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Ultrasound irradiation combined with hydraulic cleaning on fouled polyethersulfone and polyvinylidene fluoride membranes

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In this study, an ultrasonic irradiation technique was utilized to mitigate the fouling of polyethersulfone (PES) and polyvinylidene fluoride (PVDF) membranes. The use of ultrasound at 20 kHz was applied to a dead-end microfiltration cell in order to mitigate fouling caused by the presence of colloidal bentonite particles. The effect of ultrasonic power and pulse duration on the permeate flux recovery was examined. Measurements indicate that an increase in ultrasonic power and longer pulse duration results to a higher permeate flux recovery. In order to reduce power consumption, a low to high power shift (LHPS) and pulsation method, were investigated. Methods of cleaning such as ultrasonic irradiation, ultrasonic cleaning with forward flushing and ultrasonic cleaning with backwashing were utilized and their cleaning efficiencies were examined. The cleaning performance was assessed using the clean water flux method and scanning electron microscope analysis of the cleaned membranes. Results showed that LHPS and pulsation method both improve the permeate flux recovery but were not able to attain the 93.97 and 74.88% flux recovery for PES and PVDF that was achieved by constant-15 W ultrasonic cleaning. In addition, forward flushing and backwashing may enhance the performance of ultrasonic cleaning at 9 W but could become disadvantageous at 15 W.

Keywords: backwashing; forward flushing; fouling; membrane; ultrasonic irradiation

1. Introduction

Membrane technology has been widely applied in water and wastewater treatment, especially in the removal of dissolved and suspended solids from industrial and municipal wastewater [1]. Its wide spread application is due to its ability in producing a high quality of permeate. A major disadvantage of membrane technology is fouling, which refers to the deterioration of membrane performance due to permeate flux decline caused by pore blocking, concentration polarization and cake formation [2]. The replacement of a membrane due to fouling is one of the largest operating costs in a water treatment plant [3]. Therefore, mitigation of membrane fouling becomes important in order to maintain the economic viability of the treatment process. Previous studies have developed a variety of approaches in reducing membrane fouling such as synthesis of new membrane materials [4], development of a new membrane unit [5] and membrane cleaning methods [3,6].

Current technologies that are used in reducing membrane fouling are chemical, hydraulic and electrical cleaning [7,8]. These technologies have certain disadvantages, where hydraulic cleaning such as backwashing and forward flushing does not remove efficiently strong adherent films or material trapped within the pores of the membrane [8]. In addition, hydraulic processes are time-consuming and often interrupt the filtration operation [1]. Chemicals used in membrane cleaning cause damage to the membrane material, resulting in secondary pollution and are expensive [7,9,10]. Application of the electrical technique is limited due to corrosion of electrodes, high power costs, and the induced electrolysis effect [1,9].

Ultrasound has been widely utilized in cleaning lenses, jewellery, semiconductor wafers and steel strips [1,7]. Ultrasonic cleaning utilizes the cavitation phenomenon, which produces turbulence and high shear velocities that dislodge particles from the membrane surface [9]. In comparison to the conventional cleaning methods, ultrasonication has several advantages such as high flux recovery, no chemical waste generated and an *in situ* operation is made possible [11]. However, ultrasonic irradiation is not economically attractive due to its high-energy consumption. In the literature, ultrasonic frequencies ranging from 70 to 1062 kHz were utilized in cleaning fouled ceramic membranes [12] while 42 kHz was applied in cleaning fouled polyvinylidene fluoride (PVDF) hollow fibre membranes [2]. Meanwhile, an ultrasonic power of 19 W was

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used for cleaning silica-fouled ceramic membranes [8] and 375 W was applied for cleaning hollow-fibre ultrafiltration membranes [1]. Due to the high ultrasonic frequency and power applied in previous studies, current investigations focused on finding ways to decrease the energy requirement by optimization, pulsation and combining ultrasonic irradiation with other cleaning methods such as hydraulic and chemical cleaning. Previous studies have focused in modifying the feed characteristics, membrane properties and operating temperature in order to enhance the use of ultrasound in membrane cleaning. On the other hand, the pulsation method, which is not widely studied, has shown potential on reducing the power consumption but its performance is slightly ineffective compared to continuous ultrasonic cleaning [12]. In addition, combining ultrasonic cleaning with backwashing, forward flushing and chemical cleaning further enhances the membrane cleaning efficiency [2,6].

Numerous studies have been performed in optimizing the membrane cleaning process using ultrasonic irradiation in terms of altering the power intensity, feed characteristics, colloidal particle size and ultrasonic frequency. Only a few studies on ultrasonic irradiation were done regarding a reduction of power consumption as well as the combined methods of ultrasonic and hydraulic cleaning. In this study, a new approach to reducing the energy consumption of membrane ultrasonic cleaning is investigated. The low to high power shift (LHPS) method (i.e. ultrasonic cleaning will start from a lower power then to its optimum power) is utilized and its performance will be compared to the pulsed method in ultrasonic membrane cleaning.

The experiments were performed using a dead-end microfiltration set-up with an ultrasonic probe system, to investigate the capability in cleaning fouled polyethersulfone (PES) and PVDF by ultrasonic irradiation. The effect of ultrasonic power and pulse duration on the permeate flux recovery was examined. In order to reduce power consumption, the efficiency of applying LHPS and pulse duration method was evaluated against continuous ultrasonic irradiation. In addition, the effectiveness of various cleaning methods such as backwashing, forward flushing and ultrasonic irradiation combined with hydraulic cleaning was also studied.

