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Increasing rejection in microfiltration using vibrating membrane

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ABSTRACT: A new method is proposed to increase rejection in microfiltration by applying membrane oscillation using a new type of microfiltration membranes with slotted pores. The oscillations applied to the membrane surface result in reducing membrane fouling and increasing separation efficiency. An exact mathematical solution of the flow in the surrounding solution outside the oscillating membrane is developed. The oscillation results in appearance of the lift velocity, which moves oil particles away from the membrane. The latter results in both reducing membrane fouling and increasing oil droplets rejection. This developed model was supported by the experimental results for oil water separation in produced water treatment. It was proven that oil droplet concentration reduced notably in the permeate due to the membrane oscillation and that applied shear rate caused by the membrane oscillation is also reduce pore blockage. New generation of microfiltration membranes with slotted pores was used in the experiments.

Keywords: Membrane oscillation; shear rate; slotted structure membrane; oil water separation and membrane fouling

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INTRODUCTION

Sea water is substantially polluted by the discharge of oily produced water, which is an essential environmental concern [1-4]. On industrial scale, oily produced water is generated in huge amount with prediction of approximately 88 billion barrels on the yearly basis worldwide [5]. There are various separation methods used for purification of the oily produced water which include pH change, gravitational method, centrifugation, biological treatment, membrane filtration and electrostatic de-emulsification [1, 6-12]. Membrane purification process is superior over other processes because it has several benefits such as it require no chemicals, low input energy, environment friendliness, high quality of permeate [11,13–16]. It is the reason why membrane separation processes got attention of researchers over recent years for treatment of produced water [17,18]. Despite of wide application of membrane purification, fouling is the major problem in membrane separation of oil from water emulsion that leads towards a considerable reduction in the permeate flux [11,19].

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Various methods of membrane separation have been tried for the separation of oil droplets and water. The method of microfiltration (MF) [20-26] and ultrafiltration (UF) [27-32] are considered superior as compared to other membrane separation methods for separation of oil from water. The reason behind that is fouling in these membranes is less than that of reverse osmosis and nano-filtration membranes for oil water separation. It has been found that MF membrane gives greater permeate flux at lower trans membrane pressure as compared to UF process.MF is more economical for oil water separation on commercial scale [33–35].Various researchers have also studied the impact of membrane pore geometry on separation efficiency [36–39]. Recently slotted pore membranes were introduced for oil water emulsion purification [24,35,40–44]. It was found that slotted pore membrane gives greater value of permeate flux at less trans-membrane pressure compared to circular pore membrane [35].

MF is widely used, however, the membrane fouling still remains the important issue to be resolved [44]. Various methods have been suggested for the reduction of the membrane fouling of increasing shear such as higher cross flow filtration and aeration. In the aeration process, bubbles are generated which results in disturbing the concentration polarization and, hence, reducing the fouling [45]. However, this process has disadvantage of higher energy consumption. In the case of cross-flow filtration, feed supplied with high tangential velocity, which results in shear rate generation over the membrane surface [46], but this approach has also limitation of high power consumption and requires multiple recirculation of feed stream [46,47]. To overcome this problem of higher energy consumption in cross flow filtration, another method referred to as dynamic microfiltration was proposed. In dynamic microfiltration, relative motion between the bulk fluid and membrane used is applied, which results in higher shear rate applied to the membrane surface. This is achieved by vibration or rotation of the membranes. Application of membrane oscillation, fouling and concentration polarization can be reduced [48-58] and just this method is considered below. It is shown below that the membrane oscillation results in a substantial decrease of the trans-membrane pressure, results in a higher rejection of oil droplets and decreases substantionly membrane fouling. It is shown below that membrane oscillation method provides lower oil concentrations in the permeate, that is, higher rejection as compared with other memthods.

Mathematical model is developed below and exact solution of the flow in a vicinity of the oscillating membrane is deduced. The deduced mathematical solution allows calculating the shear applied on the membrane surface. It is shown that the imposed oscillations allow reduce membrane fouling and increase rejection of oil droplets, which is caused by lift velocity. It was shown that because of the oscillation, which are applied over the membrane's surface, lift velocities are developed, which move the droplets from the membrane's surface. The effect of membrane pore blocking was studied at various shear rate . Experimental investigation of the oscillating membrane fouling is undertaken, which is an agreement with the theoretical prediction. The prediction of permeate concentration of oil droplets was made.

A new type of membrane with slotted pores was used for investigation of oscillation influence, which were not used for this purpose before.

