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Ultrafiltration of Simulated Oily Wastewater Using the Method of Taguchi

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Abstract— A study on the treatment of simulated oily wastewater for that generated from the process of fuel oil treatment at the gas turbine power plant was performed. The feasibility of using hollow fiber ultrafiltration (UF) membrane in a pilot plant was investigated. Three different variables: pressure (0.5, 1, 1.5 and 2 bars), oil content (10, 20, 30 and 40 ppm), and temperature (15, 20, 30 and 40 °C) were manipulated with the help of Taguchi method. Analysis of variable (ANOVA) and optimum condition was investigated. The study shows that pressure has the greatest impact on the flux of UF process. It was noticed that more than 96% oil removal can be achieved and flux of 624 L/m2.hr by UF process and that the fouling mechanism of UF process follows the cake/gel layer filtration model. Process optimization was conducted with a confirmation test. It was concluded that the observed values are within ±5% of that the predicted which reflects a strong representative model.

Index Terms— ANOVA, Hollow Fiber, Membrane, Oily Wastewater, Taguchi, Ultrafiltration

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1 INTRODUCTION

A variety of industrial sources generates large amounts of wastewaters daily. An important fraction of these are the oil in water (O/W) emulsions for which current treatment technologies are often costly and ineffective [1]. Oily wastewaters are produced by various processes and plants such as oil refineries, petrochemical plants, and metalworking plants. These wastewaters create a major ecological problem throughout the world [2]. Another source of oily wastewater is the effluent of gas turbine power plants running on crude oil at which the main source of oily wastewater is the fuel treatment process [3]. Oil in water can exist as free, dispersed, emulsified and dissolved oil. The first two forms can be separated from wastewater by simple physical processes. However, emulsified or dissolved oil is more difficult to remove [4]. Conventional oily wastewater treatment methods include gravity separation and skimming, dissolved air flotation, de-emulsification, coagulation, and flocculation. These methods have several disadvantages such as low efficiency, high operation costs, corrosion and recontamination problems [5]. With the remarkable development in membrane filtration technology these processes now exist as an efficient aid that may have all the features required by the industrial standards and environmental regulations. Hence, it is increasingly being applied for treating wastewater from different sources. Membranes have several advantages that made it applicable across a wide range of industries, such advantage like the quality of treated water (permeate) is more uniform regardless of influent variations, no chemicals are needed and the possibility for in-process recycling [6]. Membrane filtration has been proven effective in treating oily water in different industries including municipal wastewater [7], [8], engine rooms [2] and industrial wastewater [9], [10]. It was also studied in many oily wastewater treatment types of research [11], [12]. Ultrafiltration (UF) processes have been introduced as solution for oily wastewater treatment in many studies [1-2], [9], [13-14]. However, UF fails when it comes to molecular rejection, i.e. salt. This is where NF and RO can be useful [15-17]. Therefore integrating UF with other type of membrane may open doors for efficient oily wastewater treatment and water reuse [18-20].

2 TAGUCHI METHOD

The conventional technique of studying the effect of multiple factors on the membrane-integrated processes may alter high cost due to a large number of runs and time besides the difficulties of interpretation of these results [21]. In such case, Taguchi approach can be applied with confined knowledge of statistics to reduce the number of runs. Hence, it was highly adopted and gained wide popularity in engineering application [22] and used in many studies related to wastewater treatment, [13], [15], [23]. Taguchi approach can be applied with confined knowledge of statistics hence, got high adaptability and gained wide popularity in engineering application [22], and used in many studies related to wastewater treatment, [13], [15], [23]. The main steps for the experimental design in Taguchi method are (1) determination the objective function, (2) identifying the control factors, (3) selection the orthogonal array (OA), (4) running the experiment, (5) analysis of the data and (6) model confirmation, [21]. Taguchi method utilizes a statistical measurement of performance known as signal-to-noise (S/N) ratio, in which signal represents the desirable value while noise represents the undesirable value. There are many different possible S/N ratios, however, two of them are applicable in the present experiments: larger is better (LTB) and small is better (STB) [22]. In this study, the larger is better (Equation 1) is the flux and TDS rejection while the smaller is better for the fouling resistant (Equation 2).

