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CLARIFICATION OF FACTORS THAT AFFECT THE FLUX PERFORMANCE OF HOLLOW FIBER MEMBRANES DURING ULTRAFILTRATION USING DESIGN OF EXPERIMENTS

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ABSTRACT

In this paper, the separation of humic substances from oily wastewater was investigated using Hollow Fiber membranes. Consideration was given to the increse of membrane permeability or flux of the Ultrafiltration process. Specifically, several factors which were temperature, pressure, time, pH and surface area of membrane, were studied. The Design of Experiments (DOE) methodology was used to investigate the effect of the factors. From the analysis of variance (ANOVA), it was determined that the pH and temperature of feed solution, time of separation process and transmembrane pressure are significant. The results of this study help to increase the permeability of membranes, thus contributing to a more sustainable filtration system.

Keywords: ultrafiltration, hollow fiber membranes, design of experiments.

INTRODUCTION

The fast changing market environment makes manufacturers have to promptly develop new products and provide high quality products to meet customers' requirements in order to maintain their competitive edge. In the process of new product development, parameter design is mostly emphasized by researchers because it is critical for practitioners to simultaneously achieve both the time-to-market reduction and the quality enhancement of products and processes. To optimize a product or process, engineers usually perform experiments to determine its parameter settings. The Design of Experiments (DOE) method has been applied to optimize parameter design in a variety of industrial applications, particularly in product development, process design and operational condition setting. The DOE method has been verified as an efficient and effective approach of designing experiments and thus is a concise and fast way of determining the optimal parameter settings of a product or process (Robert *et al*., 2003). Yong and Hahn (2007) mentioned that the systematic DOE method represents an effective and widely used tool which reduces cycle time, improves reliability, reduces process variability, and increases overall product quality. DOE is helpful in getting a rooted understanding of the fundamental processes or allowing suggestions for future experiments (Posada and Buckley, 2004).

Recently, membrane separation has become one of the most successful and advanced techniques of separation in various industries. Separation and purification processes using membrane technology are gaining popularity in many chemical and food processing sectors as well as in wastewater treatment industries (Grazyna *et al.,* 2006). The credibility of membrane technology in separating multi-component mixtures into two or more desired products has become a more desirable choice considering its abilities and advantages. The technology offers several advantages over and above the traditional techniques, including low energy requirement and low temperature operation (Sulaiman *et al*., 2001). The membrane separation and filtration processes comprise a continuum of processes designed to separate particles or solutes of different sizes by utilizating membranes with appropriately sized pores (Ohya, 1976). The processes are Microfiltration (MF), Ultrafiltration (UF), Nanofiltration (NF) and Reverse Osmosis (RO) in order of decreasing pore size. A membrane has the ability to filter a component more efficiently because of differences in physical and/or chemical properties between the membrane and the solute.

Generally, a 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 (Canas and Benavente, 2002). 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. This study focuses on ultrafiltration and its application in wastewater treatment. 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. An ultrafiltration membrane has a pore size of about 0.01-0.1 μm and thus it prevents particles, colloids, microorganisms and dissolved solids that are larger in dimension than the pores in the membrane surface from passing. 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 (Causserand *et al*., 2002; Platt *et al*., 2002; Laine *et al*., 1990; Pradanos *et al*., 2002).

Ultrafiltration processes have been widely applied to a variety of fields. More specifically, in the area

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of industrial wastewater treatment, they have been applied to tannery wastewater in order to recycle trivalent chromium (Fabiani *et al*., 1996; Shaalan *et al*., 2001), to remove colour from tannery wastewater (Alves and De Pinho, 2000), to reduce organic polluting compounds in olive-mill wastewater (Turano *et al*., 2002) and even in artificial kidney mechanisms (Serra *et al*., 1998). Therefore, the great usage of ultrafiltration in industrial operations generates the need for a useful tool for the determination of membrane performance and the minimization of operating costs

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 the 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.

