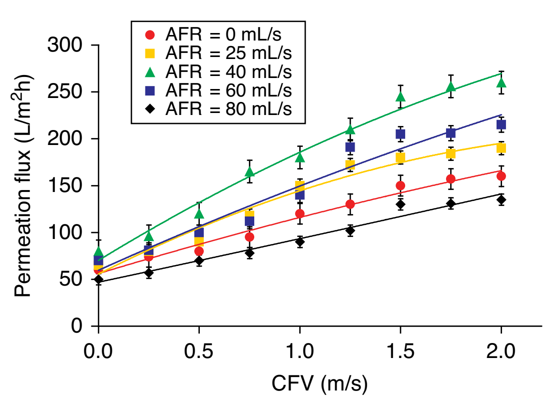

Fig. 8. Effect of CFV on permeation flux at different air flow rate (AFR) (TMP = 3 bar, T = 30°C).

One effect is that the cake/gel thickness is mainly controlled by CFV so that higher CFV leads to form thinner cake/gel layer and produce higher permeation flux. The other is that bigger air bubble are easier to be taken away by the speeding fluid while smaller grains tend to stay on the membrane surface, which makes the, increasing turbulence, fouling resistance lower and increases permeation flux. In particular air sparging with CFV could increase velocity and turbulence near the membrane surface, thus limiting the boundary layer and/or thickness and decline of concentration polarization. There is another hypothesis that even the cake/gel layer porosity may change due to preferential separation of large particles in flow stream and development of a denser cake/gel layer [31,32].

5. Conclusions

Membrane fouling is a major obstacle in its industrial application in many filtration processes. Air sparging was shown to be effective in solving the fouling problem. In this condition, air sparging could be a proper choice to enhance the permeation flux.

The effects of operating conditions, such as air sparging flow rate, CFV and TMP, on the fouling resistance and permeation flux in air sparged cross flow MF have been investigated. The final permeation flux increased with increasing air sparging flow rate, CFV and TMP. The air sparged could significantly enhance permeation flux, e.g., the permeation flux increased 170% when the air sparged flow rate increased from 0 to 40 ml/s. The best results are found in the slug flow regime ($\varepsilon = 0.415$). In conclusion, increasing the shear stress and generating turbulence with increasing air flow rate were efficient ways to enhance permeation flux in industrial oily wastewater treatment. The results show that, air sparging in to the feed stream increases turbulence near the membrane surface as well as the CFV, thus limiting the cake/gel layer thickness.

Briefly, the result shows that the use of the air sparging as a turbulence promoter during cross-flow MF of an industrial oily wastewater can provide permeation flux values up to 1.7 times higher compared to the permeation fluxes obtained during operation without using the air sparging. Air sparging method is indeed worth for further development due to its low operating cost, effective, efficient and easy operating conditions.

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