2. Experimental

2.1. Chemicals and equipment

Bentonite and $Al_2(SO_4)_3$ were procured from Sigma–Aldrich (USA) while HCl (37% fuming) and NaOH pellets (99% purity) were obtained from Merck (Germany). All reagents were of analytical grade and used without further purification.

GE Osmonics manufactured the PES and PVDF membranes with nominal pore size of 0.45 μm, effective area of 1.96×10^{-3} m^2 and 5.0 cm diameter. Turbidity of the synthetic raw water was measured using Hach 2100P turbidimeter. The average particle size as well as the particle size distribution curve was obtained using the Malvern 2000 particle size analyser. The surface morphology of fresh and fouled membranes was examined using a scanning electron microscope (SEM S-4800 Hitachi, Japan).

2.2. Preparation of synthetic raw water (clay solution)

Synthetic raw water was prepared by adding 20 g bentonite to 2 L of tap water. It was stirred at 200 rpm for 15 min to uniformly distribute the particles, and the mixture was left for 24 h. Approximately 1.8 L of supernatant was decanted and vigorously agitated at 200 rpm for 10 min. The suspension was used as stock solution. About 200 mL of the bentonite stock solution was diluted to 2 L with tap water. The general characteristics of the synthetic raw water are shown in Table 1.

2.3. Dead-end microfiltration system

The dead-end microfiltration set-up used in this study is shown in Figure 1.

The set-up is composed of a feed tank, membrane, stirrer, ultrasonic probe, permeate tank, electronic scale, computer and vacuum pump. The feed tank, which is made of Plexiglas, has a 12 L capacity. The stirrer is used during coagulation and flocculation of synthetic raw water. Subsequent to flocculation, it is continuously operated at 30 rpm to keep the flocs suspended. A horn-type ultrasonic probe (Sonicator3000, Misonix, USA), is positioned about 2.5 cm above the membrane surface. The feed water passes through the membrane using the vacuum pump operated at constant 50 kPa. The permeate was drawn from the membrane using vacuum suction produced by the vacuum pump (Stainless Vacuum Pump 60 Hz Rocker 400), where the pressure could be controlled digitally. The permeate enters the tank, which was placed over an electronic balance (High Precision Balance BH-3000 Series). The electronic scale measures the cumulative mass of the permeate, and then transmits it to a computer where it is recorded at specific time intervals.

Table 1. Characteristics of synthetic raw water.

Parameters	
Turbidity (NTU)	160 ± 10
pH	8.2–8.4
Conductivity (μS m^{-1})	358.37
TDS (mg L^{-1})	241.07
TSS (mg L^{-1})	201.80
Total Hardness (mg L^{-1})	185.08
Ca^{2+} (mg L^{-1})	51.60
Mg^{2+} (mg L^{-1})	13.61
K^+ (mg L^{-1})	17.96

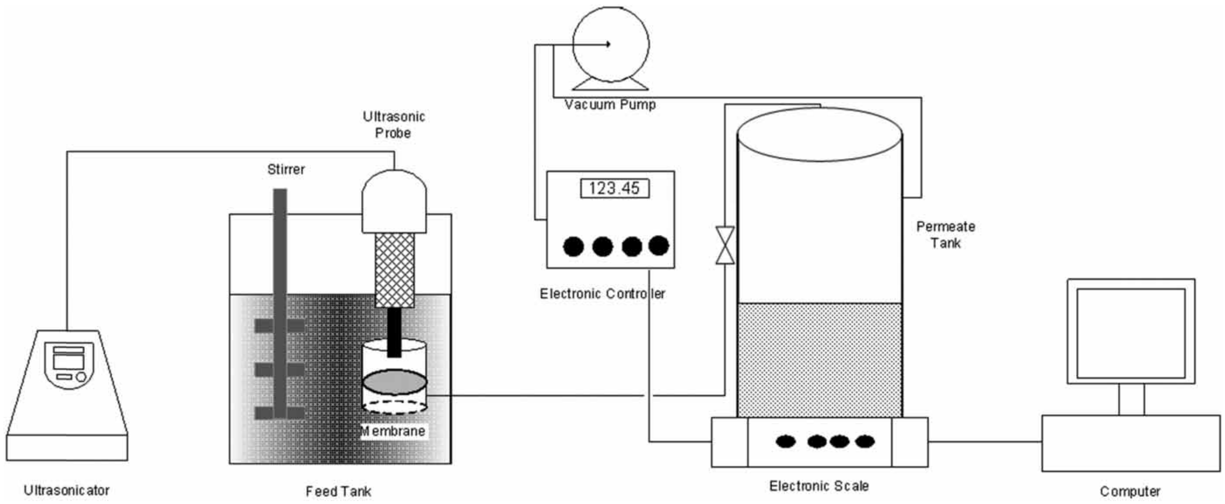

Figure 1. Schematic diagram of the ultrasound and dead-end microfiltration experimental set-up.

