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# Membrane Defouling Using Microbubble Generated by Fluidic Oscillation

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## Abstract

Impurities and colloidal substances are two of many fouling conditions that reduce the membrane filtration performance used in wastewater treatment. This study investigates the potential of fluidic oscillation generated microbubbles (MBs) to defoul the filtration membrane. Cartridge filters of microfiltration (MF) of 1 $\mu$ m pore size were fouled using surface seawater collected from the Hull coastal area. The seawater was circulated at 5.8L/min to actuate colloidal substance deposition on the membrane surface. The recorded feed channel pressure drop ( $\Delta P$ ) across the membrane filters shown rapid fouling occurred in the first 8 hrs of the circulation. Fluctuations of  $\Delta P$  during the next 8hrs were observed showing the colloids filling the pores of the membrane, remaining steady for two hours showing membrane was completely fouled. The filtration membrane was cleaned and defouled using fluidic oscillator generated MBs. The fouled membranes were sparged with 1L/min of air scouring for ~1 to ~2hrs to remove the deposited colloids and impurities on the surface of the membrane. The membrane, analysed under Scanning Electron Microscopy (SEM), UV<sub>254</sub> and EC meter, shows the extent of MBs mediated removal of the deposited colloidal particle from the membrane surfaces. This study found that the highest defouling rate occurs with MBs generated by fluidic oscillator (closed vent), followed by MBs generated by fluidic oscillator (opened vent) and MBs generated without fluidic oscillator are 0.00953, 0.00622, and 0.00341 bar/min, respectively.

**Keywords:** Microbubble, Defouling, Membrane

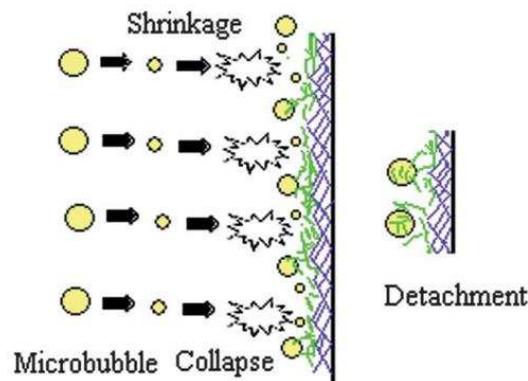
## INTRODUCTION

Membrane filtration approaches such as microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO) are very important for wastewater treatment and waste recovery (Giacobbo et al., 2015; Bhattacharya et al., 2013; Galanakis et al., 2013). They provide many advantages such as high selectivity, capacity and feasibility. However, they are easily fouled by biofouling, organic and colloidal substances which restrict the permeation rate and reduce the process efficiency. In general, fouling is usually caused by the deposition of small colloidal particles on the membrane surface and inner walls of membrane pores which results in the formation of cake layer (Zhao & Yu, 2014; Stephen P. Chesters et al., 2013). Conventional defouling methods such as chemical cleaning and pre-treatment usually are destructive and cause waste problems. Recently, innovative studies were conducted to explore the potential of MBs to clean the filtration membrane. Mechanisms such as creating MB pulsating-like action (Wilson et al., 2013), the behaviour of MBs by adsorption (Hiroyuki et al., 2015), and swarm velocity (Lee & Lee, 2002) clearly described the role of MBs in cleaning applications. Agarwal et al. (2012) and Wibisono (2014) listed four steps of cleaning using MB: 1) generation of smaller MBs 2) MBs burst to generate high-pressure spot and shear force 3) continuous biofilm matrix disruption 4) biofilm detachment. Based on the cleaning mechanisms mentioned, it is important to generate smaller microbubble high-pressure spots, however, it is unlikely to generate small MBs by only depending on the various size of pores, shear and material of diffuser system. Zimmerman et al. (2009) generate smaller size of MBs from the diffuser pore using fluidic oscillation by oscillating the feed air stream by pinching off the bubbles known as a hemispherical cap. Thus, fluidic oscillation microbubble is

generated to assist and compare with the conventional bubble cleaning method to restore membrane performance. MF membrane used as pre-treatment for desalination usually have shorter filter lifetimes due to fouling (Baker, 2004). This research mainly to exploit the advantage of using a cheaper way of producing smaller microbubbles in cleaning MF membrane (Zimmerman et al., 2011). Using MB cleaning, the performance of filtration membrane has been developed positively by prolong the membrane life and alleviating energy consumption (Wibisono et al., 2015; Fazel et al., 2013). Environmentally, this research will significantly bring the food and chemical industries towards green waste management by reducing the waste production and replacement of chemical cleaning agents (Stephen P Chesters et al., 2013; Mercier-Bonin et al., 2004).

