Journal of Engineering Science and Technology
Special Issue on SOMCHE 2014 & RSCE 2014 Conference, January (2015) 1 - 13

ULTRAFILTRATION MEMBRANE FOULING AND CLEANING: A CASE STUDY IN HUTAN LIPUR PERANGIN SIK, KEDAH

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Abstract

Membrane technology is being widely applied all over the world in various field including biotechnology, pharmacy, food, drinking water production, wastewater treatment and many more. However, instead of the advantages in membrane separation, there are still limitations in the application such as flux decline due to membrane fouling which results in lower production and higher energy consumption to maintain the flux. Membrane fouling has been one of the main challenges when the rejected particles accumulated on the membrane surface or inside the pores. Membrane cleaning is necessary to minimize fouling effect to the membrane. This current study aims to determine the dominant fouling mechanism in ultrafiltration treatment of raw surface water which was conducted in Hutan Lipur Perangin Sik, Kedah. From the experimental data and model prediction fitting curve, cake formation has been found as the dominant fouling mechanism. Physical and chemical cleaning has been done to confirm the fouling type and minimize membrane fouling. Backwashing was chosen for physical cleaning varying different backwash pressure while five types of chemical cleaning agents have been used. It has been concluded that backwashing at 2 bar for 0.5 minutes is the appropriate method which can recover the permeate flux up to 82.96% recovery compared to chemical cleaning.

Keywords: Fouling mechanism, Backwashing, Ultrafiltration, Membrane fouling, Flux recovery.

1. Introduction

Malaysia is blessed with an abundant supply of water with 21,536 m^3 per capita water resources per year [1]. However, due to its growing economy, Malaysia will

Nomenclatures

J_A	Flux after cleaning
J_B	Flux before cleaning
J_0	Initial flux
J_t	Flux at time t
K	Coefficient
t	Time

Abbreviations

MF	Microfiltration
NF	Nanofiltration
PAN	Polyacrylonitrile
RO	Reverse osmosis
SEM	Scanning electron miscroscope
TMP	Transmembrane pressure
UF	Ultrafiltration

need to be more efficient in the water resources management and supply. The main source of raw water supply in Malaysia comes from rivers, storage dams and groundwater. As stated in the National Water Resources Study (Peninsular Malaysia) 2000-2050 [2], the government is focussing on identifying the demand and water resources to meet future needs in Peninsular Malaysia as well as to determine the availability of water resources up to 2050. Around 85% of Malaysia's raw water supply comes from rivers and stream. However, there are few rivers which are polluted thus the water cannot be used directly and the application of treatment system is needed.

Studies have been done to find out the most effective method to treat raw water supply. Previously, conventional water treatment method has been used for quite some time until the membrane technology is developed. For conventional water treatment method, combination of physical separation techniques for particle removal while biological and chemical treatments are carried out to remove suspended solids, organic matter and dissolved pollutants. Recently, membrane separation processes has become a convincing technology which provides effective solutions to meet human, environmental and industrial needs.

Membrane separation processes can be used for a wide range of applications and can offer significant advantages over conventional separation such as distillation and adsorption since the separation is based on physical mechanism. Membrane separation replaces or enhances the conventional water treatment methods by applying selective permeable barriers, with pore size to permit the passage of water molecules but small enough to retain a wide range of particulate and dissolved compounds depending on their nature. Most membrane separation processes do not occur chemical, biological or thermal changes of the component. In recent years, membrane separation such as microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO) have been applied in drinking water production for human consumption. MF and UF are recognized as the most attractive processes due to their low operating pressure requirement in treating and producing drinking water [3].

However, membrane fouling has been one of the main challenges due to the rejected particles accumulated on the membrane surface or inside the pores [4]. Membrane fouling might cause the increase in energy consumption during the membrane process for long term operation [5]. Previous studies by other researchers reported that the source of fouling in surface water is coming from natural organic matter (NOM) [6-8]. NOM is a mixture of organic compounds due to chemical and biological degradation of plant and animal residues which occur naturally in all surfaces water [9].

