Ultrasonication Effects on Ultrafiltration Membrane Cleaning and Fouling Mitigation

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Abstract**—**Mitigation of fouling on hollow fiber ultrafiltration membrane using ultrasonication has been carried out. The effects of different contact times, at constant frequency and power of 28 kHz and 60 Watt, respectively on membrane cleaning with and without chemical agents were studied. Results showed that the best optimal membrane cleaning achieved were sonicating in 15 minutes without any chemical agents, with 57% recoveries. It should be noted that the presence of the chemical agent increased the effectiveness of ultrasonic cleaning compared to using water. The best combination method recovered the initial flux to at 67% using 1M of NaOH and 10 min sonication. This is likely the consequence of expanded cavitations movement occurring in the more surface-dynamic result.

Keywords**—**sonication, sludge solution, chemical cleanings, fouling.

I. Introduction

MEMBRANE fouling is one of the major problems encountered in membrane filtration, occurs by irreversible deposition of retained particles, colloids, macromolecules, salts, etc. Consequently, fouling causes significant decreased in permeate flux as a results of concentration polarization, plugging of membrane pores and adsorption of fouling material on the membrane surface or in the pore walls (1).

Currently, the most common membrane cleaning technologies for hollow fiber ultrafiltration membrane include hydraulic backwashing, chemical cleaning, electrical cleaning and mechanical cleaning (2). Through these methods are satisfiable but there are some drawbacks and limitations. Repeating back flushing in the hollow fiber membrane (3-5) results in degradation of maximum flux and often time consuming which could interrupt the total operation process. Chemical cleaning involves applying strong chemicals may damage the membrane materials and cause secondary pollution (6). Problems with other cleaning techniques include chemical costs, waste disposal and significant capital investments for equipment (7).

Alternatively, ultrasound has been widely applied as a cleaning method due to cavitation phenomena (8). Ultrasound cleaning is the use of an aqueous medium aided by ultrasound to remove the soluble and insoluble foreign particles through dissolution and displacement. Ultrasound is sound transmitted at frequencies beyond the range of human hearing. Ultrasonic energy is generated by piezoelectric transducer, which is powered by a generator. Various studies have been conducted using ultrasound as a method of mitigating membrane fouling over the last decade. The frequency of the ultrasound driving is an important parameter, which was mainly applied in the range of 20 to 100 kHz to create cavitation and scrubbing action, and power intensity was up to 1500 W. Various studies reported the enhancement of permeate flux using ultrasound (9-11), which attributed to phenomena related to bubble oscillations, acoustic streaming, and heating (12,13,14), which increase flux by affecting the concentration polarization at the membrane surface (15). Cavitation and acoustic turbulence generated by ultrasound are generally regarded as the major mechanisms of detaching particles and other foulants from membrane surfaces.

Kobayashi *et al.* (16) investigated the effect of permeate flux with ultrasound irradiation at three different frequencies of 28, 45 and 100 kHz. It should be noted that lower frequency of 28 kHz was more effective in cleaning fouled membranes. Similar results were obtained by Lamminen *et al.* (17). Maskooki *et al.* (18) tested three ultrasound frequencies as well as a combination of these both with and without a chemical cleaning agent (ethylene-diamine-tetra-acetic-acid, EDTA) and found that the best cleaning performance achieved when the frequency was alternated between 28 to 45 and 100 kHz. Chu *et al.* (19) reported that strong turbulent eddies of size 5 to 100 microns are induced around the collapsing bubbles and cavitation has been known to occur more rapidly at frequency between 20 to 40 kHz.

This study was to focused on establishing effective membrane cleaning procedures to maintain and/or produce a high permeate flux recovery for the hollow fiber membrane fouling during the filtration process of activated sludge solution. The cleaning procedures which, incorporated chemical (alkaline solutions), physical (backwashing and ultrasonic application) were conducted at room temperature.

A schematic diagram of the experimental setup was shown in Figure 1, where the feed solution was circulated in the tank by using peristaltic pump. During the experimental run, the retentates was recycled back to the feed tank. Permeate was collected and the volumes recorded for every 20 min interval for permeate flux calculation. It was subsequently poured back to the feed tank to maintain constant volume of the feed tank.