### Membrane fouling and defouling

Fouling usually caused by the deposition of small colloidal particles on the inner walls of membrane pores. The blockages are a build-up of particles in the form of a cake layer on the membrane surface and membrane pore opening. The effect of permeation flux reduction due to fouling is twofold. First, pore blocking and cake formation lead to the increase in flow resistance. After that, the presence of colloidal particles deposited on the membrane surface hampers liquid mixing. Thus, a relatively high concentration of solutes persists near the membrane surface which causes the reduction of the solvent flux crossing the membrane (Henry et al., 2012).



**Figure 1: Schematic illustration of biofilm detachment by collapsing MBs (Agarwal et al., 2012)**

The potential of microbubbles for removing the biofilm depends on the internal pressure of the bubble. As governed by the Young-Laplace equation for spherical microbubbles (Eq.1), the internal pressure of the bubble depends on the diameter of the bubble. The smaller the diameter of the bubble lead to the higher internal pressure and subsequently bubble collapse resulted in higher energy. High energy generated allows more detachment of the biofilm and the cleaning efficiency. Fig. 1 above illustrates the pressure waves are distributed over the domain of the self-collapsing bubbles eventually dispel the fixed biomass from the membrane surface. The detachment of the biofilm is further simplified in Fig.2 below.

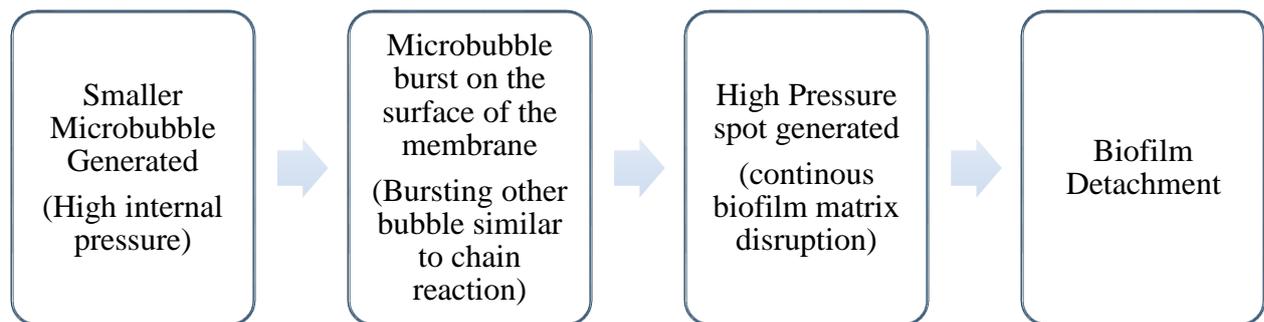
$$P = P_L + \frac{4\sigma}{d_b} \quad (\text{Eq. 1})$$

P: Gas Pressure

$P_L$ : Liquid Pressure

$\sigma$ : Surface Tension of the Liquid

$d_b$ : Bubble Diameter



**Figure 2: Simplified biofilm detachment using microbubble**

### **Fluidic Oscillator Microbubble Defouling**

The mechanisms of using MBs to defoul filtration membrane exemplified in Fig.1 and 2 show great potential in utilizing MBs to controlling and preventing membrane fouling. Lu et al., (2008) concluded that higher gas flowrate and smaller bubble sparging limit fouling better on hollow fibre MF membranes. MBs applied to MF membrane successfully enhance membrane performance by reducing TMP more effectively, enhance the critical flux, and induce lighter cake formation (Hwang & Wu, 2009; Lu et al., 2008; Yu et al., 2003). Lee et al., (2014) found that MBs are able to adhere to particulates and colloidal matters, thus causing them to float, disrupt the gel layer, and provide pyrolytic decomposition of protein.