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MATHEMATICAL MODEL

In Figure 1 a schematic diagram of the membrane oscillating along z-axis is presented. The velocity distribution was generated because of oscillating membrane, which results in the shear rate creation. The oscillation results in a lift velocity of the oil droplets, which moves away the fouling materials from surface of membrane and reduce the concentration polarization in the vicinity of the membrane. As a result, both fouling and concentration polarization are reduced. The model is developed in this section allows calculating the shear rate over the membrane's surface caused by membrane oscillations. The dimension of the membrane module is supposed much bigger as compared with the dimension, **δ**, of the region caused by the oscillations (Figure 1), hat is, the membrane can be assumed to be flat. The oscillating membrane is completely immersed in the liquid. It is assumed also that the Reynolds number is small. In this case Navier-Stokes equations are reduced to only one equation:

$$\frac{\partial V_z}{\partial t} = \nu\left(\frac{\partial^2 V_z}{\partial y^2}\right), \qquad (1)$$

where, V_z is z component of the fluid velocity, which depends on the y only (Figure 1), t and y are time and co-ordinate (Figure 1), ν is a kinematic liquid viscosity. The displacement of membrane which is oscillating along z-axis is as follows (Figure 1):

$$Z(t) = A\sin(\omega t),$$

where A is an amplitude of oscillations and ω is the frequency of oscillations.

The following boundary conditions should be satisfied for the liquid velocity, determined by Eq. (1):

$$V_z = A\omega\cos(\omega t),\ y=0, \qquad (2)$$

$$\text{At } y \to \infty \qquad V_z \to 0,\ y \to \infty. \qquad (3)$$

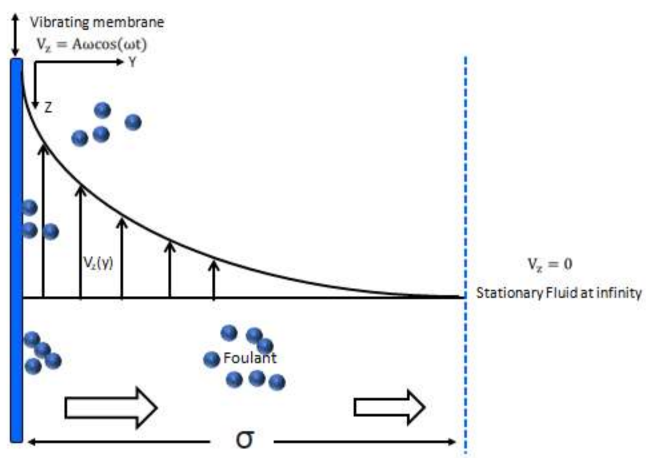

Figure 1. Velocity distribution of fluid & motion of foulants in the vicinity of submerged oscillating membrane.

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Exact solution of Eq. (1) satisfying boundary conditions (2)-(3) is as follows:

$$V_z = A\omega \exp(-\alpha y)\cos(\alpha y - \omega t)\,, \qquad (4)$$

where $\alpha = \sqrt{\omega/2\nu}$. See Appendix for details of the derivation.

According to Eq. (4) the influence of oscillations is extended into the bulk of the liquid on the distance $\delta \sim \frac{1}{\alpha} = \sqrt{2\nu/\omega}$, that is, decreases with increasing frequency of oscillations.

We define the shear rate as $\gamma(t,y) \;\; = \frac{dV_z(t,y)}{dy}$. Using Eq. (4) we conclude:

$$\gamma(t,y) \;\; = \frac{dV_z(t,y)}{dy} = -A\omega\alpha \exp(-\alpha y)[\cos(\alpha y - \omega t) + \sin(\alpha y - \omega t)] \qquad (5)$$

The shear rate over the surface of the oscillating membrane can be obtained by finding the value for boundary at y=0. This gives the shear rate over the surface of the oscillating membrane:

$$\gamma_s = A\omega\alpha \quad [\sin(\omega t) - \cos(\omega t)] \qquad (6)$$

The shear applied on the membrane surface is proportional to the frequency of oscillations according to Eq. (6). This shear rate prevents pore blocking. The latter has a direct experimental confirmation (see Figure 7), which proves that pore blocking is reduced linearly with oscillation frequency in accordance with Eq. (6). In the absence of the oscillations, the only force acting on the oil droplet is the drag force, which pushes the droplet through the membrane into the permeate. However, when the membrane is oscillating this results in generation of the shear of various intensities and the lift force, which moves the oil droplet away from the membrane surface. Figure 2 shows the act of forces (drag and lift forces) in the case of membrane oscillation. Lift force is the consequence of the applied oscillations of the membrane surface. The intensity of the lift force is higher at the membrane surface and gradually decreases when moving away from the membrane surface.