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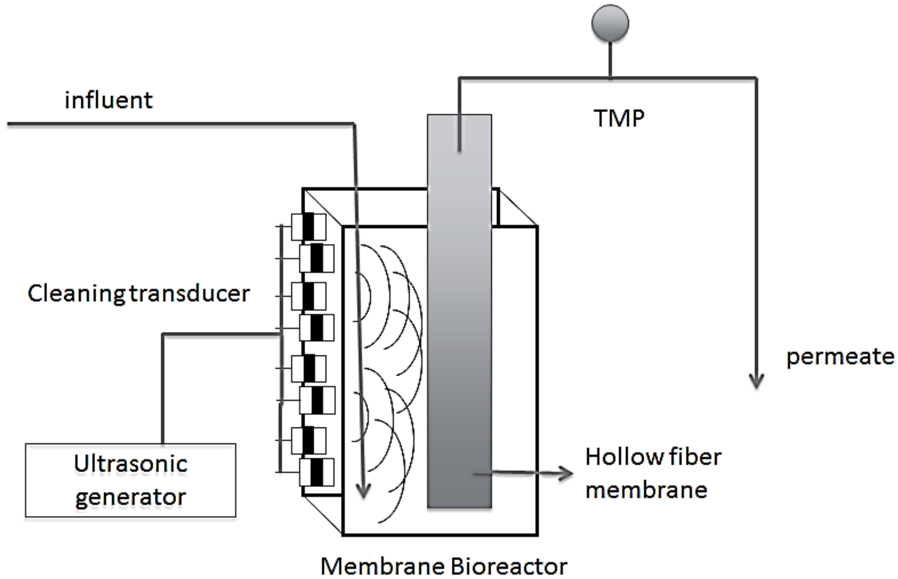


Fig. 1: Cleaning transducers for membrane fouling mitigation.

II. Literature Review

A. Experimental Apparatus

A plate type hollow fiber ultrafiltration membrane was used in this study. It was connected in series of membrane bundles with dimensions of 0.5 m height and 1.2 m width, giving an effective membrane area of 8 m^2. The feed solutions were prepared with the characteristics as mention in Table I.

TABLE I: Characteristics of Feed Solution.

Parameter	Bulking sludge solution	Granular sludge solution
Turbidity (NTU)	> 900	> 900
TSS (mg/L)	3100	3200
pH	8.41	8.18
Oxidation reducion potential (mV)	18.1	37.7
Conductivity (mS/m)	77	76
Dissolved oxygen (mg/L)	> 2.0	> 2.0
Total dissolved solids (g/L)	1.8	0.77

In this study, the membrane was completely submerged in a stainless-steel module of 40L capacity and eight units of piezo-electric ultrasonic transducers were attached at nominal power of 60 W output for an emission of 28 kHz each to generate ultrasonic waves and micro-bubbles. Table II shows the specification of the ultrasonic transducer. The first stage involved filtering the sludge solution for about 30 min so that substantial decrease in the permeate flux was observed. In the second stage, DI water was used to rinse the hollow fiber. The membrane flux was kept constant at about 20 L/m^2.h. The membrane permeate was extracted intermittently (7min on/4min off) with a negative pressure pump and being recycle back to the tank to maintain the water level. Trans-membrane pressure was used as the indicator of fouling in membrane. Cleaning of fouled membrane using sonication with and without chemicals were investigated at the selected contact time.

TABLE II: Specification of 28 kHz 60W Ultrasonic Transducer.

Parameter	Value
Frequency (kHz)	28
Power (W)	60
Capacity	3800pt (8pzt)
Radiating surface (mm)	59
Resonance resistance (Ω)	10 – 20
Piezo-ceramics size	38*15*5
Length (mm)	68
Power supply	Input: 220V – 240V AC
Weight (g)	500g/piece

B. Analytical Method

All activated sludge characteristics including MLSS and MLVSS were measured according to Standard Method (APHA, 1995). The membrane fouling was observed with scanning electron microscope couple with energy dispersive x-ray spectroscopy (SEM-EDX) (Thermo Scientific, accelerating voltage of 20 kV, Universiti Putra Malaysia). Before SEM-EDX analysis, samples were Au-Pd coated. TMP values were also measured.

