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A study of operational factors for reducing the fouling of hollow fiber membranes during wastewater filtration

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Abstract

Fouling of membranes is the most important problem in wastewater filtration since the sustainability of the process is highly influenced by the fouling rate. Possible ways for fouling control are through feed water pre-treatment, operational conditions and membrane cleaning. This paper studies the operational conditions that affect the fouling of hollow fiber ultrafiltration membranes during the separation of wastewater. Specifically, several factors which are temperature, pressure, time, pH and surface area of membrane, are studied. The Design of Experiments methodology is used to investigate the effect the factors. The results of this study help to reduce the fouling of membranes, thus contributing to a more sustainable filtration system.

*Keywords:*Ultrafiltration; Hollow fiber membrane; Design of experiment methodology;

1. Introduction

Fouling of membranes is a significant issue for the efficiency of membrane filtration in wastewater treatment systems. Research on ultrafiltration membranes water treatment is hot in the field especially in production sectors. With the decrease in the price of materials, a growing number of membranes applied in domestic water treatment have received good results, but pollution is the important bottleneck that restricts the promotion of this technology.

Fouling occurs when the components filtered from the feedstream collect near the membrane/fluid interface. The earliest stage of the fouling process is characterized by concentration polarization (CP) associated with the boundary layer, in which a gradient of excluded products forms near the membrane surface [1-2]. Under some conditions, the excluded products can associate with the membrane surface or membrane pores, forming what is generically known as a fouling layer (Fig. 1). There are several types of fouling that can be divided into reversible and irreversible fouling based on the attachment strength of particles to the membrane surface. This fouling layer will affect the separation performance by decreasing flux and selectivity as well as serving as a source of pollution. Foulants at the membrane surface or within the membrane pores will exert some influence on the passage of solutes through the membrane, either because of the porosity of the cake or blocked membrane is lower than that of the clean membrane or because the charge properties of the fouled membrane are very different than those of the unfouled membrane [3-5].

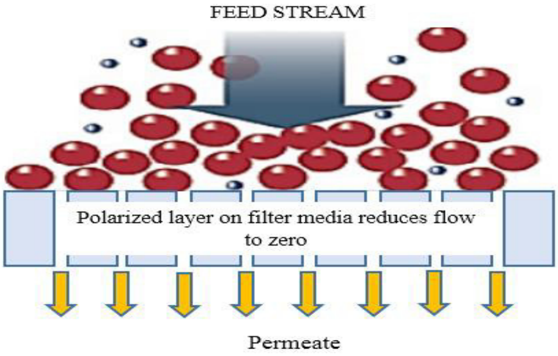

Fig. 1. Membrane fouling layer

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Generally, membrane can be defined as a barrier, to separate two phases and to restrict transportation of various chemicals in a selective manner. Meanwhile, synthetic membrane can be defined as an interphase that separates two phases and restricts the transport of various chemical species in a rather specific manner [6]. The small pores of the membranes can serve as a physical barrier, preventing the passage of certain materials such as salt, bacteria and viruses while allowing the free passage of water and air. The desalination of water using ultrafiltration is a well-known use of membranes as a filter.

Ultrafiltration is basically a size-exclusion based, pressure driven membrane separation process. Ultrafiltration membrane has pore size in the range of typically 2 to 20 nm (20 to 200A) and is capable of retaining chemical species with a molecular weight from 1000 to 1,000,000 Daltons. This membrane does have a very thin and fine porous surface layer supported by a microporous substructure. The surface layer acts as the separator, while the substructure beneath it provides the mechanical strength needed [7-9].

The loss of membrane permeability during ultrafiltration of particles (which is attributed to the adsorption or deposition of particles on the membrane) depends primarily on the interaction of the membrane with the components of wastewater solution, as well as on the properties of the material of which the membrane has been made. In addition, there are another two contributing factors that should be monitored which are the conditions of the process and the properties of the solution.

When the water to be treated contains humic substances, the adsorption or deposition of organic matters on the membranes may be affected by a variety of factors such as pH and temperature of feed solution, time of separation, pressure and surface area.

Therefore, fouling control strategies and sustainable development are very important missions for the community research and technology evaluation programme because water is an important resource for human life. Fouling control strategies are able to decrease energy demand, increase membrane lifetime and reduce other operational costs. Nowadays, modern fouling control approaches focus on changing filtration process variables including alteration of feed water quality [10-12].

The purpose of this study is to examine the factors that can reduce the fouling of hollow fiber ultrafiltration membranes during the separation of wastewater.