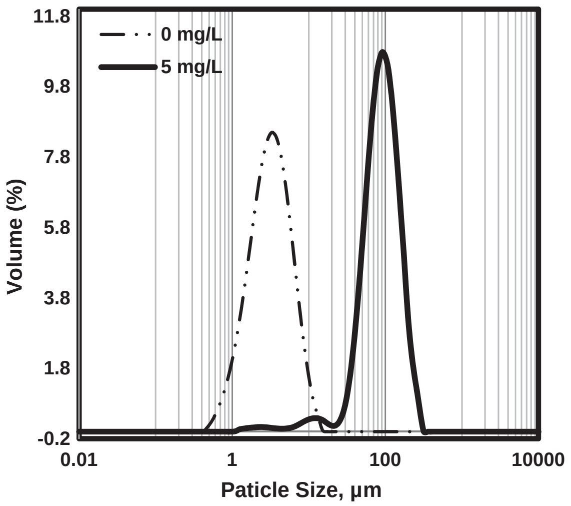

Figure 2. Particle size distribution of flocs under 0 and 5 mg L^{-1} $Al_2(SO_4)_3$ concentration.

2.4. *Determination of flux recovery*

The experiment consists of 5 stages: coagulation and flocculation, initial clean water flux, fouling, membrane cleaning and final clean water flux.

2.4.1. *Coagulation and flocculation of synthetic raw water*

About 8.0 L of synthetic raw water was placed in the feed tank. The temperature and pH of the feed water were maintained at 25°C and pH 7.0 ± 0.1 using 0.5 N NaOH and 0.5 N HCl. About 5 mg L^{-1} of $Al_2(SO_4)_3$ was added and stirred at 200 rpm followed by slow mixing at 30 rpm. The particle size distribution of non-coagulated and coagulated (5 mg $L^{-1}$$Al_2(SO_4)_3$) raw water is shown in Figure 2.

In raw water, the particle sizes range from 0.48 to 13.18 µm. Upon addition of $Al_2(SO_4)_3$, an enlargement of particles was observed with a corresponding shift in the distribution curve in the range from 4.37 to 316.23 µm.

2.4.2. *Initial clean water flux*

The membranes were stabilized by passing deionized (DI) water through the membrane for 5 min or until the flux became constant. The operating pressure was maintained at 50 kPa. The steady-state flux represents initial clean water flux.

2.4.3. *Fouling*

Fouling of the membrane was carried out for 15 min using the coagulated synthetic bentonite solution, maintaining a trans membrane pressure of 50 kPa. Preliminary experiments were performed (Figure 3), where the optimum dosage of 5 mg L^{-1} $Al_2(SO_4)_3$ for coagulation/flocculation and microfiltration showed that after 15 min of fouling, the permeate flux became constant.

2.4.4. *Membrane cleaning*

2.4.4.1. Ultrasonic cleaning. The fouled membrane was placed in a DI water bath and ultrasonically cleaned for 5 min at 25°C using a probe type sonicator. Preliminary experiments were carried out; where 5 min is the shortest cleaning time that would recover the permeate flux (Figure 4).

The effect of ultrasonic power and pulse duration on percentage flux recovery was examined by varying the power from 3 to 24 W and pulse duration from 1 to 10 s, respectively. A blank experiment was carried out by submerging the fouled membrane in DI water for the same cleaning time of 5 min.

The effect of LHPS was done by setting the ultrasonic cleaning at low power in the range of 3 to 9 W for 2.5 min followed by membrane cleaning at optimum ultrasonic power for 2.5 min.

2.4.4.2. Combined cleaning. Forward flushing and backwashing were combined with ultrasonic cleaning, where same method and conditions for fouling and cleaning were applied. Forward flushing and backwashing utilized a flow rate of 0.5 L min^{-1} of the permeate water for 1 min.

2.4.4.3. Final clean water flux. DI water was allowed to pass through the cleaned membranes for 5 min or until steady-state flux was achieved. This would represent the final clean water flux.

2.5. *Calculation of flux recovery*

The permeate flux recovery was calculated using Equation (1):

$$\%\text{recovery} = \frac{J_{fw}}{J_{iw}} \times 100 \tag{1}$$

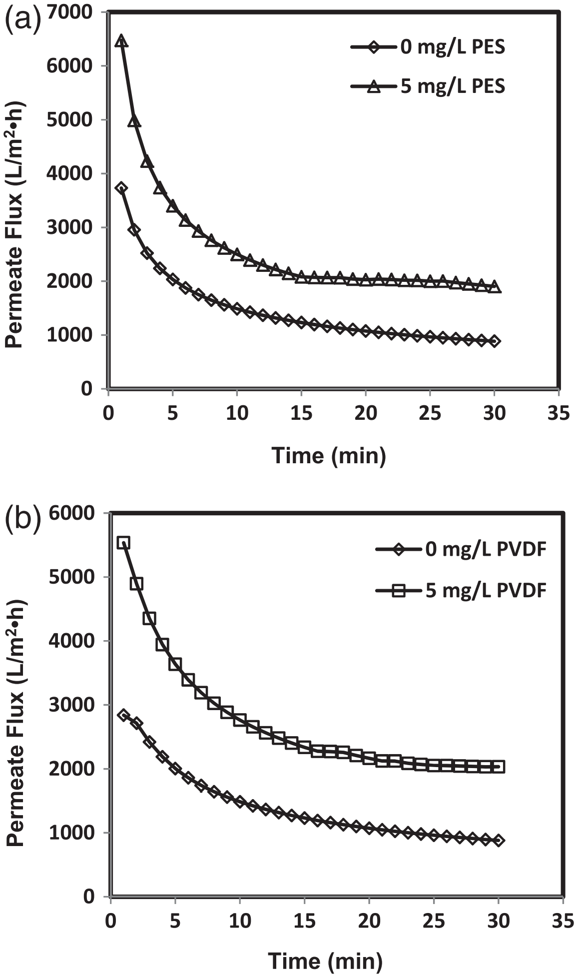

Figure 3. Permeate flux under different $Al_2(SO_4)_3$ concentration using (a) PES and (b) PVDF membrane.

where J_{iw} is the initial water flux (m^3/m^2 s), which is determined by filtering DI water until constant flux is achieved. Meanwhile, J_{fw} is the final water flux (m^3/m^2 s), which is obtained by passing through DI water in the fouled membrane after removal of the cake formed on the membrane.