Good filtration process conditions are able to decrease energy demand, increase membrane lifetime, reduce other operational costs and reduce fouled membranes during the process. (Peiris *et al*., 2012; Seidel and Elimelech, 2002; Busch and Marquardt, 2009).

The purpose of this study is to examine the factors that can affect the permeate volume flux of hollow fiber ultrafiltration membranes during the separation of wastewater by using the DOE method.

EXPERIMENT

Membranes

In the experiment, polymer pellets comprising 15.25% polyethersulfone (PES) concentration were used as the main membrane forming material, 66.43% 1-methyl-2-pyrrolidone (NMP) was used as a solvent to dissolve the polymer without further purification, 14.3% polyethylene glycol (PEG 400) was used as an additive to enhance PES membrane properties and 4.02% of water was utilized. PES hollow fiber membranes were produced based on conditions proposed by (Noor Adila, 2013) during the spinning process. The spinning condition has been chosen based on the best response for flux and rejection.

In particular, Table-1 shows some conditions that have been used during the spinning process by (Noor Adila, 2013). From the table, data no. 6 has been chosen as the spinning condition because of stable flux and 100% rejection.

Table-1. Setting for spinning condition.

Exp. Run	DER (cm^3min^{-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

Note:
DER : dope extrusion rate
AGL : air gap length
CBT : coagulation bath temperature
BFR : bore fluid ratio
PT : post-treatment time

Preparation of hollow fiber membrane modules (Potting procedure)

Membrane modules 22cm in length were potted. Then, the fibers were carefully prepared in U-shape and threaded through the tube sheet until about 3cm protruded from the bottom end of the silicone tube. Figure-1 illustrates the schematic diagram of the hollow fiber membrane module and its housing. At the bottom end, the fibers were placed in the end cap and sealed with polyurethane resin (Loctite^R E-30CL epoxy adhesive). The resin was injected via a syringe into the bottom of the silicone tube until it reached the top of the tube sheet and sealed the fibers in the tube sheet. The syringe was then removed and the silicone tube was nipped with a flow reducer or a metal clip to prevent resin leakage. The modules were left for one day in order to allow the resin to cure. 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.

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Figure-1. U-shape ultrafiltration membrane module.

Ultrafiltration process

The ultrafiltration membranes and the resulting flux were investigated in a pilot-scale experiment (Figure-2). The experiment was carried out in a cross flow filtration set up. Since the outer layer of the hollow fiber surface was the selective layer, the feed solution was pumped into the shell side of the ultrafiltration module and permeate liquid flowed out from the lumens of the fibers. A total of two bundles of fibers were potted for testing each experimental setting. Based on this, the significance of the replication error in comparison to the model dependent error can be examined.

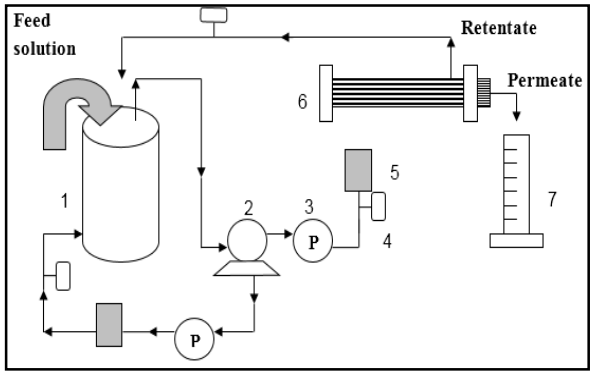

Figure-2. Schematic set-up of ultrafiltration unit. (1) Feed tank; (2) pump; (3) pressure gauge; (4) control valve; (5) flow meter; (6) hollow fiber membrane module; (7) measuring cylinder.