Therefore studies on membrane fouling and cleaning are carried out in order to recover the permeate flux to its initial flux. The cleaning method can be categorized into few types including physical and chemical cleaning. There are many types of physical cleaning such as hydraulic cleaning, air sparge and vibration. For drinking water production, hydraulic cleaning by backwashing are mostly applied [8]. During chemical cleaning, the cleaning chemicals react with the foulant by different chemical reactions such as hydrolysis, oxidation, saponification and chelation depends on the cleaning agent used.

Conventional drinking water treatment system is not economical and not appropriate to be applied for rural area. Rural residents normally rely on raw surface water without any treatment due to the location of distribution area. However, raw surface water is generally contaminated and harmful for human consumption. Many cases related with waterborne disease has been reported. Lack of suitable pre-treatment, inappropriate cleaning procedures for membrane cleaning are among the reasons that limit the application of membrane technologies for drinking water production in remote areas [9].

Thus the aim of this research is to study the type of fouling and determine the appropriate cleaning method for the membrane based on the membrane fouling analysis. For drinking water production from raw surface water, an integrated membrane system has been designed and installed near Hutan Lipur Perangin Sik, Kedah where the raw water from the top of Hutan Lipur Perangin Sik is fed to the system. By using the integrated membrane system, several equipments with different function are combined in one treatment system thus the treated water produced is safe for consumption and have better quality. Generally, the integrated membrane system might combine a membrane process with a conventional treatment units or any membrane process incorporated with another membrane process. A pre-treatment process is needed prior to membrane separation process in order to protect the membrane and extend the membrane's life span followed by post-treatment process to enhance the water quality produced.

2. Materials and Method

2.1. Membrane

Polyacrylonitrile (PAN) hollow fibre membrane was used in this study, which is commercially employed in ultrafiltration for drinking water production. Membrane characteristics data is summarized in Table 1.

Table 1. Membrane Characteristics Data.

Parameter	Membrane
Type	Hollow fiber
Material	PAN
Molecular weight cut off, Da	50000
Length, m	0.69
Number of hollow fiber	2500
Hollow fiber diameter, mm	2
Membrane surface area, m^2	10.87
pH-range of operation	4 to 9
Temperature-range, ^0C	5 to 45

2.2. Ultrafiltration treatment

Ultrafiltration process was performed using a pilot scale rig as shown in Fig. 1. Raw water is fed directly from the reservoir on top of Hutan Lipur Perangin Sik, Kedah through the sand filter as the pre-treatment unit and then filtered by the hollow fibre ultrafiltration module at transmembrane pressure (TMP) of 1 bar for 24 hours. Permeate was collected in the permeate tank which also will be used for backwashing.