3. Results and discussion

3.1. *Effect of ultrasonic power*

In order to study the effect of ultrasonic power on flux recovery, membrane cleaning was performed under varying power from 3 to 24 W with a sonication time of 5 min. As shown in Figure 5, there is an increase in the permeate flux recovery with an increase in ultrasonic power.

As the power applied to the membrane system was increased, a high-pressure amplitude of the sound wave is

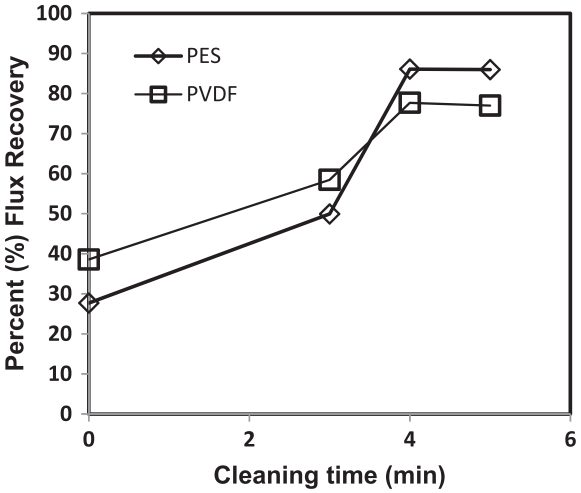

Figure 4. Permeate flux recovery of PES and PVDF using different ultrasonic cleaning time.

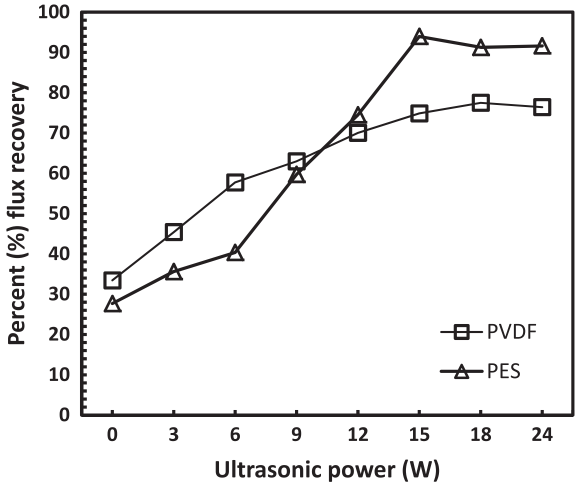

Figure 5. Effect of ultrasonic power on permeate flux recovery of PES and PVDF membranes.

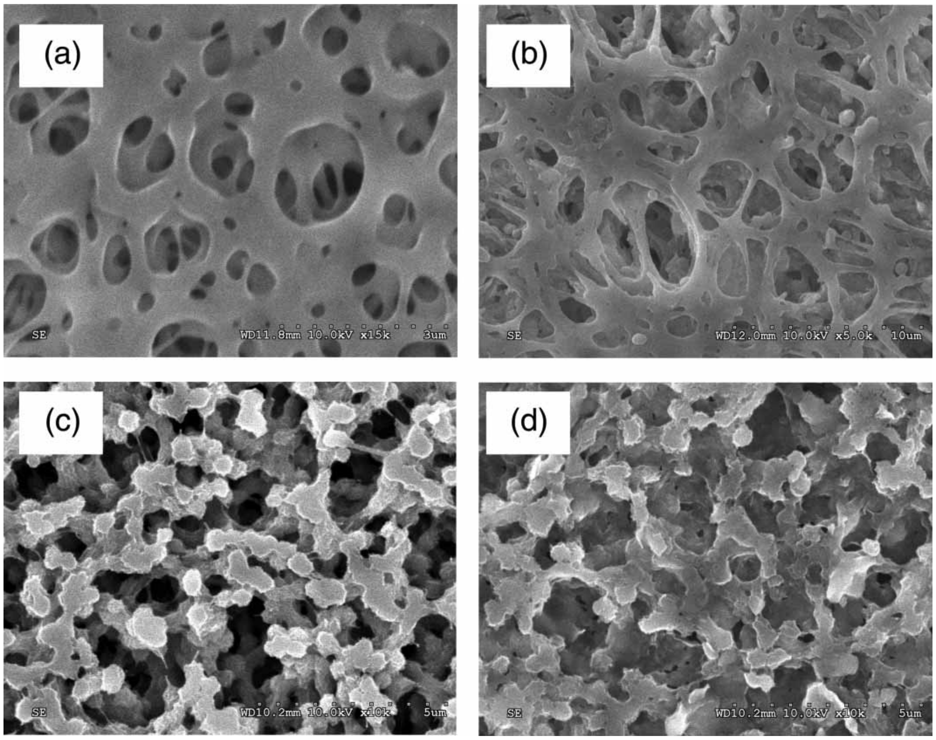

Figure 6. SEM images: (a) unfouled PES; (b) cleaned PES at 15W; (c) unfouled PVDF; and (d) cleaned PVDF at 15 W.

created, causing an increase in the number of cavitation bubbles formed and greater size of cavitation zone [7,13,14]. There is also an increase in the hydrodynamic turbulence caused by the high intensity of the implosion of bubbles. This is due to the greater number of bubbles that are able to collapse and an increase in the absorption of the acoustic energy of the system [7,15].