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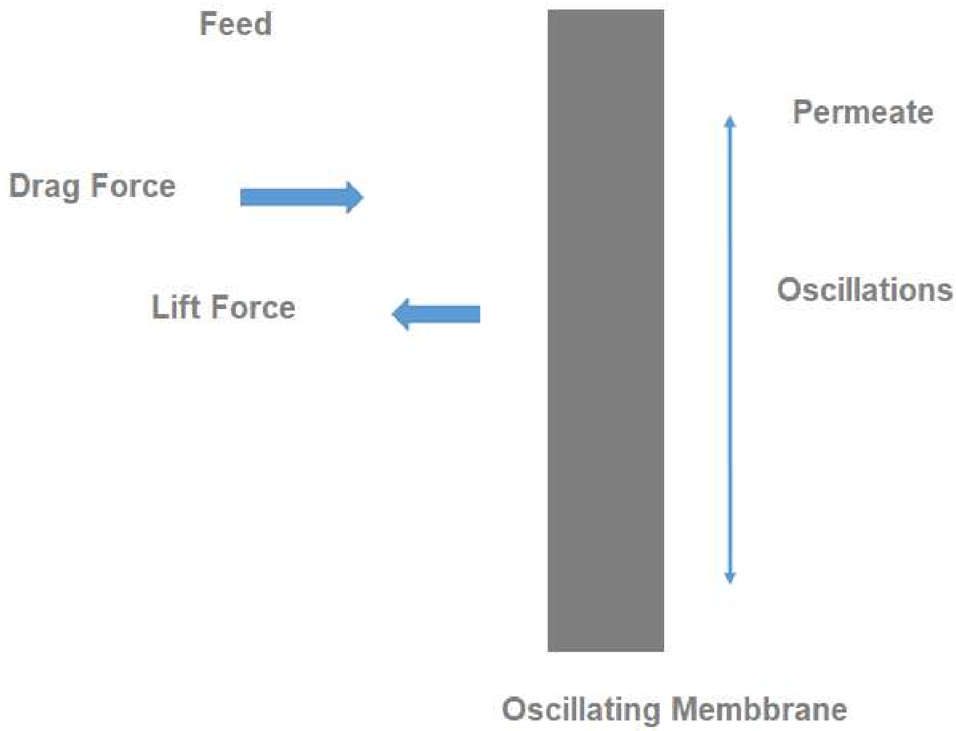

Figure 2. Drag force tries to push the drops to the permeate side, while the lift force comes from the membrane oscillation acts in the opposite direction to the drag force.

Expressions for the drag force and lift force are given below:

$$F_d = k_w 12\pi\eta R_{sp} U \quad (7)$$

where F_d is the drag force, k_w is a wall correction factor, for a similar system k_w value 4.3 was used [24]; η is the dynamic viscosity of the liquid, R_{sp} is the radius of the droplet and U is the permeate velocity of the liquid, caused by applied cross-membrane pressure difference [24].

The lift force is given by the following expression [46]:

$$F_l = 81.2\left(\left(\rho_w \eta |\gamma|^3\right)^{0.5}\right) R_{sp}{}^3, \quad (8)$$

where F_l is the lift force, ρ_w is the density of the water at room temperature and γ, according to Eq. (6), is the applied shear rate [46]. Eq. 8 shows the lift force is determined by the applied shear rate and drop size. It means that for a given shear rate, the shear force will be more effective for larger droplets [46]. Note, according to Eqs. (6) and (8) the lift force decreases exponentially away from the oscillation membrane surface.

The shear rate is produced over the surface of membrane because oscillation creates the lift force which tends to move the droplets from the surface of membrane. As a result, number oil drops which could be deposited

 doi:10.20944/preprints202108.0393.v1

on the membrane surface and block membrane pores is reduced. So, fouling also gets reduced. The model is validated with the experimental results in the following section. It is shown below the less concentration of oil droplets in permeate stream, which is due to the shear rate applied over the membrane surface.

(9)

According to Figure 2 the drag velocity of oil droplets towards the oscillating membrane is reduced (or even becomes negative, that is, away from the oscillating membrane), which is caused by the applied membrane oscillations.

EXPERIMENTAL METHODOLOGY

OSCILATING MEMBRANE FILTRATION

The food blender at high speed was used for the oil droplets formation from vegetable oil. The real produced water was also used in the experiments. The Coulter Mullisizer II was used for the examination of oil droplets size distribution in both cases. Size scale of oil droplets in both cases was determined in the range of 1 µm to 15 µm. The slotted porous membrane of 4 µm pore size in width was utilized for the process of oil and water separation (Figure 3). This membrane was made up of Nickel (Ni) whose surface was modified with Poly tetra fluoro ethylene (PTFE) by Micropore Technologies, UK. The picture of membrane was captured by scanning electron microscopy shown in Figure 3.

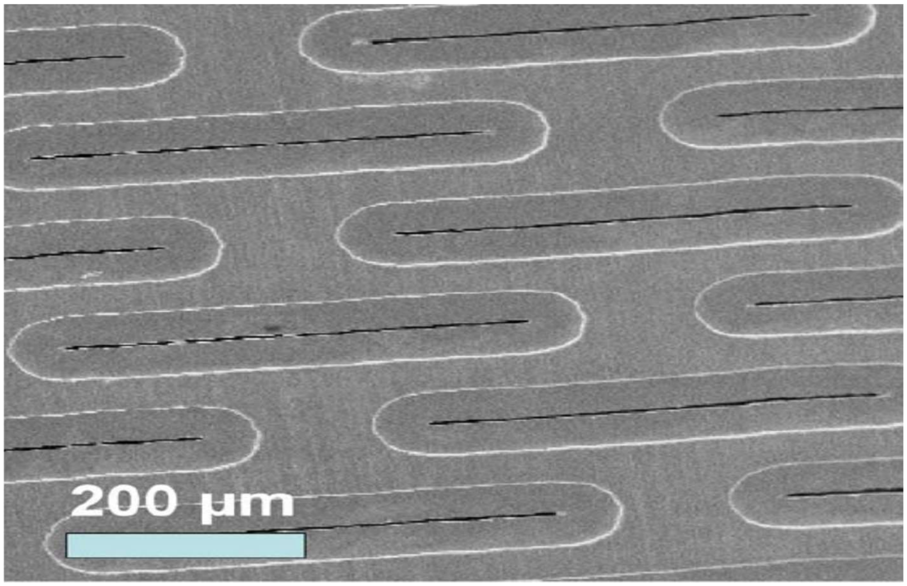

Figure 3. Scanning Electron Microscopy (SEM) Diagram for Slotted Porous Structure Membrane.