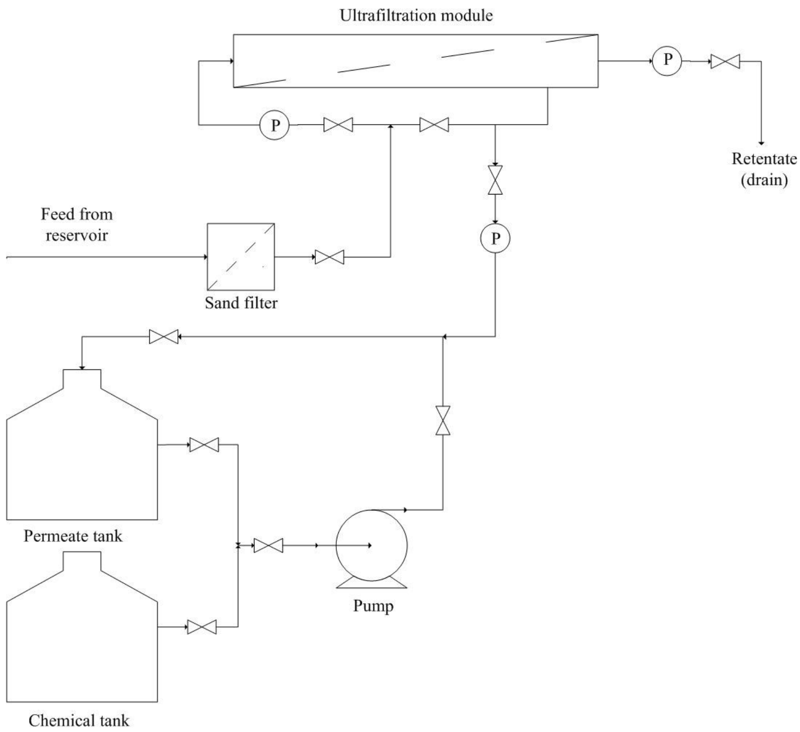


Fig. 1. Schematic Diagram of Ultrafiltration Rig for Drinking Water Production.

2.3. Membrane fouling study

Fouling occurred in most filtration process where the flux reducing during the filtration. In this research, the type of membrane fouling will be studied which are reversible and irreversible fouling. The source of fouling, fouling reduction and the appropriate cleaning for membrane will be discussed.

The fouling mechanism will be determined through experiment as well as using following models [10]:

$$\ln (J) = \ln (J_0) - K_c t \tag{1}$$

$$\frac{1}{J^{1/2}} = \frac{1}{J_0^{1/2}} + K_s t \tag{2}$$

$$\frac{1}{J} = \frac{1}{J_0} + K_i A t \tag{3}$$

$$\frac{1}{J^2} = \frac{1}{J_0^2} + K_{cf} t \tag{4}$$

where J_0 is the initial flux, J is the flux, K is the coefficient. The predicted flux from each model is compared to the experimental flux in the graph to determine the most suitable fouling mechanism. where J_0 is the initial flux, J_t is the flux at the time t, K is the coefficient. The predicted flux from each model is compared to the experimental flux in the graph to determine the most suitable fouling mechanism. Once the type of fouling is determined, the membrane cleaning will be done based on the source of fouling.

Model 1 contributes to the complete pore blocking and Model 2 describes the standard pore blocking mechanism. As for Model 3, it is based on the particles adsorption on the membrane pores contributed to intermediate pore blocking. Model 4 assumes that the entire surface is covered by a layered of particles and that the cake resistance is proportional to the cumulative permeated volume. The predicted flux is compared to the experimental flux in the graph to determine the most suitable fouling mechanism.

2.4. Membrane cleaning study

2.4.1. Physical cleaning

Physical cleaning was done by backwashing using permeate water for 0.5 minute every four hours of filtration. Backwashing pressure was chosen between 2 bar and 3 bar to determine the best backwashing pressure with acceptable flux recovery.

2.4.2. Chemical cleaning

For chemical cleaning, different cleaning agents were tested to recover the permeate flux back to the initial flux. The choice of the cleaning agents was based on literature [11-14]. The fouled membranes were immersed in five different cleaning agents which are NaClO, H_2O_2, NaOH, HNO_3 and $C_6H_8O_7$ with 0.25M concentration for 5 minutes. Results from chemical cleaning were finally compared to physical cleaning to determine the appropriate cleaning method for UF membrane.

2.5. Percentage of recovery

The effectiveness of the cleaning procedure in reducing fouling and recovering membrane performance was determined by comparing permeate fluxes before and after membrane cleaning. The percentage of recovery is calculated for each types of cleaning methods and conditions [15, 16].

$$Flux\ recovery, (\%) = \frac{J_A - J_B}{J_0 - J_B} \times 100 \quad (5)$$

Where J_A is the permeate flux after cleaning, J_B is permeate flux of before cleaning and J_0 is the initial permeate flux.