Zimmerman & Tesar, (2010) patented a method of producing smaller microbubble using fluidic oscillator. This device acts as amplifier by oscillating the gas passing through. Zimmerman, (2014), listed several application of using smaller bubble generated by fluidic oscillator to strip components of liquid such as gas transfer in bioreactors, anaerobic digesters, and particle separation. Various applications of fluidic oscillator generated microbubble have been studied- - better oil emulsion separation (Hanotu et al., 2013), higher separation efficiency via microbubble distillation (Al-yaqoobi et al., 2016), better algal growth (Kamaroddin et al., 2016), and efficient yeast recovery (Hanotu et al., 2014).

In this paper, the study of defouling will be conducted using MBs generated by fluidic oscillation. A fluidic oscillator connected to the diffuser as shown in Fig. 3 is able to produce smaller bubble size (Hanotu et al., 2013). Instead of relying on the structure of porous material for the nozzles to generate smaller bubble, fluidic oscillations divert the jet overcoming the coanda effect to enable the pinch off of the hemispherical cap of bubble formation, resulting in nearly mono-dispersed, uniformly released microbubbles (Zimmerman et al., 2008). This device has no moving parts and is able to produce smaller microbubble at higher energy efficiency (Tesar, 2014).

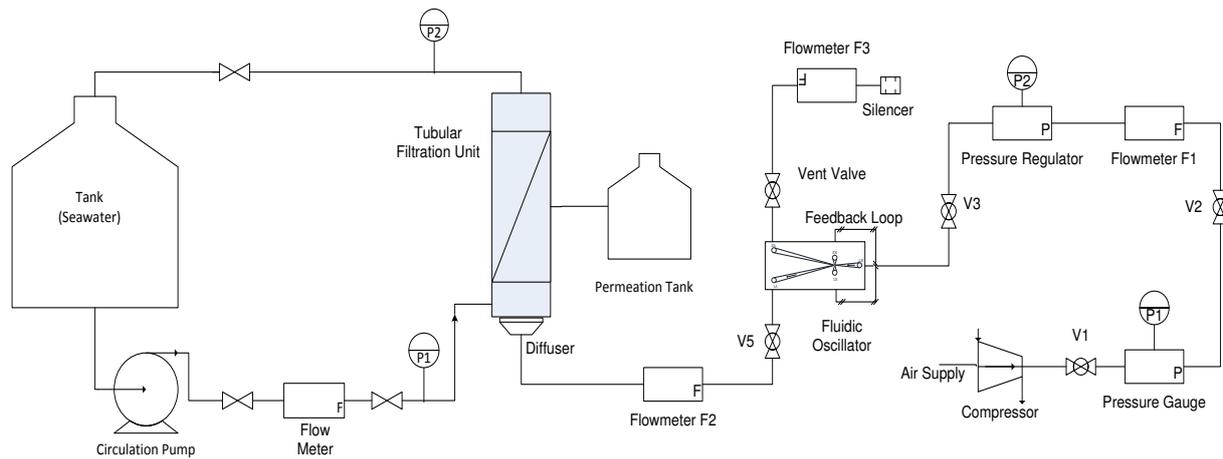
## MATERIALS AND METHODS

### Experimental design and setup

In this study, two main phase of experiment were conducted.

- I. Membrane fouling by circulating the seawater – mainly increase in pressure drop
- II. Microbubble sparging for membrane defouling – optimised membrane performance

### Microfiltration membrane defouling



**Figure 3: Schematic configuration of the membrane defouling system**

The filtration system was developed to remove colloidal substances from surface seawater and circulated at 5.4-5.8L/min. Filtration housing is fitted to a 10inch tubular MF membrane as shown in Table 1. Pressure drop of the filtration system recorded every 1 minutes. A flowmeter and pressure transducers (P1 and P2) are connected and recorded using an Arduino Data Logger. For the experimental start-up, the tubular unit was circulated with tap water for 24hr to allow soaking process and pressure to balance before being fed with seawater. 5ml of the effluent sample is collected at one minute intervals from the beginning and with every imposed pressure drop level. One tubular unit runs for 3 days. Once the pressure drop is constantly above 1.4bar, the system was sparged with microbubbles from air scouring unit. The air scouring unit consist of a control box (pressure regulator, valve, pressure gauge) were connected to the fluidic oscillator and diffuser. Air flow rate injected approximately 1L/min using alumina diffuser produces 100-1000 $\mu$ m size of microbubble to defoul the filtration membrane. The fluidic oscillator was operated with feedback loop length of 50mm. MBs sparged the membrane and membrane sample are analysed by SEM.