The membrane was connected with oscillating arm, then oscillating arm is initiated with electrochemical oscillator in order to create shear rate over the membrane surface. The flow diagram of oscillating microfiltration membrane with oscillating system for separating oil and water is shown in Figure 4. The oscillating frequency and amplitude scale for membranes was

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varied in between 0 to 10 Hz and 0 to 10 mm, respectively. As a result, the shear rate was produced in the vicinity of the membrane outer surface. The constant flux through the membrane was created using positive displacement (PD) pump.

Filtration experiments with the oscillating sotted pore membrane at various intensities were applied to both crude oil and vegetable oil emulsions. The membrane oscillation in vertical direction results in the shear rate creation.

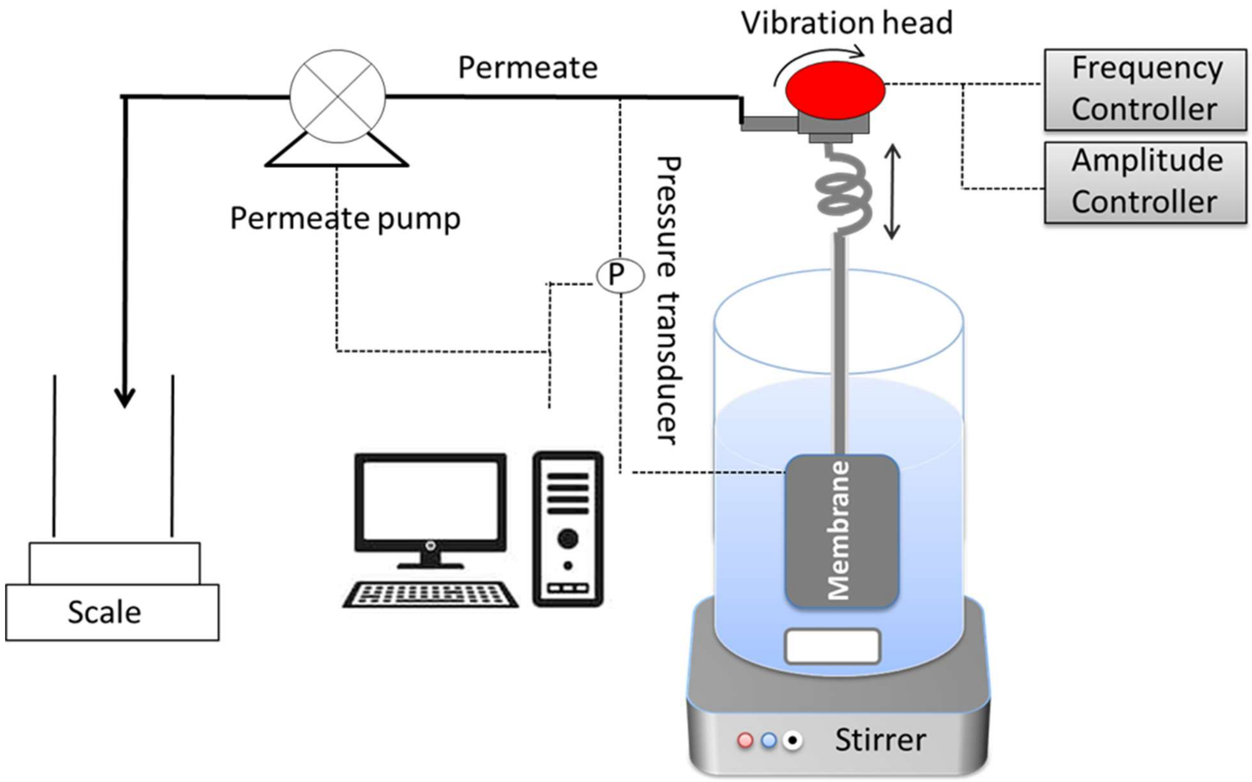

Figure 4. Flow diagram of the membrane oscillation devise.

MATERIALS USED

The drops of crude oil and vegetable oil (stabilized by Tween 20) were used. In both cases oil droplets showed the interfacial tension around 30 and 4 mn/m, accordingly. The length and width values of slotted pores on the membrane surface were 400 μm and 4 μm, respectively. The total area of the slotted pre membrane was $1.6\text{x}10^{-9}$ m^2.