III. Results and Discussion

Flux decay rates were greatest at the beginning of each operation but reduced significantly when TMP reach 30 kPa in 30 min. This indicates that fouling occurred rapidly when membrane module was put into operation.

A. Membrane Cleaning by Sonication

The effects of ultrasonication cleaning at different contact times, at constant frequency and power of 28 kHz and 60 W, respectively on membrane cleaning with and without chemical agents were studied. A hollow fiber membrane was filtered with sludge solution at high pressure (approx. 30 kPa) in 10 min to foul the membrane. Membrane filtration operated in longer pumping time with high trans-membrane pressure would lead to faster fouling. Membrane fouling is characterized in general as a reduction of permeate flux through the membrane, as a result of increase flow resistance due to pore blocking, concentration of polarization and cake formation. Pore blocking and cake formation can be considered as two essential mechanisms of membrane fouling.

TABLE III: EFFECTS OF ULTRASONIC CLEANING TIME ON PERMEATE FLUX.

Sonication time (min)	Initial water flux ($L/m^2.min$)	Water flux after fouling ($L/m^2.min$)	Water flux after ultrasonic cleaning ($L/m^2.min$)	Flux recovery (%)
5	1.87	0.10	0.90	48
10	1.87	0.12	1.07	57
15	1.80	0.05	1.05	58

Table III shows the effects of sonication time on the recovery of permeate flux at 5, 10 and 15 min of contact time. Significant improvement of permeate flux were observed. The best contact time recorded was at 10 min using water as solution. However, increasing sonication time to 15 min, improved flux recovery up to only 58%.

B. Membrane Cleaning by Sonication With NaOH as Cleaning Agents

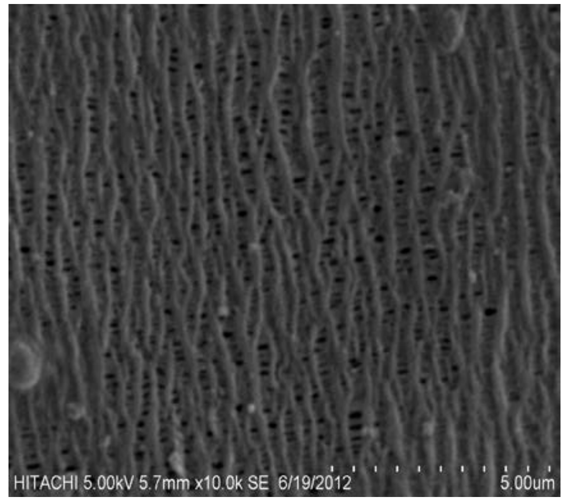
Experimental were repeated using NaOH as chemical agents (based on the best flux recovery previously) and sonication time at 5, 10 and 15 min, and results are presented in Table IV. It appears that 10 min of sonication is an appropriate duration as shorter and longer sonication durations did not give improvement in cleaning.

TABLE IV: EFFECTS OF ULTRASONIC CLEANING TIME ON PERMEATE FLUX.

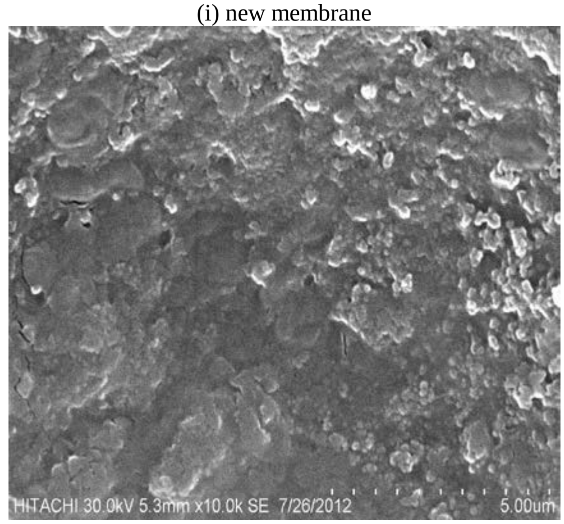
Sonication time (min)	NaOH	Water flux before sonication cleaning ($L/m^2.min$)	Water flux after ultrasonic cleaning ($L/m^2.min$)	Flux recovery (%)
5	1.0 M	1.82	4.14	56
10	1.0 M	1.83	5.55	67
15	1.0 M	1.82	5.35	66