### Main membrane characteristics, pore sizes, and composition

Table 1 presents the information of the main membrane characteristics. Microfiltration membrane (MF) of 1micron pore size operated using crossflow configuration at room temperature during summer time. Seawater fed to the membrane are filtered specifically for the impurities and colloidal contained.

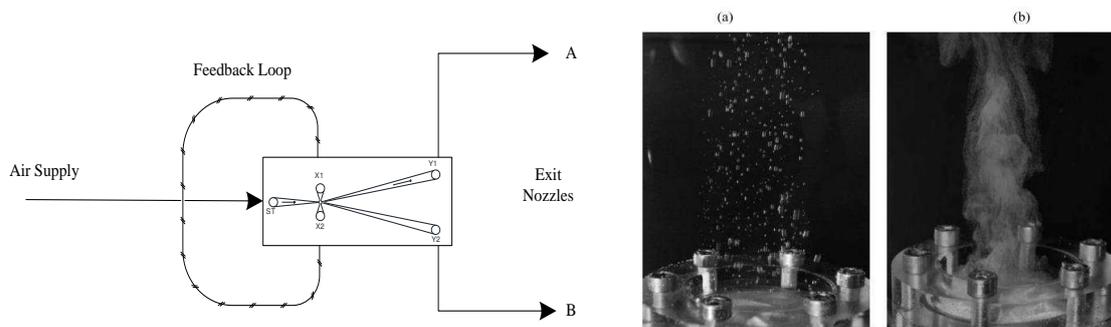
### Seawater and membrane sources

Seawater was collected from the East Riding of Yorkshire, England at Spurn and stored at room temperature (21 $\pm$ 4 $^{\circ}$ C) prior to all test. UV<sub>254</sub>, and pH of the seawater were at 0.034cm<sup>-1</sup>, an 8.0 respectively.

**Table 1.** Main membrane characteristics and operating conditions

	Properties
Type	Sediment cartridge filter
Material	propylene
Micron rating	1micron
Cartridge dimension	ID:30mm; OD:65mm; L: 255mm
Flow type	Inside-out
Membrane configuration	Tubular
Membrane System Setup	Cross-Flow
Temperature	Room ~22.7-25.1(°C)
Pressure initiation	2bar

### Fluidic Oscillator



**Figure 4: Fluidic oscillator schematic with images of bubbles (a) coalescent and non-uniform bubble without fluidic oscillator (b) Uniform size and non-coalescent bubbles with fluidic oscillator (Hanotu, 2013)**

For microbubble generation, fluidic oscillator is connected to the air sources as shown in Fig. 4. There are many advantages using the fluidic oscillator in term of cost-effectiveness, robustness, reliability, immobile parts and no requirement of electricity (Zimmerman et al., 2011). The bubbles generated by fluidic oscillation have low energy consumption that distinguish the method from other method such as ultrasonic and rotary disk which require a significant supply energy (Abdulrazzaq et al., 2015). Zimmerman et al., (2008) explained this device acted as a fluidic amplifier with a potential to pinch off hemispherical cap bubble. This early break off of bubble formation at the diffuser aperture offers smallest possible bubble size. Fig.2 and Eq.1 illustrates the relationship of the smaller microbubble to have higher surface area to volume ratio which lead to momentum transfer rates, especially for scrubbing the surface of membrane (Abdulrazzaq et al., 2015; Agarwal et al., 2012). A fluidic oscillator mainly consists of 3 parts: one inlet for air supply, two mid ports for the feedback loop, and two exit ports as the oscillation channel outlet. The arrangement as shown in Fig. 4 oscillates the gas flow between two paths under constant pressure of gas (Zimmerman & Tesar, 2010). A remarkable feature of this system is that the frequency of the oscillation is adjustable by manipulating the air flow rate and the length of the feedback loop (Tesař, 2014). Fig. 4 shows the effect of microbubble size generated with (a) and without fluidic oscillator (b).