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$$\left(\frac{S}{N}\right)_{LTB} = -10\log\left[\frac{1}{n}\sum_{i=1}^{n}\frac{1}{y_i^2}\right] \quad (1)$$

$$\left(\frac{S}{N}\right)_{STB} = -10\log\left[\frac{1}{n}\sum_{i=1}^{n}y_i^2\right] \quad (2)$$

Where S is the signal, N is the noise, n is the repetition number of each experiment with the same conditions, yi is the response of experiment

3 FOULING RESISTANCE AND FILTRATION MODEL

Permeate flux and fouling resistance are key factors for UF and NF process evaluation. Flux shows the amount of permeate rate. Fouling resistance shows the significance of cake/gel layer on the membrane surface and its effect on flux decline. Fouling resistance (Rf) was calculated as following [24]:

$$R_f = \frac{TMP}{\mu}\left(\frac{1}{J_{ww}} - \frac{1}{J_{wi}}\right) \quad (3)$$

Where: TMP: is the transmembrane pressure, μ is the water viscosity, Jwi is the initial water flux, Jww is the water flux after fouling. Membrane physical structure has an important influence on flux. If the pores are larger than the size of oil droplets, these droplets may enter the pores causing irreversible fouling. When the membrane pores are smaller than the droplets in the feed, these particles/oil droplets accumulate over the membrane surface causing the formation of a cake/gel layer. During membrane filtration, the degree of fouling depends on upon three main factors: 1) Operation factors 2) feed properties and 3) membrane properties. the operational parameters are such an important factors in deciding the rate of membrane fouling, in particular, increasing pressure enhances formation of the cake/gel layer of higher density and finally, leads to complete pore blocking [25].

Most models of membrane fouling correlate the permeate flux with time in terms of a quadratic and/or exponential relationship by assuming pore blockage, adsorption, gel-polarization, and bio-fouling [10]. The filtration models are listed in Table 1.

TABLE 1 EQUATIONS OF FILTRATION MODELS

Filtration Model	Fouling Mechanism	Ref.
$Ln(J) = Ln(J_0) - K_b t$	Complete pore blocking	[26]
$1/J^{1/2} = 1/J_0^{1/2} - K_s t$	Standard pore blocking	[27]
$1/J = 1/J_0 - K_i t$	Intermediate pore blocking	[28]
$1/J^2 = 1/J_0^2 - K_c t$	Cake filtration	[29]

The standard blocking mechanism occurs when the oil droplets are smaller than that of the membrane pores which leads to an internal pore blocking. The complete blocking mechanism occurs when the oil droplets size is greater than that of the membrane pores. As results, particles/oil droplets do not enter into the membrane pores and do not permeate through the membrane. The Intermediate blocking mechanism occurs when the size of oil droplets is similar to that of membrane pores leading to the Membrane pores to be blocked near their entrances on the feed side. The cake formation mechanism occurs when the size of oil droplets is much greater than the pore size; hence they are unable to enter the membrane pores. Factors affecting this type of mechanism are oil droplets deformation, cake compression, and cake/gel layer thickness.

4 EXPERIMENTAL WORK

4.1 Wastewater Feed

Oily wastewater feed used in this experiment was prepared using untreated crude and reverse osmosis permeates water. The mixture was then agitated for one minute using 10,000 rpm homogenizer type Ultra Turrax T46/6 by Janke and Kunkel KG. An emulsifier with hypophilic-lipophilic balance (HLB) value of 7 was added as a 1% as weight percentage to the untreated crude to ensure emulsion stabilization, the emulsifier is a proper quantities mix of Tween 85 and Span 80 both by Thomas Baker, the selection of desired HLB value and the weight percentage was based on some experiments done to evaluate the emulsion stability. It was noticed that with the above-selected conditions the emulsion can still stable for more than two weeks of observation. TDS value was controlled using lab grade NaCl by Sigma-Aldrich.