RESULTS AND DISCUSSION

The size distribution is established based on oil droplets mass in the permeate stream shown in Figures 5 and 6. Figures 5 and 6 show that there is a considerable reduction in the concentration of oil droplets in permeate caused by membrane oscillations are applied. The reduction in concentration of permeate was found to be a linear function of applied shear rate. The interfacial tension has played as an important part to reduce the oil droplets concentration. At low value of interfacial tension, flattening and deformation

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of oil droplets was easier, which tends to move droplets into the permeate. While at high value of interfacial tension, the droplets became stiffer, their deformation and flattening was not so easy and this results in a lower droplet penetration into the permeate.

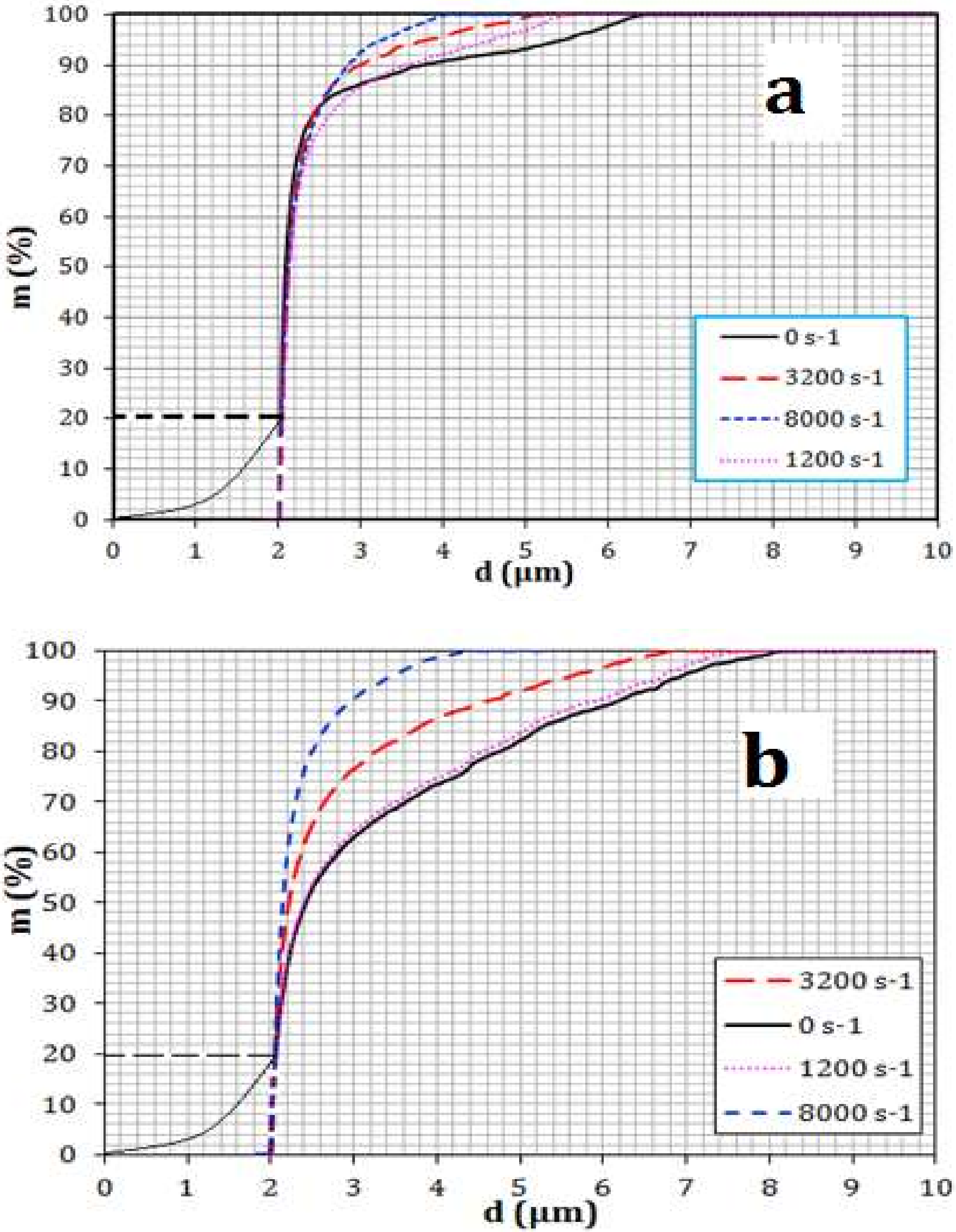

Figure 5. Size Distribution of mass of oil droplets stabilized by Tween 20 in the permeate at (a) at 200 l/m²h.& (b) 1,000 l/m²h. [44].