## MBs operating conditions

MBs were generated using the scouring unit which is connected through the alumina diffuser at the bottom of the filtration housing. The air was injected through the diffuser at flow rate and pressure of 1 l/min and 2.2bar respectively. The bubble size generated are in the range of 100-1000micron. The following MBs conditions were generated:

- I. 1L/min of flow with slightly open vent valve
- II. 1L/min of flow with fully closed vent valve
- III. Non-fluidic oscillator generated microbubbles

## Data Collection and Measurement

### *Arduino Pressure Transducer and flowmeter*

Two pressure transducers were installed at the inlet, P1 and outlet, P2 to measure the pressure drop while the flowmeter connected after the circulation pump. The analogue reading for both of this instruments are connected to Arduino Uno data logger. The data were collected at 1minute intervals using the PLX-DAQ excel sheet.

### *Nutrition Controller*

Continuous monitoring of the pH value, Total Dissolve Solids, TDS (EC) and temperature of the feed were inferred using the Continuous Monitor Hydroponics trimeter. The sensors were placed in the feed tank. The nutrition controller collected the pH value, TDS and temperature of the system. The sample was collected and analysed using spectrophotometer.

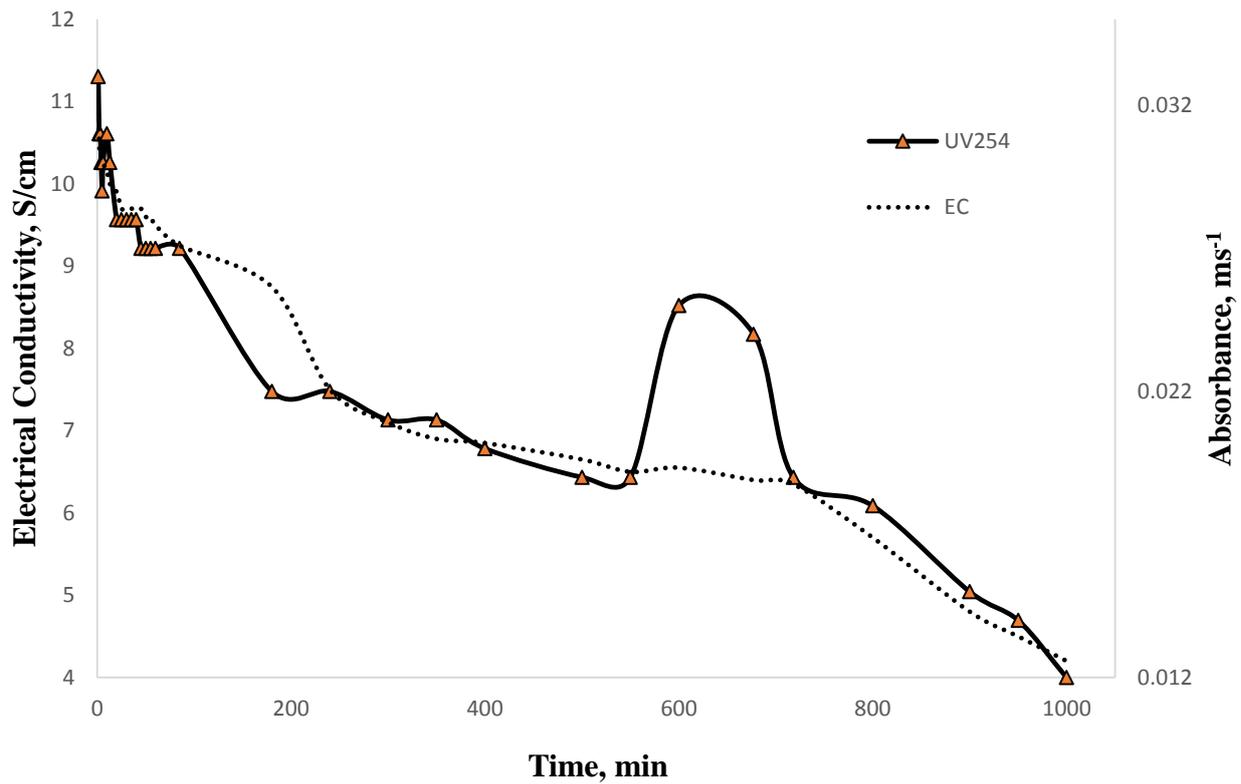
### *UV/Vis and SEM*

The UV absorbance of the water was measured at 254nm using UV/Vis spectrophotometer (Jenway 6705). At the beginning of the experiment, during microbubble sparging, and if there were fluctuations of P2, UV absorbance were tested at the intervals of 1 minute for twenty minutes. With steady pressure drop, the absorbance measured in the interval of 30mins to 1hr. The surface of the membranes after the experiment was dried at 50°C for one night and coated using gold. The gold coated membrane surface was examined for the colloidal deposition and removal under the Scanning Electron Microscope (SEM).