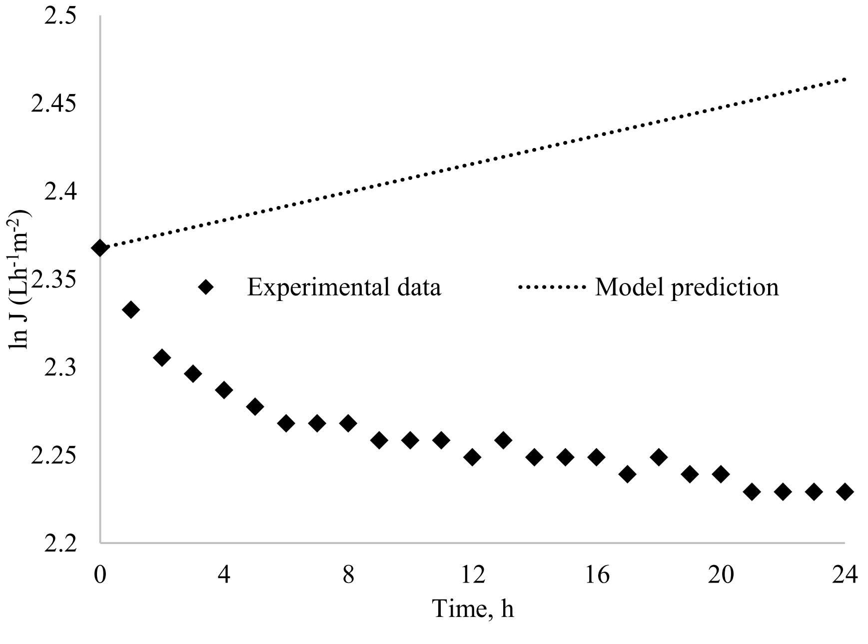
3. Results and Discussion

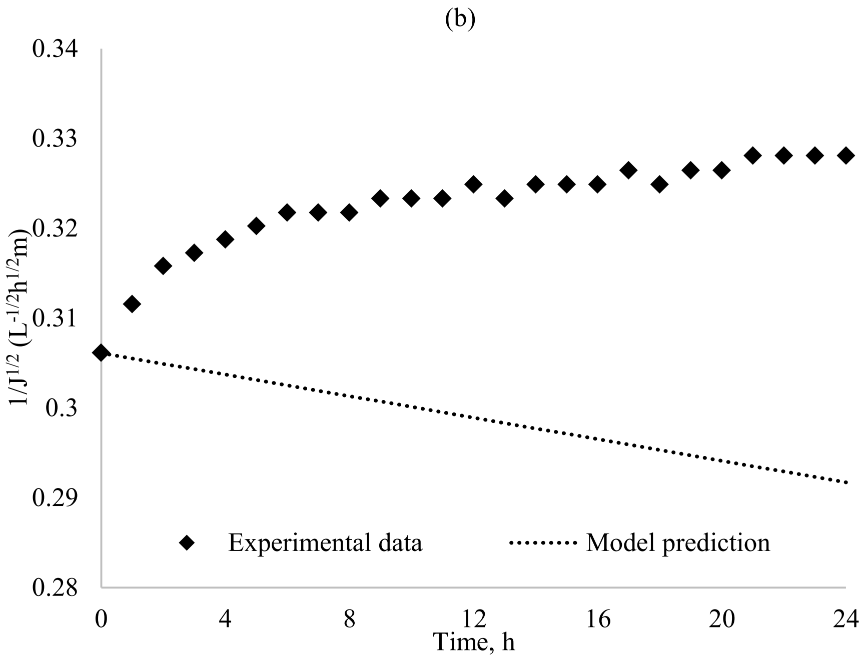
3.1. Membrane fouling study

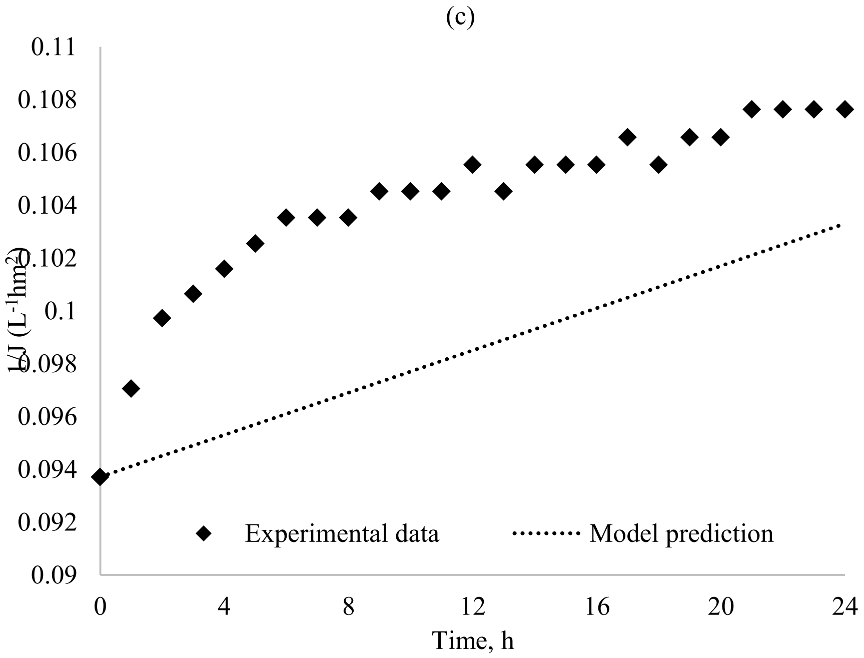
Ultrafiltration has been done using UF membrane for 24 hours at TMP 1 bar. The experimental data is recorded in Figs. 2 (a-d) with four model prediction fitting for membrane fouling study. Membrane fouling study was done in order to identify the types of fouling occurred during ultrafiltration.

For fouling mechanism analysis, four fouling models were used including complete pore blocking, intermediate pore blocking, standard pore blocking and cake filtration. Figs. 