There is no significant increase observed in the flux recovery when the power applied is greater than 15 W. Instead, a slight decrease in permeate flux recovery is observed when power applied on membrane cleaning is higher than 15 W. This could be due to the excess power applied generating smaller particles that could block the membrane pores, causing a decrease in the permeate flux.

During the application of low ultrasonic power to the fouled membrane, PVDF has a slightly higher flux recovery over PES. This indicates that ultrasound at low power can only remove particles on the top most layer of the fouled material but it cannot fully remove the particles on the membrane surface. Upon the application of high power of 12 W or greater, PES has attained a higher flux recovery compared to PVDF, implying that PVDF is more difficult to clean.

Figure 6 shows the surface morphology of PES and PVDF, both clean and fouled. The PVDF membrane exhibits a rough and irregular morphology while the PES membrane has a smoother surface.

A rough surface would produce more tangential colloidal forces that could help immobilize other particles onto the membrane [16]. Surface roughness of a membrane also enhances the attachment of the colloidal particles onto the surface either through an increase in the attachment rate between colloidal particles (coagulation) or between a particle and the surface (deposition). Preferably, the colloidal particles accumulate and deposit in the 'valleys' of rough membranes [17]. In addition, a rough surface would provide greater contact area that would increase the rate of attachment by providing more opportunity for the particle to deposit onto the membrane surface [16]. All these effects contribute to severe fouling of a membrane with an irregular and rough surface. From Figure 6(b), the application of ultrasonic irradiation using 15 W was sufficient to restore the fouled PES membrane. On the other hand, Figure 6(d) illustrates that the membrane pores of PVDF remain clogged and some particles are still attached on the membrane surface even after ultrasonic cleaning. Therefore, PVDF proves to be more difficult to clean over PES.

3.2. *Effect of power shift*

According to the previous section, the optimum ultrasonic power in membrane cleaning is 15 W. In order to determine the effect of the power shift on the permeate flux recovery of the membrane a low ultrasonic power was combined with 15 W. The total applied sonication time is 5 min, where low power is applied for the first 2.5 min followed by the 15 W ultrasonic power for the succeeding 2.5 min. The combination of low to high power may significantly reduce the power consumption of the ultrasonic unit.

Table 2. Comparison of the permeate flux recovery of PES and PVDF membranes using LHPS and HLPS methods.

Power shift	% permeate flux recovery			
	LHPS		HLPS	
	PES	PVDF	PES	PVDF
3–15 W	78.78	67.33	63.19	67.21
6–15 W	85.07	73.78	73.75	69.33
9–15 W	88.15	73.79	80.01	72.05
Continuous 15 W	93.97	74.78	93.97	74.78

Table 2 shows the effect of low to high power shift (LHPS) and high to low power shift (HLPS) on the permeate flux recovery.

In PES using the LHPS method, the permeate flux recovery was observed to increase as the applied low power was increased from 3 to 9 W. The combination of 9–15 W provided the highest flux recovery of 88.91%, which is still inferior in comparison to the 93.97% flux recovery obtained using constant 15 W ultrasonic cleaning. On the other hand, cleaning PVDF membrane provided almost similar values of 73.79% flux recovery using LHPS (9–15 W) with the 74.78% flux recovery obtained from continuous 15 W ultrasonic cleaning. This implies that despite any type of ultrasonic cleaning method used, PVDF proved to be more difficult to clean due to its rough, irregular surface in comparison to PES. In Figure 7, the SEM images of PES membrane after ultrasonic cleaning using continuous (15 W) and LHPS (9–15 W) method are shown.

The surface of the cleaned membrane using LHPS method (Figure 7(b)) appeared to have some particles that are still attached onto the membrane surface. On the other hand, application of continuous ultrasonic cleaning at 15 W caused the appearance of clear pore structure of the membrane (Figure 7(a)). A constant application of 15 W for 5 min was sufficient enough to break up, disperse and completely remove the fouling from the membrane surface.

A HLPS method was also utilized in membrane cleaning and its efficiency is compared to the LHPS method. A similar trend with the LHPS method was observed, as the low power was increased, the permeate flux recovery also increases. From Table 2, it is evident that LHPS is a more effective method of membrane cleaning in comparison to HLPS due to its higher permeate flux recovery.

In PES, the flux recovery using 15 W ultrasonic cleaning is 5.06% higher compared to the LHPS method that utilized a combination of 9–15 W. The power combination of 9–15 W only utilized 80% of the total power used by the constant 15 W ultrasonic cleaning. Although the difference in power saving is larger when compared to the difference in flux recovery, it cannot be concluded that 9–15 W is a more cost-effective method. Other parameters should be considered regarding the microfiltration cycle such as pumping cost and membrane life span. Meanwhile, in PVDF, similar flux recovery values of 73.79, 72.05 and 74.78% were obtained using LHPS, HLPS and continuous 15 W cleaning, respectively.