## RESULTS AND DISCUSSIONS

### **Effect of microbubble on fluid properties**

Fig. 5 shows the Electrical Conductivity (EC) value collected from the trimeter nutrition controller. Both of the value decreases over time indicates that membrane was fouled. The rapid decrement of the absorbance and EC showing the dissolved solids were deposited on the surface of the membrane during the first 500minutes which is roughly after 8hrs of circulation. The values remain constant for about ~2hr showing the membrane filtration efficiency has dropped due to its ability to filter more particles is now limited. This finding is in agreement to the study conducted by (Gwenaelle et al., 2017) which stated that fouling could be initiated just after 15minutes of filtration. When the microbubbles were introduced to the system after the 700<sup>th</sup> minute, the absorbance value varies from 0.019ms<sup>-1</sup> to 0.0225ms<sup>-1</sup>. It could be assumed that some of the deposited particles on the membrane surface scrubbed by the microbubbles were recirculated through the filtration system. Both values, however, continue to decrease over time and after MB's treatment.



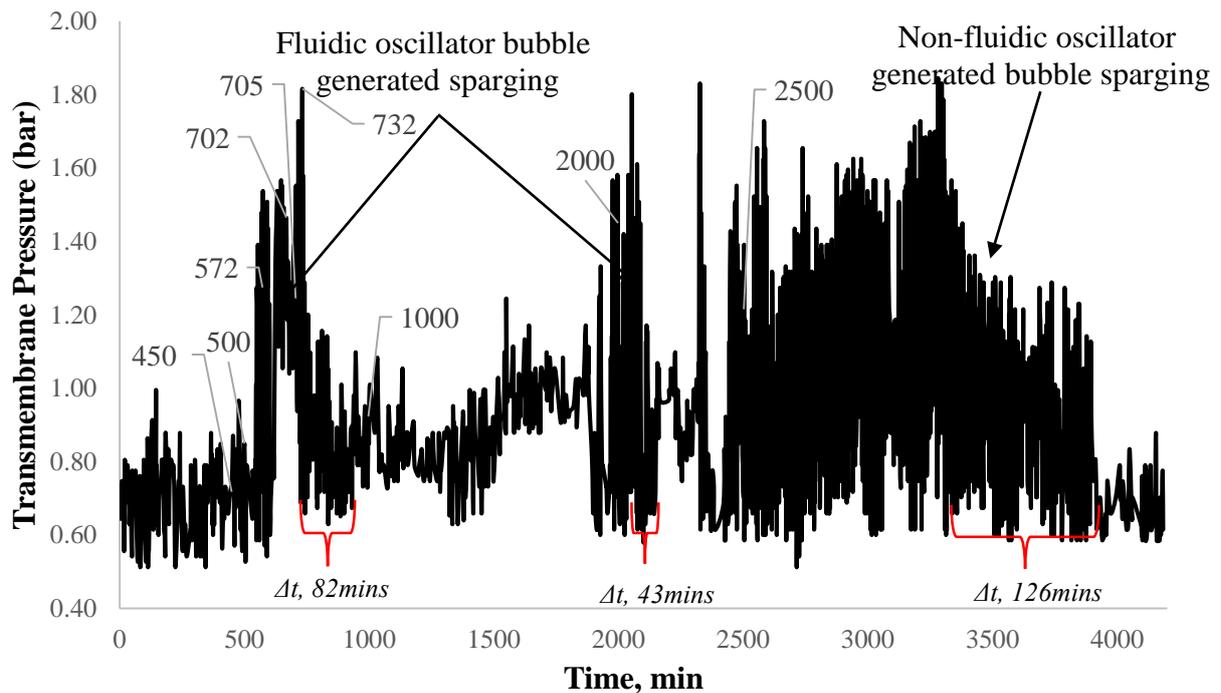
**Figure 5: UV<sub>254</sub> and EC value**

### **Overall fluidic oscillator generated microbubble cleaning**

Pressure is the main probe for examining the properties and results of this study. Increasing in transmembrane pressure drop (TMP) means that the filter is continuing filtering the impurities from the seawater and particles were deposited on the surface of the membrane. In this study, the microbubbles were introduced at the 600<sup>th</sup> minute to remove the deposited particles from the surface of the membrane.