2 (a-d) shows the experimental data and four model prediction curve to fit the experimental results according to Eqs. (1)-(4).

(a)




(b)
0.34
0.33
0.32
0.31
0.3
0.29
0.28
1/J^1/2 (L^-1/2h^1/2m)
Experimental data
Model prediction
0
4
8
12
16
20
24
Time, h


(c)
0.11
0.108
0.106
0.104
0.102
0.1
0.098
0.096
0.094
0.092
0.09
1/J (L^-1hm^2)
Experimental data
Model prediction
0
4
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16
20
24
Time, h

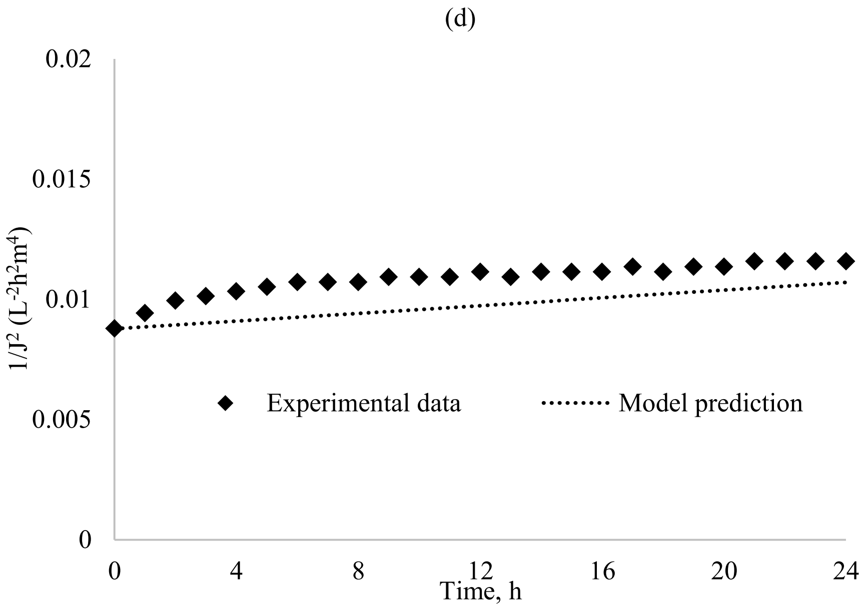

Fig. 2. Experimental Data and Fouling Model Fitting for a) Complete Pore Blocking b) Standard Pore Blocking c) Intermediate Pore Blocking d) Cake Filtration.