Nomenclature

A	pH of feed solution
B	temperature of feed solution
C	time of separation process
D	transmembrane pressure
E	surface area of membrane
FI	fouling index
J	permeate volume flux
DER	dope extrusion rate
AGL	air gap length
CBT	congulation bath temperature
BFR	bore fluid ratio
PT	post treatment

2. Experimental Work

2.1. Membranes

The experiment involved hollow fiber ultrafiltration membranes. PES hollow fiber membranes were produced based on conditions proposed by an earlier research team [13]. The best response of flux condition and the highest percentage of rejection have been chosen to determine the spinning condition.

Table 1. Setting for spinning condition.

Exp. Run	DER ($cm^3 min^{-1}$)	AGL (cm)	CBT (^0C)	BFR (NMP/H_2O)	PT (Immerse in MeOH), (h)	Flux (LMH)	Rejection (%)
1	6	2	30	70:30	6	23.11	90.01
2	6	2	18	70:30	2	5.36	92.99
3	6	2	30	0:100	2	17.05	96.18
4	6	2	18	0:100	6	5.25	97.66
5	2	0	30	0:100	2	26.36	99.95
6	6	0	30	0:100	6	28	100

From the Table 1, data no 6 has been chosen as the spinning condition because of stable flux and 100% rejection.

Membrane modules 22cm in length were prepared. Then, the fibers were carefully prepared in U-shape and threaded through the tube sheet as in Fig. 2, until about 3cm protruded from the bottom end of the silicone tube. At the other end, the fibers were placed in the end cap and sealed with polyurethane resin. The modules were left for one day in order to allow the resin to cure before being removed. The resin flush with the tube sheet end was cut with a new razor blade so that the fiber bores were exposed to permeate flow.

Fig. 2. U-shape ultrafiltration membrane module

2.2. UF process

The fouling of the ultrafiltration membranes and the resulting variation of separation properties were investigated in a pilot-scale experiment (Fig 3).

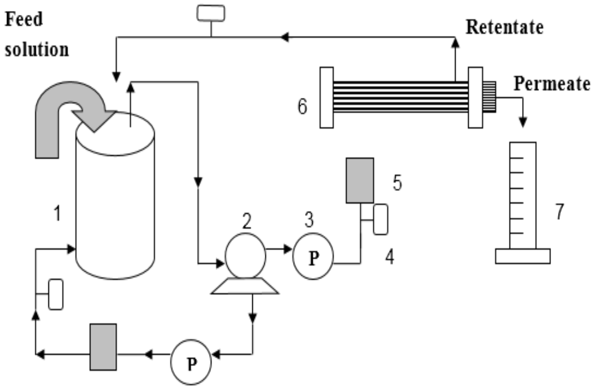

Fig 3. Schematic set-up of ultrafiltration unit. (1) Feed tank; (2) pump; (3) pressure gauge; (4) control valve; (5) flow meter; (6) hollow fiber membrane module; (7) measuring cylinder.

Before starting the process, the pH and temperature of the wastewater are varied or adjusted based on selected values. Then the wastewater will be fed into the feed tank. Transmembrane pressure is adjusted during the filtration process to push or pull the permeate through the membrane. Permeate is collected and then measured using a measurement cylinder. As for membrane, its surface area will be adjusted during the potting process by varying the number of membranes.

2.3. Feed Solution

In the experiments, cutting oil wastewater was used as a feed solution to determine the proneness of membrane fouling. The concentration of oil in the feed solution amounted to $0.25g/l^3$

0.1 N Nitric acid and 0.1 N NaOH were used to adjust the pH level.

2.4. Analytical method

The separation and transport properties of the membrane with respect to the feed solution were determined by measuring the permeate volume flux which was calculated as:

$$J = \frac{L}{At} \tag{1}$$

where J is the permeate volume flux (LMH). L denotes the volume of the permeate sample collected within time(t), and A indicates the effective membrane surface (m^2).

The proneness of the membranes to fouling was established in terms of fouling index (FI), calculated as follows:

$$FI = \frac{J_h}{J_0} \tag{2}$$

where J_h is the distilled water flux and J_0 denotes the distilled water flux across a new membrane [14].

2.5. Experimental Design Setup

In this study, in order to minimize the number of experiments but still capable of quantifying the effect of each variable, a statistical method of half factorial design was applied. The experimental plan was based on a two level, half factorial designs with resolution V and four center points for curvature evaluation. In this experiment, five variables were evaluated, each at two levels: low and high. They were pH and temperature of feed solution, time, transmembrane pressure and surface area of membrane as shown in Table 2. A total of 20 experiments were conducted and the Design Expert software was used to analyze the results.

Table 2. Parameter settings of membrane ultrafiltration process.

Level	pH	Temperature (^{0}C)	Time (min)	TMP (bar)	Surface area (m^2)
High (+)	13	45	35	3	0.074
Center point	8	35	25	2	0.058
Low (-)	3	25	15	1	0.042

3. Result and discussion

After conducting the separation process with different parameter values, the results of fouling index were obtained as provided in Table 3. Fouling index was calculated based on Eq. 2.