Membrane System

Fig.(1) shows a schematic view of the experiment setup. The system consists of one PVC type hollow fiber UF membrane with molecular weight cutoff of 50K Dalton and surface area of 2 m2. The UF membrane model BN-90 and was supplied by Guangzhou Chunke Environmental Technology Co. Ltd. from China. A 100-liter glass tank was used as feed tank. A centrifugal pump type PKm 90 by Pedrollo Co. was used as UF feed pump. Pressure gauges are installed at the module inlet and rejection stream, flow meters used to measure permeate and rejection flow rate, throttle valve used at the rejection stream to control the pressure. Three control factors were chosen in this work: temperature, pressure, and oil concentration, while the time and TDS were kept constant at 30 minutes and 150 ppm, the factors and their levels are shown in the table (2). The chosen of the above operation condition was based on real wastewater collected from gas turbine power plant's wastewater treatment facility where its oil contents are 39 ppm, TDS is 150 ppm. The Taguchi design of Experiment (DOE) was used and an orthogonal array of 16 runs (L16) was selected as the least number of experiments can be performed to evaluate the effects of above different factors in the UF process. Flux and removal efficiency were evaluated as in Eq. 4 (flux calculations) and Eq.5 (removal efficiency):

$$J = \frac{Q_p}{A_m} \quad (4)$$

$$\text{Removal \%} = \frac{C_i - C_p}{C_i} \times 100 \quad (5)$$

Where, J = flux, (L/hr.m^2), Qp = Permeate flow rate (L/hr) and A_m = surface area of membrane (m^2), C_i and C_p are initial and permeate concentration of the property respectively.

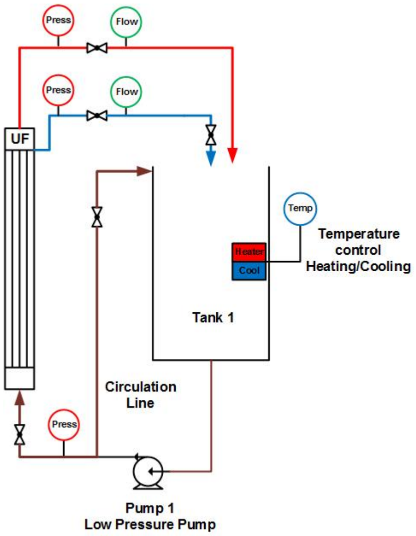

FIG. 1 EXPERIMENT SETUP

TABLE 2 FACTORS USED WITH THEIR LEVELS

Level / Parameter	1	2	3	4
Temp (ºC)	15	20	30	40
P (bar)	0.5	1	1.5	2
TDS (ppm)	150	150	150	150
Oil (ppm)	10	20	30	40

5 RESULTS AND DISCUSSIONS

Table (3) represents the experimental results for UF process. It was found that oil removal for UF process exceeds the 96% for all the experimental runs, hence it was not considered as a response and was not included in the optimization process. Fig. 2 represent the main effect plot for S/N ratio using the "larger is better ". The figure indicates that maximizing pressure and temperature will increase the S/N ratio.

Fig. 3 represents the effect of temperature and pressure on oil removal. It was found that higher pressure will lead to lower oil removal; this may be attributed to the fact that the increase in pressure may deform the oil droplet and push it through the pores. The temperature effect on oil removal is increasing at elevated pressure. For example, the increase in temperature from 20 to 30 ºC will decrease the oil removal by 0.2% and 2% at a pressure of 0.5 and 2 bars respectively. The negative effect of temperature on the oil removal is due to the pore opening and reduction in oil viscosity.

Fig. 4 represents the Flux at different temperature and oil values. The figure indicates that the oil content decreases the flux linearly. The figure also indicates that the increase in oil concentration will decrease the percentage increase of flux with temperature. For example, the increase in temperature from 20 to 30 ºC will increase the flux by 7% when the oil contents are 10 ppm, however, the increase will only be 1.7% when the oil concentration is 30 ppm. This is a result of the cake layer formation which is higher when the oil concentration is high.