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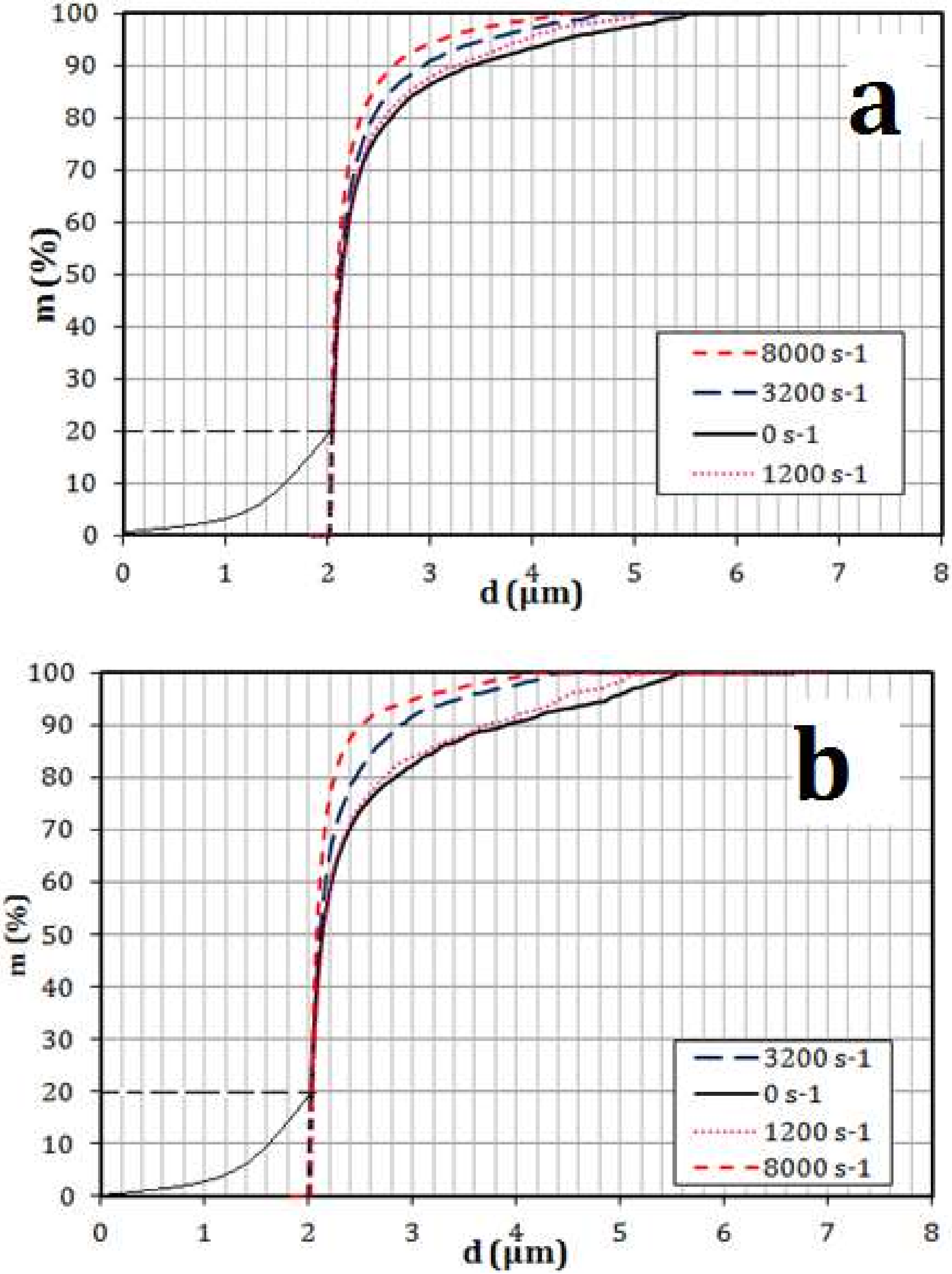

Figure 6. Size distribution of mass of crude oil droplets in the permeate at (a) 200 lm^2/hr (b) 1000 l/m^2h [44].

The blocking of pores (slots) of membrane was investigated experimentally by applying oscillation shear rate over the membrane surface. In these experiments, droplets of crude oil and vegetable oil (stabilized by Tween 20) were used. Like concentration in permeate, it was found that blockage of membrane pores were also reduced by applied oscillating