3.3. *Effect of pulse duration*

Another economical method to mitigate membrane fouling is the intermittent use of ultrasonic energy [18]. In this experiment, 15 W of ultrasonic power is intermittently applied under varying pulse duration from 1 to 10 s, with constant 5 s pulse interval with a total cleaning time of 5 min. Figure 8 shows the effect of pulse duration on the percent flux recovery at a constant pulse interval of 5 s. It could be observed that the permeate flux recovery increases as the pulse duration was increased caused by longer ultrasonic cleaning time.

The flux recovery becomes constant at pulse duration of 5 s, where further increase in the pulse duration has insignificant effect on the flux recovery. The maximum flux recovery using intermittent pulsed ultrasonication is 83.65 and 73.29% for PES and PVDF, respectively. In PES, the efficiency in flux recovery using intermittent ultrasonication is inferior in comparison to that of the continuous and LHPS ultrasonic method, which makes it the least effective method. However, in PVDF, 73.29% flux recovery using pulsed ultrasonication is almost similar in value to that of continuous 15 W (74.78%), LHPS (73.79%) and HLPS (72.05%). This indicates that regardless of the ultrasonic

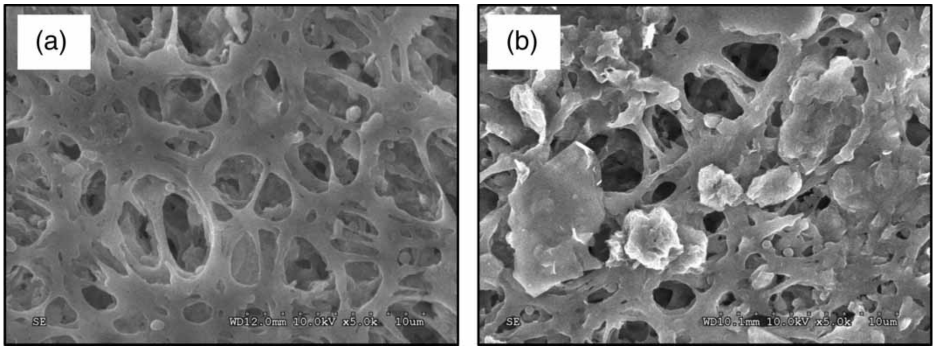

Figure 7. SEM micrograph of cleaned PES membrane using (a) continuous 15W and (b) LHPS at 9–15 W.

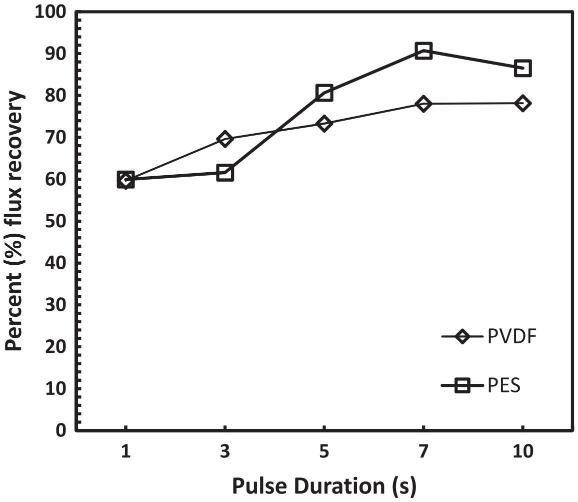

Figure 8. Effect of pulse duration on flux recovery for PES and PVDF membrane.

Table 3. Flux recovery of the fouled PES and PVDF membranes after cleaning using combined ultrasonication and hydraulic cleaning.

	% flux recovery	
Cleaning method	PES	PVDF
Backwashing	61.51	41.30
Forward flushing	66.43	54.72
Ultrasonic at 9 W	59.73	62.99
Ultrasonic at 9 W + backwashing	79.07	73.67
Ultrasonic at 9 W + forward flushing	74.39	60.55
Ultrasonic at 15 W	93.97	74.88
Ultrasonic at 15 W + backwashing	93.78	74.21
Ultrasonic at 15 W + forward flushing	81.51	73.00

cleaning method applied; almost the same flux recovery values were obtained.

3.4. Ultrasonic cleaning combined with hydraulic cleaning

In order to compare the efficiency of different cleaning methods, PES and PVDF membranes were fouled for 15 min using pre-coagulated bentonite solution. The membranes were cleaned using different methods for a total of 5 min for each cleaning process. The effectiveness of membrane cleaning by several methods like backwashing, forward flushing, ultrasonic irradiation or combination method (ultrasonication and backwashing, ultrasonication and forward flushing) was examined and the results in terms of flux recovery are listed in Table 3.

Forward flushing and backwashing both yield low flux recovery where backwashing appears to be the least effective method of membrane cleaning for PES and PVDF. This result is contrary to previous studies where backwashing is a more effective method than forward flushing in cleaning the fouled ultrafiltration polysulfone membrane used to treat a water reservoir in China and reclamation of municipal wastewater [19]. In backwashing, the backwash water flows against the direction of the permeate, where pores are cleaned and flushed from inside out, which removes particles clogged within the membrane pores [20]. Whereas, forward flushing brings the feed water forward to the membrane in order to remove any cake formation. In this study, the slightly higher recovery of forward flushing over backwashing may be due to the pre-coagulation method (5 $mg\,L^{-1}$ $Al_2(SO_4)_3$) performed before the microfiltration step. The enlargement of the bentonite particles due to coagulation favours the deposition of cake layer over pore blocking. During backwashing, the permeate water may have difficulty in penetrating the cake layer, which is usually dense and non-porous, on the membrane surface since less water could have push through the membrane pores from the permeate side [21]. In addition, the backflow water from the permeate to feed side causes expansion and fluidization of the cake layer [20], where the detached particles could redeposit on the membrane surface. On the other hand, forward flushing provides surface shear force through pumping of the permeate water on the feed side, where cross-flow velocity causes the cake layer to be flushed away from the membrane [20,22]. In addition, the total duration of hydraulic cleaning was only 1 min, which indicates that the cleaning time might be insufficient for backwashing to effectively remove membrane fouling. As shown in Figure 9, the SEM micrograph of the cleaned PES using backwashing has a few particles left on the membrane surface when compared to the clear pore structure of PES that was cleaned using forward flushing.