Complete pore blocking may occur if the sizes of particles in the raw water feed are larger than the membrane pore size thus the particles cannot pass through the membrane pores. Experimental data fitting to complete pore blocking model is shown in Fig. 2(a). Standard pore blocking involving internal fouling where the sizes of the particles are smaller than the membrane pore. Therefore the small particles can be adsorbed to the membrane pores. Figure 2(b) shows the experimental data fitting for standard pore blocking model.

For the particles which have the same molecular sizes with membrane pore size, intermediate pore blocking may occur due to the deposition of the particle entering the membrane pores. The experimental data were better fitted to the intermediate pore blocking model as shown in Fig. 2(c) compared to the complete pore blocking and standard pore blocking model. However Fig. 2(d) shows the best fitting of the experimental data with the cake filtration model. Cake filtration occurs for the particles which are unable to enter the membrane pores thus caused layers of cake and cover the membrane surface.

From the data fitting, R^2 value for each model prediction has been calculated as shown in Table 2.

Table 2. R^2 Value for Each Model Fitting.

Fouling mechanism	R^2 value
Complete pore blocking	0.7728
Intermediate pore blocking	0.7811
Standard pore blocking	0.7892
Cake filtration	0.8049

Table 2 shows the R^2 values from the data fitted for each model. All the models gave high R^2 values. However cake filtration model has the highest R^2 value approaching 1 which indicates that the layer cake formation on the membrane surface is the dominant fouling mechanism during filtration.

Cake filtration has been found as the major cause of fouling for surface water filtration. This is accordance to the findings of many researchers that surface water contains natural organic matter (NOM) which composed variety of particulate and soluble organic compound [17-18]. Leiknes et al., (2004) reported that NOM was the major foulant during drinking water treatment from surface water [19].

Previous study mentioned that there are three blocking filtration mechanisms and a cake filtration mechanism in the blocking filtration law but only one of these mechanisms is often employed to fit the filtration data for the entire range [20]. Iritani et al., (2013) reported that pore blocking was significantly produced higher contribution to the overall flux decline compared to cake filtration [21]. Vela et al., (2009) found that initial pore blocking which occur at the beginning of the ultrafiltration experiment was the main cause of the difference observed between experimental results and theoretical predictions for short time scales [22].

3.2. Membrane cleaning study

3.2.1. Physical cleaning

Backwashing has been done after 4 hours of filtration for 0.5 minute at two different pressures, 2 and 3 bars to study the effect of backwashing pressure on the flux recovery. The results of flux recovery are shown in Fig. 3.

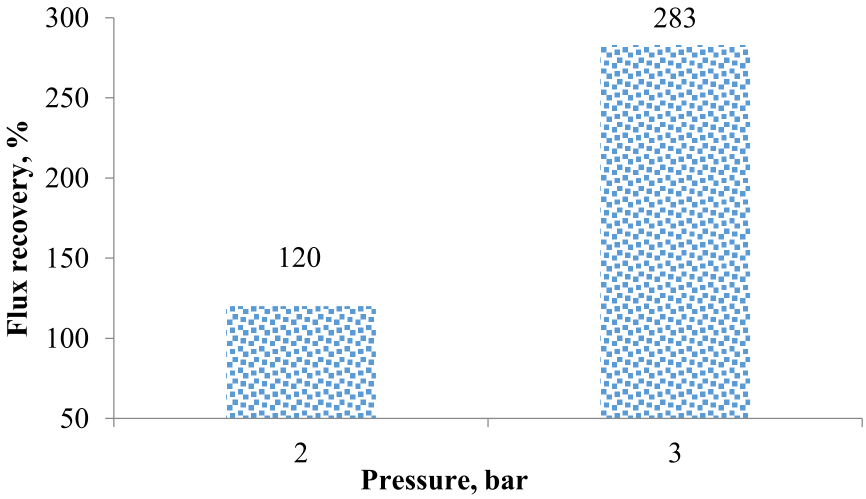

Fig. 3. Flux Recovery at Different Backwashing Pressures.