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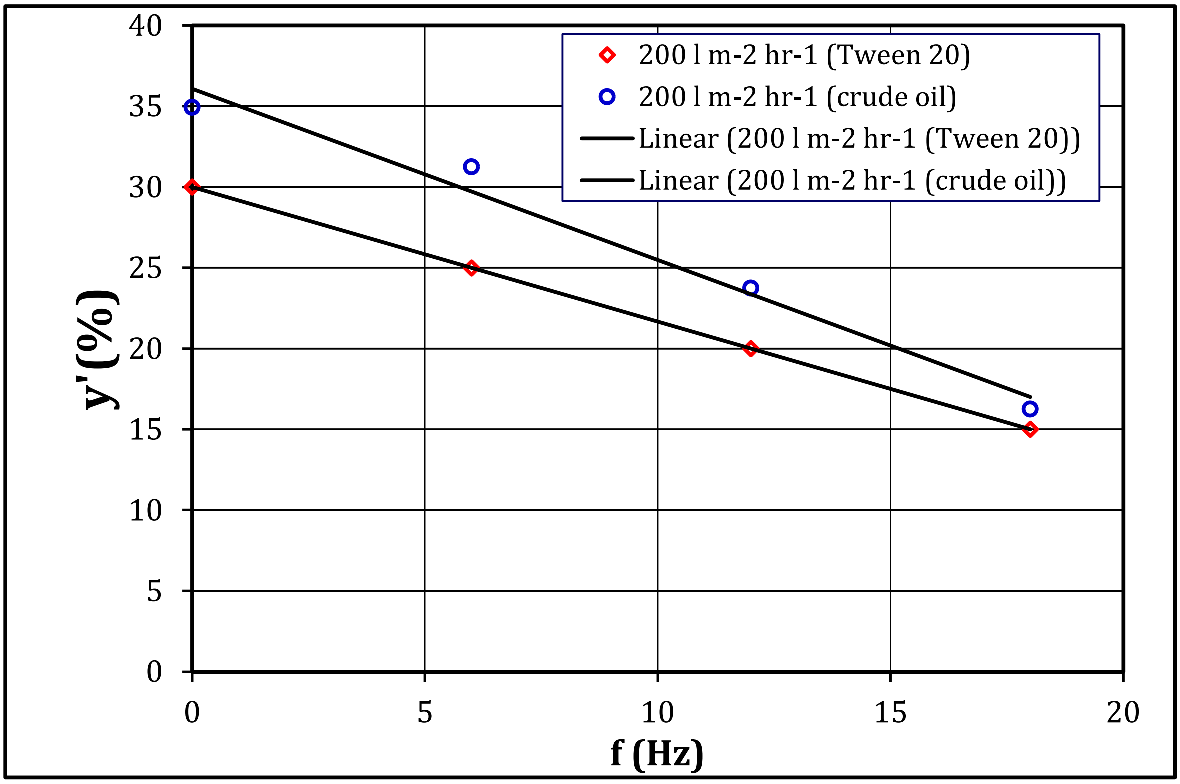
membrane. It was found that the blockage of pores was appreciably reduced by oscillations applied as shown in Figure 7. This reduction in blockage was found to be a linear function of oscillating frequency of membrane. It means the greater value of oscillating frequency of the membrane, the lower blockage of pores is. It was shown that the interfacial tension was essential parameter in pores blockage of membrane. Oil droplets having low interfacial tension can be deformed easily and tends to move into the permeate, which results in less pore blockage. But on the other side, at high interfacial tension, blockage in pores of membrane was found higher. This made the drops stiffer and it became more difficult to push the undeformed drops through pores, which results in more pore blockage [44].



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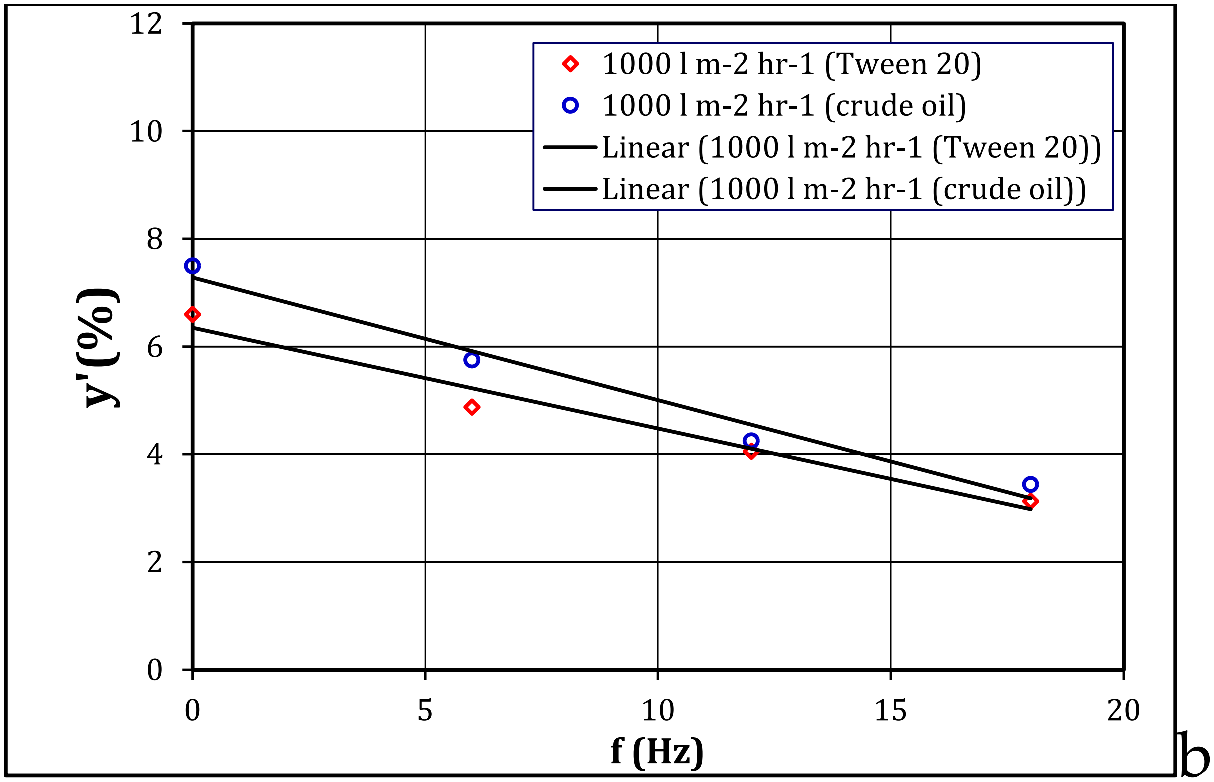

Figure 7. Impact of oscillation frequency, f(HZ), on the pore blockage area, y', in percentage. Stable vegetable oil droplets (stabilized by Tween 20) & crude oil droplets at (a) 200 l/m^{2}h (b) 1,000 l/m^{2}h.