Ultrasonic cleaning at 9 W provided flux recovery of 59.73 and 62.99% for PES and PVDF, respectively. The permeate flux recovery of ultrasonication at 9 W significantly improved when combined with hydraulic cleaning. However, the combination of ultrasonic irradiation with backwashing provided a higher flux recovery over ultrasonic irradiation combined with forward flushing. The application of ultrasound has loosened the cake layer, and then backwashing lifted and removed the loosened and trapped particles from the membrane surface and within its pores [2,21]. Meanwhile, similar values of permeate flux recovery were obtained for ultrasonic irradiation (15 W) and ultrasonic irradiation (15 W) plus backwashing for PES. In addition, similar flux recovery values were achieved as well for ultrasonic irradiation (15 W), ultrasonic irradiation (15 W) plus backwashing and ultrasonic irradiation (15 W) plus forward flushing for PVDF. This indicates that ultrasonication (15 W) alone can effectively remove the foulants on the membrane and within its pores, where the aid of backwashing (for PES and PVDF) and forward flushing (for PVDF) is not necessary. Meanwhile, the combination of forward flushing with ultrasonication at 15 W (for PES) and 9 W (for PVDF) caused a decrease in the permeate flux. The application of ultrasonication causes the cake layer to loosen and break into smaller particle

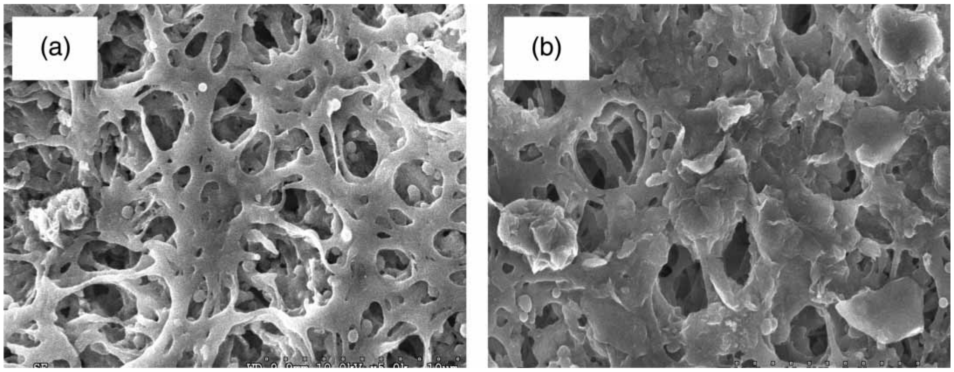

Figure 9. SEM images of fouled PES membrane cleaned by (a) forward flushing and (b) backwashing.

sizes [2]. As forward flushing was applied, the smaller particle sized-foulants were brought towards the feed side of the membrane surface, which led to clogging of the membrane pores [20,23]. Thus, combination of ultrasonication, at 15 W for PES and 9 W for PVDF, with forward flushing became disadvantageous. In addition, flux recovery of 73.67% attained using ultrasonic irradiation (9 W) plus backwashing has very similar values with those obtained from ultrasonic irradiation (15 W) and its combination with backwashing and forward flushing, which validates the difficulty in cleaning PVDF. Contrary to the results of previous investigations, this study has deduced that combination of hydraulic cleaning with ultrasonication does not always provide good impact on the flux recovery.

4. Conclusion

In order to reduce the fouling of polymeric microfiltration membrane, ultrasonic cleaning was utilized and its effects in improving the filtration process through permeate flux recovery was examined. Two polymeric membranes, PES and PVDF, were utilized in a dead-end microfiltration to investigate the effect of ultrasonic irradiation on membrane cleaning. Results showed that there is significant improvement on the recovery of permeate flux as the applied ultrasonic power and pulse duration were increased. The improvement in permeate flux recovery is attributed to longer cleaning times and an increase in the number of cavitation bubbles and acoustic energy in the system. In continuous ultrasonic cleaning, results show that the membrane can be effectively cleaned at an ultrasonic power of 15 W. The optimal power of 15 W was combined with low ultrasonic power in order to determine the effects of LHPS and HLPS on the permeate flux recovery. LHPS has greater permeate flux recovery compared to HLPS, implying that the top layer of the cake formed could be removed using low ultrasonic power. In the LHPS method, the highest flux recovery achieved was 88.91% in cleaning PES at 9 W for 2.5 min followed by 15 W for 2.5 min. A PES membrane has better flux recovery over PVDF due to the inherent rough surface of PVDF. Surface roughness causes severe membrane fouling due to increase in the rate of particle attachment onto the membrane surface.