During backwashing, the pressure on the permeate side of the membrane was set higher than the pressure within the membranes thus by flushing the membrane with high pressure water will remove the foulants accumulated on the membrane surface and pores.

Introducing higher pressure during backwashing might affect the membrane pore size. Nakatsuka et al. (1996) mentioned that the backwashing need to be twice the filtration pressure. For this experiment, TMP used for the filtration was 1 bar [23]. Thus choosing 2 bar is appropriate for backwashing to avoid damage to the membrane due to higher pressure applied during backwashing. This backwash pressure will be tested on another fouled membrane with higher flux reduction. Results are shown in Fig. 4.

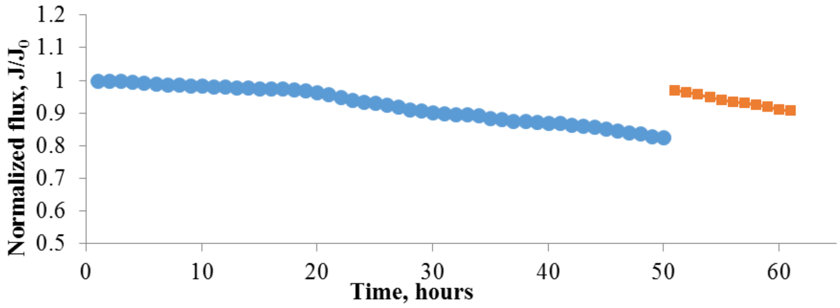

Fig. 4. Normalized Flux and Flux Recovery after 50 Hours Filtration.

Figure 5 shows the normalized flux and flux recovery after 50 hours filtration at TMP 1 bar. Flux was reduced 17.5% from its initial flux after 50 hours of filtration. After backwashing for 0.5 minute at 2 bar, the permeate flux increased from 0.82 to 0.97 from the initial flux. It shows that permeate flux can be recovered by backwashing at 2 bar for 0.5 minute at higher fouling rate.

3.2.2. Chemical cleaning

Figure 5 shows the percentage of recovery for different cleaning agents. Sodium hypochlorite (NaClO) gave the highest recovery rate of 79.3% while for acid cleaning agents, the highest percentage of recovery was obtained by using HNO_3 with 78.86%. The result shows that the oxidation reaction occurs between NaClO and foulant reduces the adhesion of fouling materials to membrane due to increasing of hydrophilicity and negative charges of the foulant. Strugholtz et al. (2005) reported that the oxidation of the aromatic humic substances contains in NOM as the foulants can be removed by an oxidizing agent such as NaClO [24]. This is also consistent with Woo et al. (2013) finding which proves that NaClO can be effectively used as the cleaning agent to remove NOM foulants from river was as feed. Besides, in the same work, they concluded that the alkaline cleaning agent is better than acidic cleaning agent [11]. Acidic cleaning agents such as nitric acid and citric acid would be very effective for removing inorganic foulants [12]. However raw water feed used is generally contains natural organic matter.