A fitting line is drawn through the effects that are close to zero. In this aspect, if the factors are not important, the points should be found close to the line (half normal) as shown in Fig 4.

Analysis of variance (ANOVA) tested on the variables with FI value as the response was conducted to determine the significance of each variable. The confidence interval was set as 95% for a variable to be considered significant.

The Model F-value of 24.19 implies the model is significant (see Table 4). There is only a 0.01% chance that a Model F-Value this large could occur due to noise. A p-value less than 0.0500 indicates the model terms are significant. In this case A,B, C, D, AD, BD are significant model terms. It was found that surface area of membrane is not a significant variable.

The Lack of Fit F-value of 1.10 implies the Lack of Fit is not significant relative to the pure error. There is a 52.52% chance that a Lack of Fit F-value this large could occur due to noise. A non-significant lack of fit is good because we want the model to be fit.

The model's coefficient of determination, R^2, is 0.9236 (see Table 5). The R^2 which is almost unity indicates that the model fairly approximated the fouling index data.

The fouling index regression model in terms of the coded factors (low = -1 and high = 1) as generated by the Design Expert software is:

$$FI = 0.53 + 0.15A + 0.048B + 0.10C + 0.046D + 0.081AD - 0.068BD \quad (3)$$

where A is pH of feed solution, B is temperature of feed solution, C is time of separation process, and D is transmembrane pressure.It should be noted, however, that curvature was significant in the model, which means a more powerful experimental design is required for better optimization e.g., with the use of response surface methodology. Based on Fig.5, it shows that the residual plot is normally distributed.

Table 3. Fouling index of the separation process.

pH	Temperature (^{0}C)	Time (min)	TMP (bar)	Surface area (m^2)	Flux (J_h)	Flux (across a new membrane) (J_o)	Fouling index (FI)
13	25	35	3	0.042	0.0639	0.071	0.9
13	25	35	1	0.074	0.4722	1.005	0.4699
3	45	35	1	0.074	0.1777	0.258	0.6886
13	45	15	1	0.074	0.1001	0.201	0.4979
13	45	15	3	0.042	0.0748	0.112	0.6678
8	35	25	2	0.058	0.0853	0.114	0.7484
3	25	15	1	0.074	0.0400	0.175	0.2286
13	45	35	3	0.074	0.8429	0.878	0.9600
3	25	35	3	0.074	0.1093	0.200	0.5466
8	35	25	2	0.058	0.4608	0.513	0.8982
3	45	15	3	0.074	0.1023	0.418	0.2448
8	35	25	2	0.058	1.0740	1.365	0.7868
13	25	15	3	0.074	0.1347	0.199	0.6768
13	45	35	1	0.042	0.6544	0.818	0.800
3	45	15	1	0.042	0.2988	0.732	0.4082
3	45	35	3	0.042	0.3110	0.900	0.3456
13	25	15	1	0.042	0.3417	0.799	0.4276
3	25	15	3	0.042	0.2032	0.795	0.2556
3	25	35	1	0.042	0.2707	0.780	0.3470
8	35	25	2	0.058	0.7128	0.821	0.8682

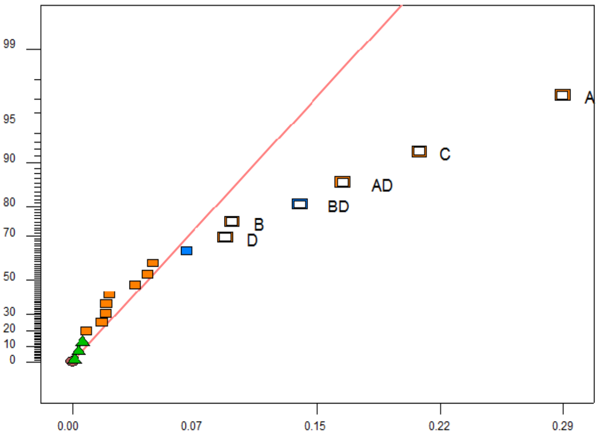

Fig. 4. Half-normal plot

Table 4. ANOVA for fouling index as the response.