Acoustic cavitation, vibration and other effects generated by ultrasound may destroy sludge floc structure and consequently reduce the average particle size and viscosity of solution. The best flux recovery observed was at 10 minutes with 67% recoveries. It should be noted that the chemical agent increases the effectiveness of ultrasonic cleaning as compared to using water. However, the increase in water flux after cleaning might due to increase in pore size of membrane due to high concentration of chemical cleaning. This is likely the consequence of expanded cavitation movement occurring in the more surface-dynamic result.

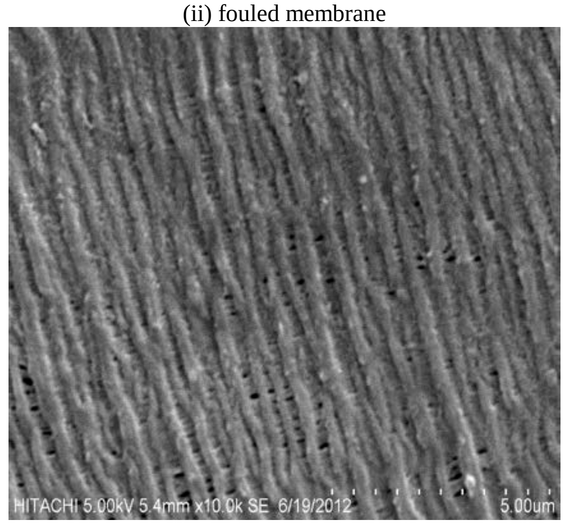
SEM analysis was performed from the membrane surface to verify that the flux enhancement is not due to the membrane destruction. The SEM images shows surfaces of clean membrane, fouled membrane, and fouled membrane after being cleaned by sonication, chemical cleaning and by the combination method. New membrane surface is observed to be porous and free of particles as shown in Fig. 2 (i). The surface of used membrane shows the presence of cake layer (Fig. 2 (ii)). Sonication cleaning removed most of the cake, and as a result, most of the pores became open (Fig. 2 (iii)). The membrane surface after chemical cleaning still have a lot of cake fragments on it as shown in Fig. 2 (iv).



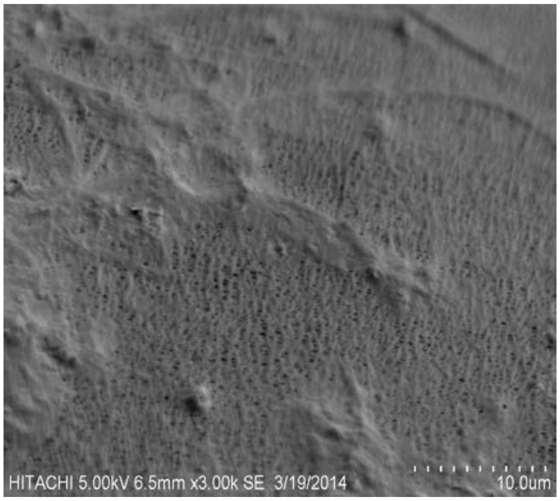
(i) new membrane



(ii) fouled membrane



(iii) cleaned by sonication



(iv) cleaned by chemical

Fig. 2: SEM image of membrane surface before and after cleaning.

IV. Conclusion

Pore blocking fouling prevails in the early stage of filtration, which causes a significant flux decline over time. Sonication method can be used to clean membrane fouling effectively. Results showed that the best removal achieved were at sonication in 15 minutes without any chemical agents, with 57% recoveries. It should be noted that the presence of the chemical agent increased the effectiveness of ultrasonic cleaning compared to using water. The best combination method recovered the initial flux to at 67% using 1M of NaOH and 10 min sonication. Therefore, it is feasible to apply low intensity ultrasound to improve membrane cleaning.

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