Fluctuations in Fig. 6 shows the following experimental configuration in order.

- I. Slightly open vent valve: Better TMP reduction
- II. Fully closed vent valve: Best TMP reduction
- III. Non-fluidic oscillator generated MBs: Slowest TMP reduction



**Figure 6: Membrane fouling and defouling with and without fluidic oscillator generated microbubble**

#### Fluidic oscillator defouling with vent valve

Fig. 6 shows the pressure drop recorded for each 1minute interval for the whole filtration cycle of 72hrs (3days). The difference between the inlet and outlet pressure gives the transmembrane pressure (TMP) across the membranes. At the 450<sup>th</sup> minute, the pressure transducer recorded small fluctuations of pressure which heralds that impurities started to deposit and filling the membrane surfaces and pores. Rapid fouling was observed from 500min to 572min and starts to record a constant pressure drop until minute 702. The TMP fluctuates at minute 732 showing the pressure fluctuations due to pressure release from diffuser once microbubbles were introduced at 705min. This preliminary data showing the positive relationship between the bubbles and cleaning due to shear forces, drag forces, and strong velocity fluctuations induced by the bubble flow (Nagaoka et al., 2006).

After the sparging processes were stopped, the TMP started to increase. At 2000 minutes, a similar trend of the fouling was observed for which the TMP remains constant showing the fouling reaching saturation. Fluidic oscillator generated MBs once again are introduced with zero vent flow to the system. This resulted in higher defouling rate; where the bubbles possess sufficient or higher shear and drag force to detach the deposited particles. Higher TMP is also recorded after 1000 and 2500 minutes showing the MBs defouling were not able to restore the performance of the membrane to its initial conditions. The slowest defouling rate was recorded as non-fluidic oscillator generated MBs were sparged to the system as shown in Fig.6. It requires approximately 125minutes to reduce the pressure drop before it was fouled. The data shows that MBs generated by fluidic oscillator without vent valve flow has the highest efficiency of the defouling followed by the MBs generated by fluidic oscillator with vent valve and MBs generated without fluidic oscillator. The defouling assumption was in line with the dissolve particles as shown in Fig. 5 where the fluid quality was improved over the time. This

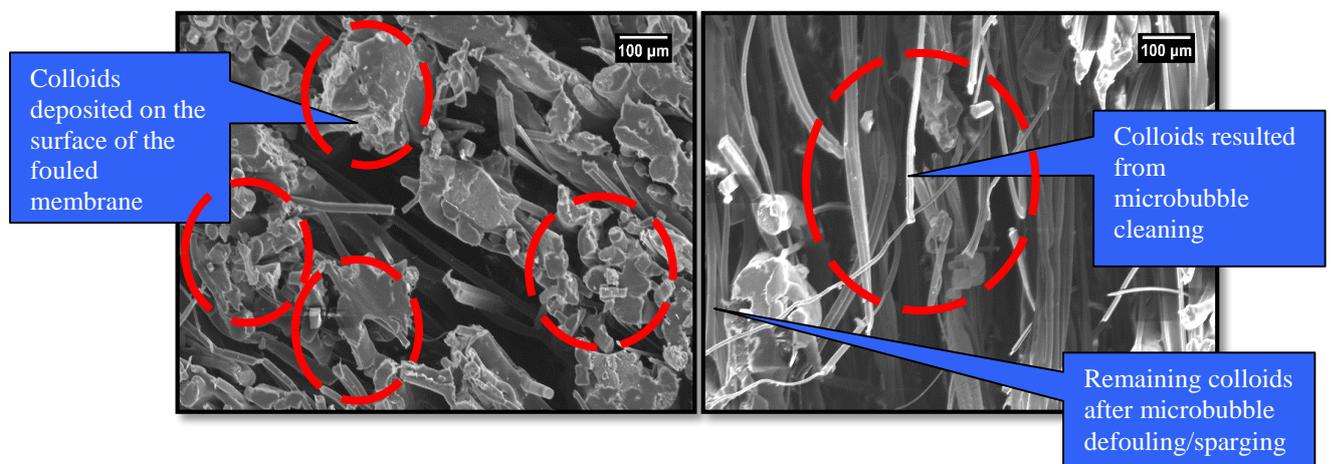
means that most of the particles were filling the pores and MBs sparging creates additional forces for better defouling.