Source	Sum of Squares	Degree of Freedom	Mean Square	F Value	p-value Prob > F	
Model	0.76	6	0.13	24.19	< 0.0001	significant
A	0.34	1	0.34	65.28	< 0.0001	
B	0.036	1	0.036	6.93	0.0219	
C	0.17	1	0.17	32.61	< 0.0001	
D	0.033	1	0.033	6.37	0.0267	
AD	0.10	1	0.10	19.89	0.0008	
BD	0.073	1	0.073	14.03	0.0028	
Curvature	0.28	1	0.28	53.84	<0.0001	significant
Residual	0.063	12	0.0052			
Lack of Fit	0.048	9	0.0053	1.10	0.5252	not significant
Pure Error	0.015	3	0.0049			
Cor Total	1.10	19				

Table 5. Summary for the experimental result

Std.Dev.	0.072	R-Squared	0.9236
Mean	0.59	Adj R-Squared	0.8854
C.V	12.28	Pred R-Squared	0.8385
PRESS	0.18	Adeq Precision	15.859

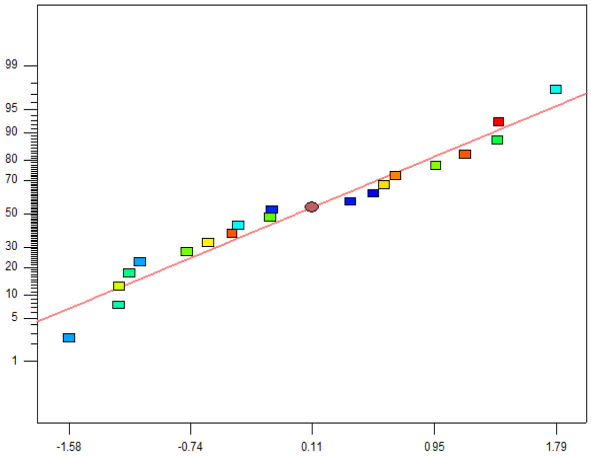

Fig. 5. Normal plot of residuals

4. Conclusion

In conclusion, the use of the design of experiments method to develop a fouling index model is proven and the derived empirical model can be subsequently used for predicting the fouling index within the separation region. The experiments show that pH and temperature of the feed solution, time of separation process and transmembrane pressure are significant factors for reducing the fouling of hollow fiber membranes during wastewater filtration. For future work, the response surface methodology will be used to get the optimum solution since the ANOVA results show the curvature is significant. The experimental results of this study can help to reduce the fouling of membranes, thus contributing to a more sustainable filtration system.

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References

[1] Chen,J., Li, Q, Elimelech M. In situ monitoring techniques for concentration polarization and fouling phenomena in membrane filtration. Advances in Colloid and Interface Sci; March 2004. p. 83-108.
[2] Bader, M.S.H. and Veentra, J.N. Analysis of concentration polarization phenomenon in ultrafiltration under turbulent flow conditions. Journal of Membrane Science 114: 1996.139-128.
[3] Schafer, A.I., Fane, A.G. and Waite, T.D.Fouling effects on rejection in the membrane filtration of natural waters. Desalination 131;2000, 215-224.
[4] Baker, R.W. Membrane Technology and Applications, England: John Wiley & Sons Ltd. 2004.
[5] S.Verlag,. Membrane SeparationProcess. Proceedings of the International Membrane Separation Processes.May 24-26. U.K: Springer-Verlag Corporation. 1989.
[6] Canas and Benavente, J. Electrochemical Characterization of an Asymmetric Nanofiltration Membrane with NaCI and KCI solution: Influence of Membrane Asymmetry on Transport Parameters. Journal of Colloid and Interface Science. 246.; 2002, 328-334.
[7] Causserand, C., Aimar, P., Vilani, C. and Zambelli, T. Study of the effects of defects in ultrafiltration membranes on the water flux and the molecular weight cut-off. Desalination 149; 2002. 485-491.
[8] Platt, S., Mauramo, M., Butylina, S. and Nystrom, M. Retention of pegs in cross-flow ultrafiltration through membranes. Desalination 149: 2002. 417-422.
[9]Pradanos, P., Arribas, J.I. and Hernandez., A. Hydraulic permeability, mass transfer and retention of PEGs in cross-flow ultrafiltration through symmetric microporous membrane. Separation Science Technology 27: 2002.2121-2142.
[10] R.H. Peiris, H. Budman, C.Moresoli, R.L.Legge. Fluorescence-based fouling prediction and optimization of a membrane filtration process for drinking water treatment. AIChE Journal 58; 2012.1475-1486.
[11] A. Seidel, M.Elimelech. Coupling between chemical and physical interactions in natural organic matter (NOM) fouling of nanofiltration membranes: implications for fouling control. Journal of Membrane Science 203; 2002. 245-255.
[12] J. Busch, W.Marquardt. Model-based control of MF/UF filtration processes:pilot plant implementation and results. Water Science and Technology 59; 2009.1713-1720.
[13] A.S.Noor Adila. Optimization of spinning condition in polyethersulfone ultrafiltration hollow fiber membrane fabrication using genetic algorithm. Internal report, Johor Bahru:Universiti Teknologi Malaysia.2013.
[14] M.K. Korbutowics, Katarzyna M.N, Tomasz W.. Analysis of membrane fouling in the treatment of water solutions containing humic acids and mineral salts. Desalination 126; 1999. 179-185.