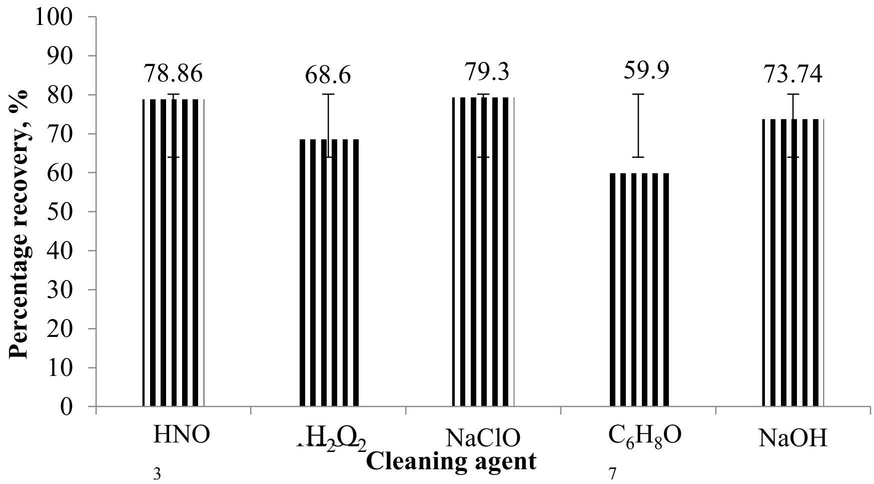

Fig. 5. Flux Recovery After 5 Minutes of Membrane Cleaning Using Different Cleaning Agents with 0.25M Concentration.

3.2.3. Comparison between physical and chemical cleaning

From both method of membrane cleaning, the recovery rate has been compared to choose the most appropriate method for membrane cleaning.

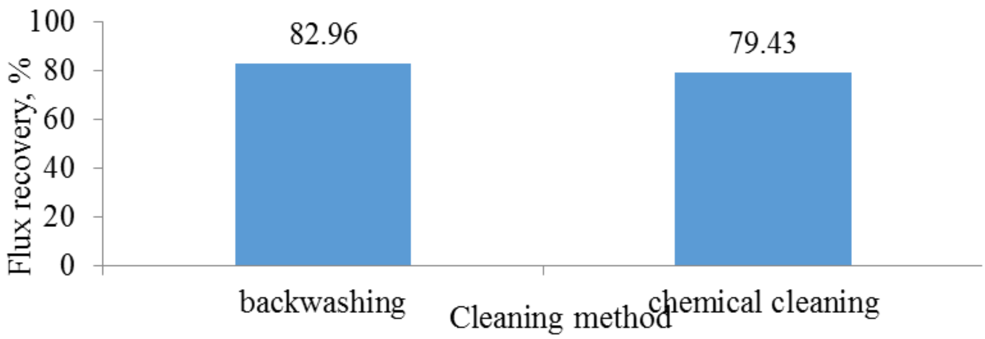

Fig. 6. Flux Recovery for Different Cleaning Method.

Figure 6 shows the percentage recovery rate comparing two membrane cleaning methods which is backwashing and chemical cleaning using 0.25M NaClO. From previous experiments, chemical cleaning using 0.25M of NaClO for 5 minutes has been chosen as the best condition for chemical cleaning while backwashing for 0.5 minute at 2 bar has been chosen based on the recovery rate after cleaning. Backwashing at higher pressure may affect the membrane pore.

From Fig. 6, backwashing method shows higher recovery percentage which is 82.96% while NaClO can only recover up to 79.43%. This might be due to the higher applied pressure during backwashing where the permeate water was flushed into the membrane at the outlet while for chemical cleaning, the membrane was immersed in the chemical. The higher pressure during backwashing cleaned the membrane pores and the membrane surface thus increased the recovery rate after cleaning. Furthermore, as a hydrophilic membrane, PAN is more easily recover permeate flux as reported by Nakatsuka et al. [23]. Thus, chemical cleaning by NaClO can be eliminated as backwashing can recover higher percentage to the initial flux. In addition, backwashing is the appropriate method for membrane cleaning as it is easier to handle. It is a cost effective method and suitable for rural area water treatment.

4. Conclusions

From the experimental data compared to model predictions data and comparison of physical and chemical cleaning on 17.5% fouled membrane, cake formation has been found as the major cause of fouling for surface water filtration. Physical cleaning by backwashing has been proved to clean the membrane based on SEM images and also recovered the permeate flux up to 82.96% compared to chemical cleaning which only recovered 79.43%.

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