### Fluidic oscillator and defouling rate

The highest TMP for each defouling were recorded at the time elapsed. Three defouling methods were applied, where the highest recorded defouling rate achieved by using fluidic oscillator (closed vent valve) at 0.00953bar/min followed by fluidic oscillator (open vent valve) at 0.00622bar/min and lowest defouling rate without fluidic oscillator at 0.00341bar/min. Assumed to have more flows with more shear and drag force, the MBs generated with fully closed vent valve by the fluidic oscillator has the highest defouling rate of 0.00953bar/min. MBs generated with flow in the vent valve by fluidic oscillator showing half of the defouling rate followed by the non-fluidic oscillator generated MBs and it is agreed by Lee et al., (2014). Wu, He, & Zhang, (2012) explained the limitation of larger size bubble on fouling control for the deposition of small particles.

Generally, the most efficient MF defouling is achieved by scouring the MF under fluidic oscillator generated MBs. This finding shows that fluidic oscillator generates smaller MBs. This results in higher efficiency of cleaning effect to scrub the colloids and impurities deposited on the surface of the MF (Lee et al., 2014; Wibisono, 2014). Zimmerman, (2014) explains the MBs generated by fluidic oscillations would inhibit repulsion between bubbles and particles for better particle separations which is also in agreement with the study conducted by Hiroyuki et al., (2015) and Agarwal, Ng, & Liu, (2011). This, however, leads to a different finding of using the fluidic oscillator towards the cleaning effect. The highest defouling observed while using oscillator without flow of air in vent valve. The basic inference from this is because more flow of air to the diffuser results in more bubbles generated compared to the oscillator with open vent valve. Manipulation of oscillator frequency by changing feedback loop length and bleeding flowrate is crucial to ensure smaller bubble generation (Brittle et al., 2015; Zimmerman et al., 2008) Fig. 4 illustrates the characteristics and function of the fluidic oscillator which need to be further investigated.

### Colloids deposition and its removal - Scanning Electron Microscopy (SEM)



**Figure 7(a) SEM images for the fouled and 7(b) defouled membrane**

Fig. 7(a) shows SEM images of the colloids deposited on the surface of the MF membrane. Because of MF has large pores, the filtration process will basically remove large size molecules

such as colloids. However, some salt particles might be also present due to the process of drying prior to SEM analysis. Fig. 7(b) shows the defouled membrane filter membrane after MB scouring. It can be clearly seen that MBs generated by fluidic oscillation scrubbed all of the impurities from the surface of the membrane. Nevertheless, not all of the impurities were removed. The result obtained is similar to the ones conducted by Gwenaelle et al., (2017) where not 100% impurities will be removed by MBs. It has also been suggested that combining MBs with other chemicals such as coagulant may help in improving the rate of impurities eliminated by MBs. The image however only showing the removal of the impurities by final sparging process (after 72hrs).

## **CONCLUSIONS**

The MBs are able to increase the effectiveness of membrane cleaning and defouling. The fluidic oscillator generated MBs resulted in higher defouling efficiency of filtration membrane. The TMPs recorded able to distinguish the relationship of fluidic oscillator and defouling rate. This, however, does not reflect the relationship of the fluidic oscillator and MBs properties towards defouling mechanisms without figurative data. Thus the following future works are very important to test all the hypotheses inferred from this study.

### *Future works*

General assumptions and preliminary relationship between fluidic oscillator and defouling were made as the membranes were defouled more rapidly with fluidic oscillations. This, however, requires further data measurement and analysis to distinguish a figurative relationship below.

- I. Installation of the bleeding valve to the fluidic oscillator outlet (system inlet).
- II. Bleeding valve flow rate to determine relationship between the flow rate of air and defouling.
- III. The oscillation frequency of the fluidic oscillator towards a better fouling and defouling.
- IV. Microbubble size distribution at different oscillation frequency.

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