Before starting the process, the pH and temperature of the wastewater were varied or adjusted based on selected values. 0.1 N Nitric acid and 0.1 N NaOH were used to adjust the pH level. In this study, a feed solution of cutting oil was used to observe the separation performance of the membrane. The concentration of oil in the feed solution amounted to 0.25g/l^3. The feed solution was supplied to the hollow fiber module by using the Hydra Cell pump, while the permeate (product) solution was discharged from the permeate (product) outlet which was open to the atmosphere. The retentate was recycled to the feed tank. Transmembrane pressure was adjusted during the filtration process to push or pull the permeate through the membrane. Permeate was collected and then measured using a measurement cylinder. As for the membrane, its surface area was adjusted during the potting process by varying the number of membranes. Flux was calculated by taking the average of two readings for each condition. Thus, the average flux data were reported.

Analytical method

The permeability (flux) of membrane was calculated by the following formula:

$$J = \frac{Q}{\Delta P \times A} = \frac{Q}{n\pi Dl\Delta P} \quad (1)$$

where

J = permeability (flux)
Q = water flux reading (L/h)
ΔP = pressure difference between the feed side and the permeation side of the membrane (atm)
A = effective membrane surface area (m^2)
N = number of fibers in the module
D = outer diameter of the hollow fiber membrane (m)
l = effective length of the hollow fiber membrane (m).

Experimental design setup

In this study, 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 statistical method of half factorial design was applied in order to minimize the number of experiments. The experimental plan was based on a two-level, half factorial design with resolution V and four center points for curvature evaluation. The statistical regression technique was applied in providing an estimate of flux and in identifying the most critical parameters in controlling flux. The work addressed the development of a mathematical model for flux to describe the relationship between the independent ultrafiltration condition variables and flux during the separation process. A total of 20 experiments were conducted and the Design Expert 6.0.5 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

RESULTS AND DISCUSSIONS

The results for flux were obtained as provided in Table-3.

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Table-3. Results of the separation process.

pH	Temperature	Time	TMP	Surface area	Flux
	(^{0}C)	(min)	(bar)	(m^2)	(LMH)
13	25	35	3	0.042	621
13	25	35	1	0.074	277
3	45	35	1	0.074	372
13	45	15	1	0.074	288
13	45	15	3	0.042	509
8	35	25	2	0.058	828
3	25	15	1	0.074	65
13	45	35	3	0.074	1300
3	25	35	3	0.074	294
8	35	25	2	0.058	800
3	45	15	3	0.074	570
8	35	25	2	0.058	850
13	25	15	3	0.074	865
13	45	35	1	0.042	442
3	45	15	1	0.042	378
3	45	35	3	0.042	602
13	25	15	1	0.042	311
3	25	15	3	0.042	545
3	25	35	1	0.042	477
8	35	25	2	0.058	828

Analysis of variance (ANOVA) tested on the variables with flux 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 75.04 in Table-4 implies that the model is significant. There is less than a 0.01% chance that a Model F-Value this large could occur due to noise. The P-value for the model is less than 0.05, which indicates that the model is significant. This is desirable because it indicates that the terms in the model have a significant effect on flux. In this case A,B, C, D, AC, AD, AE, BC, BE, and DE are significant model terms. Values greater than 0.10 indicate the model terms are not significant. The Curvature F-value of 243.64 implies that there is a significant curvature (as measured by the difference between the average of the center points and the average of the factorial points) in the design space. The Lack of Fit F-value of 5.28 implies that the Lack of Fit is not significant relative to the pure error. There is a 10.15% 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.

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Table-4. ANOVA for flux as the response.

Source	Sum of Squares	Degree of Freedom	Mean Square	F Value	p-value Prob > F	
Model	$1.193*10^6$	11	$1.085*10^5$	75.04	< 0.0001	Significant
A	$1.073*10^5$	1	$1.073*10^5$	74.20	< 0.0001	
B	63252.25	1	63252.25	43.76	0.0003	
C	45582.25	1	45582.25	31.53	0.0008	
D	$4.543*10^5$	1	$4.543*10^5$	314.27	< 0.0001	
E	1332.25	1	1332.25	0.92	0.3690	
AC	14400.00	1	14400.00	9.96	0.0160	
AD	98910.25	1	98910.25	68.43	< 0.0001	
AE	$1.498*10^5$	1	$1.498*10^5$	103.61	< 0.0001	
BC	73984.00	1	73984.00	51.18	0.0002	
BE	69169.00	1	69169.00	47.85	0.0002	
DE	$1.153*10^5$	1	$1.153*10^5$	79.74	< 0.0001	
Curvature	$3.522*10^5$	1	$3.522*10^5$	243.64	< 0.0001	Significant
Residual	10118.50	7	1445.50			
Lack of Fit	8859.50	4	2214.88	5.28	0.1015	Not significant
Pure Error	1259.00	3	419.67			
Cor Total	$1.555*10^6$	19				