Table 1. Number of crude oil drops (30 ° American Petroleum Institute) and concentration of oil in the feed and permeate obtained at various frequencies of oscillation and 400 l m^{-2} hr^{-1} flux rate.

Frequency (Hz)	No of drops per 0.4 ml sample	Concentration of crude oil in the feed (ppm)	Concentration of crude oil in the permeate (ppm)
25	1020	400	5
21	1606	400	7
0	5092	400	21

Table 1 shows number of crude oil drops in the permeate and permeate concentration obtained at different shear rates. It is clear that both number of drops in the permeate and crude oil concentration in the permeate were influenced by the applied shear. No of drops and crude oil concentration reduced with the applied shear rate.

CONCLUSIONS

An analytical solution has been developed which allows to calculate the velocity and shear rate distribution in the vicinity of the oscillating membrane and on the membrane surface itself. It is shown that the oscillations result in creation of a lift force, which acts in opposite direction as compared with drag force. This results in lower number of oil droplets reaching the oscillating membrane, and hence, less oil droplet penetrating into the permeate (higher

 doi:10.20944/preprints202108.0393.v1

rejections) and lower membrane fouling. The analytical study was validated against the experimental results, which proves that oscillation of membrane reduced the concentration of the oil droplets in permeate and reduce the membrane fouling. This reduction in the oil droplets concentration in the permeate was found to be a linear function of oscillating frequency of the membrane. The oscillations reduce the blockage of pores, the greater the shear rate intensity, lesser pore blockage was noticed.

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NOMENCLATURE:

A	Amplitude of oscillation (m)
Z(y,t)	Instantaneous displacement of an immersed oscillating membrane along z-axis (m)
t	Time (s)
V_z	Velocity of the fluid in z direction (m/s)
y	Lateral coordinates (m)
z	Directional Coordinate (m)

Greek Symbols

γ	Shear rate (1/s)
γ_s	Shear rate at membrane surface (1/s)
μ	Fluid dynamic viscosity (kg/ms)
υ	Fluid kinematic viscosity (m^2/s)
ϱ	Fluid density (kg/m^3)
ω	Angular Frequency (rad/s)
i	$\sqrt{-1}$
$\Re e$	Real part

7. LIST OF ABBREVIATIONS

MF	Microfiltration
NF	Nano Filtration
RD	Rotating Disk system
RO	Reverse Osmosis
RMF	Rotating Membrane Filter system
TMP	Trans Membrane Pressure
UF	Ultrafiltration
VHFM	Vibrating Hollow Fiber Module System
VSEP	Vibratory Shear-Enhanced Processing System

Appendix

Solution of the Eq. (1) is obtained using (a) a method of separation variables and (b) solution is tried in complex variable form, but in the end the real part of the deduced solution satisfies the initial equation and boundary conditions (2) and (3).

Solution of Eq. (1) is presented in the following form

$$V_z = \Re e f_1(t) f_2(y) \qquad \text{(A.1)}$$

Substitution into Eq. (1) and separating variable results in:

$$\frac{f_1'(t)}{f_1(t)} = \nu f_2''(y)/f_2(y)$$

The left- hand site of the latter equation depends on time only while the right-hand site on y only. The latter is possible only if both sides are constants. Having in mind that both sides should be oscillatory functions of ω, this constant is equal to iω, where i is the imaginary unit. The latter results in the following two equations

$$\frac{f_1'(t)}{f_1(t)} = i\omega$$

$$\frac{\nu f_2''(y)}{f_2(y)} = i\omega$$

Solution of these equations is as follows

$$f_1(t) = e^{i\omega} = cos\omega t + i\ sin\omega t \qquad \text{(A2)}$$

$$f_2(y) = A_1 e^{(\alpha+i\alpha)y} + A_2 e^{(-\alpha-i\alpha)y} = A_1 e^{\alpha y}(\cos\alpha y + i\sin\alpha y) + A_2 e^{-\alpha y}(\cos\alpha y - i\sin\alpha y) \qquad \text{(A3)}$$

where A_1 and A_2 are integration constants. From the condition (3) we conclude that $A_1=0$, otherwise the solution does not tend to zero at y tents to infinity. From the condition (2) we conclude that $A_2 = A\omega$. Hence,

$$f_2(y) = A\omega e^{-\alpha y}(\cos\alpha y - \mathrm{i}\sin\alpha y) \qquad \text{(A4)}$$

Substitution of expressions (A2) and (A4) into Eq.(A1) results in solution given by Eq. (4).

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