The combined method of ultrasonic cleaning (9 W) and backwashing proved to be the most effective in the recovery of permeate flux while backwashing alone was the least effective method. The combination of ultrasonic (15 W) cleaning and backwashing provided no significant improvement in flux recovery when compared to ultrasonic cleaning at 15 W. Meanwhile, ultrasonic (15 W) cleaning combined with forward flushing causes the permeate flux to decline. Overall, combining ultrasonic and hydraulic cleaning does not always have a positive effect on the permeate flux recovery.

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References

[1] X. Li, J. Yu, and A.G.A. Nnanna, *Fouling mitigation for hollow-fiber UF membrane by sonication*, Desalination 281 (2011), pp. 23–29.
[2] A.L. Lim and R. Bai, *Membrane fouling and cleaning in microfiltration of activated sludge wastewater*, J. Membr. Sci. 216 (2003), pp. 279–290.
[3] H. Ma, C.N. Bowman, and R.H. Davis, *Membrane fouling reduction by backpulsing and surface modification*, J. Membr. Sci. 173 (2000), pp. 191–200.
[4] P. Wang, K.L. Tan, E.T. Kang, and K.G. Neoh, *Plasma-induced immobilization of poly(ethylene glycol) onto poly(vinylidene fluoride)microporous membrane*, J. Membr. Sci. 195 (2002), pp. 103–114.
[5] R. Bai and H.F. Leow, *Microfiltration of activated sludge wastewater-the effect of system operation parameters*, Sep. Purif. Technol. 29 (2002), pp. 189–198.
[6] K. Parameshwaran, A.G. Fane, B.D. Cho, and K.J. Kim, *Analysis of microfiltration performance with constant flux processing of secondary effluent*, Water Res. 35 (2001), pp. 4349–4358.
[7] M.O. Lamminen, H.W. Walker, and L.K. Weavers, *Mechanisms and factors influencing the ultrasonic cleaning of particle-fouled ceramic membranes*, J. Membr. Sci. 237 (2004), pp. 213–223.

[8] D. Chen, L.K. Weavers, and H.W. Walker, *Ultrasonic control of ceramic membrane fouling: Effect of particle characteristics*, Water Res. 40 (2006), pp. 840–850.

[9] J. Li, R.D. Sanderson, and E.P. Jacobs, *Ultrasonic cleaning of nylon microfiltration membranes fouled by Kraft paper mill effluent*, J. Membr. Sci. 205 (2002), pp. 247–257.

[10] X. Chai, T. Kobayashi, and N. Fujii, *Ultrasound-associated cleaning of polymeric membranes for water treatment*, Sep. Purif. Technol. 15 (1999), pp. 139–146.

[11] S. Muthulumaran, K. Yang, A. Sueren, S. Kentish, M. Ashokkumar, G.W. Stevens, and F. Grieser, *The use of ultrasonic cleaning for ultrafiltration membranes in the dairy industry*, Sep. Purif. Technol. 39 (2004), pp. 99–107.

[12] M.O. Lamminen, H.W. Walker, and L.K. Weavers, *Cleaning of particle-fouled membranes during cross-flow filtration using an embedded ultrasonic transducer system*, J. Membr. Sci. 283 (2006), pp. 225–232.

[13] H.M. Kyllönen, P. Pirkonen, and M. Nyström, *Membrane filtration enhanced by ultrasound: A review*, Desalination 181 (2005), pp. 319–335.

[14] T. Kobayashi, X. Chai, and N. Fujii, *Ultrasound enhanced cross-flow membrane filtration*, Sep. Purif. Technol. 17 (1999), pp. 31–40.

[15] T.G. Leighton, *The Acoustic Bubble*, Vol. 10, Academic Press, San Diego, 1997.

[16] M. Elimelech, X. Zhu, A.E. Childress, and S. Hong, *Role of membrane surface morphology in colloidal fouling of cellulose acetate and composite aromatic polyamide reverse osmosis membranes*, J. Membr. Sci. 127 (1997), pp. 101–109.

[17] K. Boussu, B. Van der Bruggen, A. Volodin, J. Snauwaert, C. Van Haesendonck, and C. Vandecasteele, *Roughness and hydrophobicity studies of nanofiltration membranes using different modes of AFM*, J. Colloid Interface Sci. 286 (2005), pp. 632–638.

[18] S. Muthukumaran, K. Yang, A. Seuren, S. Kentish, M. Ashokkumar, G.W. Stevens, and F. Grieser, *The ultrasonic cleaning for ultrafiltration membranes in the dairy industry*, Sep. Purif. Technol. 39 (2004), pp. 99–107.

[19] J.P. Chen, S.L. Kim, and Y.P. Ting, *Optimization of membrane physical and chemical cleaning by a statistically designed approach*, J. Membr. Sci. 219 (2003), pp. 27–45.

[20] J.M. Arnal, B. Garcia-Fayos, and M. Sancho, *Membrane cleaning*, Expanding Issues in Desalin. 3 (2010), pp. 63–84.

[21] F.T. Reguyal, *Ultrasonic irradiation combined with hydraulic cleaning of fouled PES and PVDF membranes*, M.S. thesis, University of the Philippines, 2011.

[22] W. Gao, H. Liang, J. Ma, M. Han, Z.L. Chen, Z.S. Han, G.B. Li, *Membrane fouling control in ultrafiltration technology for drinking water production: A review*, Desalination 272 (2011), pp. 1–8.

[23] N.N. Li, A.G. Fane, W.S.W. Ho, and T. Matsuura, *Advanced Membrane Technology and Applications*, John Wiley & Sons, Inc., Hoboken, NJ, 2008.