Based on Table-5, it shows that the model's coefficient of determination, R^2, is 0.9916. The R^2 which is almost unity indicates that the model fairly predicts the flux data. The Pred R-Squared of 0.9074 is in reasonable agreement with the Adj R-Squared of 0.9784. Adeq Precision measures the signal to noise ratio. A ratio greater than 4 is desirable. The ratio of 40.062 indicates an adequate signal. The flux regression model in terms of the coded factors (low = -1 and high = 1) as generated by the Design Expert software is:

$$J = 494.74 + 81.87A + 62.87B + 53.37C + 168.50D + 9.13E + 30.00AC + 78.63AD + 96.75AE + 68.00BC + 65.75BE + 84.88DE \quad (2)$$

where A is pH of feed solution, B is temperature of feed solution, C is time of separation process, D is transmembrane pressure and E is surface area of membrane. It should be noted, however, that curvature is 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. 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 plot) as shown in Figure-3. Based on Figure-4, it shows that the residual plot is normally distributed.

Table-5. Summary for the experimental results.

Std.Dev.	38.02	R-Squared	0.9916
Mean	561.10	Adj R-Squared	0.9784
C.V	6.78	Pred R-Squared	0.9074
PRESS	$1.44*10^5$	Adeq Precision	40.062

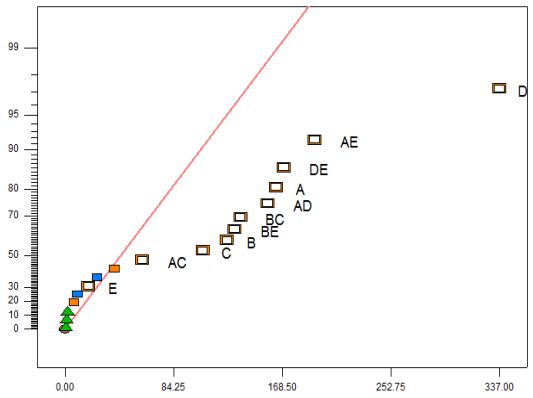

Figure-3. Half-normal plot of residuals for flux.

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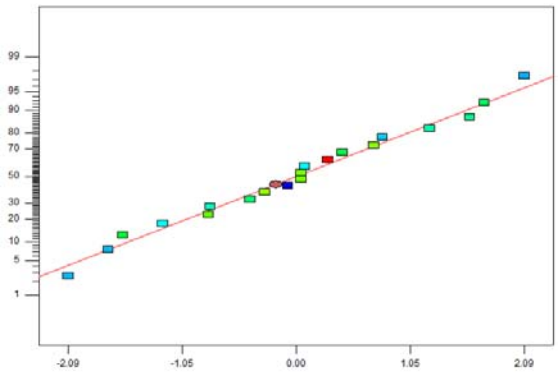

Figure-4. Normal probability plot of residuals for flux.

CONCLUSIONS

In conclusion, the use of the statistical DOE method to develop a flux model for hollow fiber membranes during the ultrafiltration process has been illustrated in this study. The derived empirical model can be subsequently used for predicting the flux value within the separation region. A half fractional factorial experiment was conducted and the results based on ANOVA show that the pH and temperature of feed solution, time of separation process and transmembrane pressure are significant factors on the flux performance of hollow fiber membranes. 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 maximize the flux performance of hollow fiber membranes, thus contributing to a more sustainable filtration system.

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