TABLE 3 RESULTS OF UF PROCESS EXPERIMENTS

T ºC	Oil ppm	P bar	Flux LMH	Oil %	Turb.%
15	10	0.5	118.3	99.8	95.1
15	20	1.0	224.3	99.6	95.0
15	30	1.5	272.8	99.5	95.4
15	40	2.0	345.9	99.6	95.9
20	20	0.5	124.9	99.7	95.4
20	10	1.0	254.8	99.6	95.1
20	40	1.5	298.4	99.4	96.7
20	30	2.0	383.8	99.0	94.7
30	30	0.5	136.3	99.7	96.6
30	40	1.0	249.1	99.2	97.5
30	10	1.5	422.7	98.5	95.0
30	20	2.0	541.3	97.3	94.2
40	40	0.5	153.1	99.5	98.3
40	30	1.0	284.9	98.4	96.0
40	20	1.5	431.1	97.6	94.5
40	10	2.0	618.3	96.0	95.0

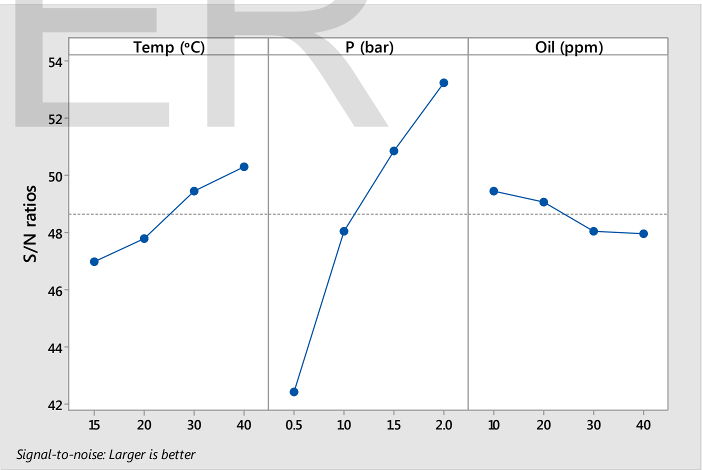

FIG. 2 S/N RATIO FOR FLUX OF UF PROCESS

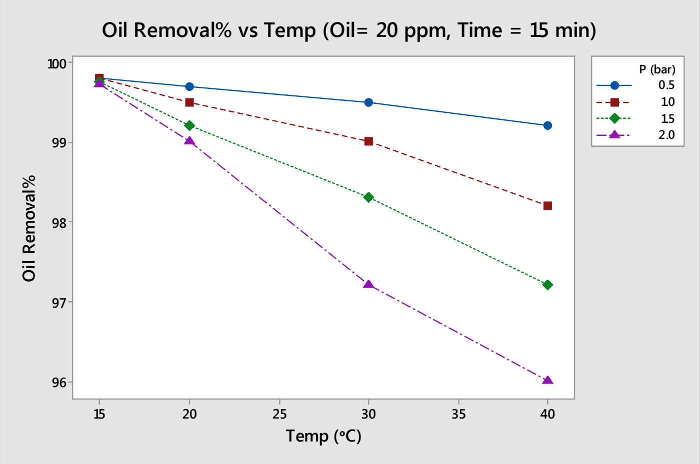

FIG. 3 EFFECT OF TEMPERATURE AND PRESSURE ON OIL REMOVAL

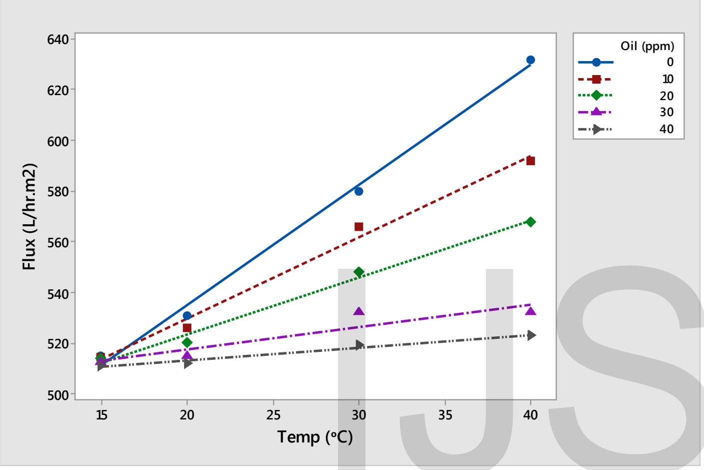

FIG. 4 EFFECT OF TEMPERATURE ON FLUX OF UF PROCESS AT DIFFERENT OIL CONTENT (P=2 BAR)

Analysis of variables was conducted for the flux data. The results are represented in Table 4. The adequacy of the suggested model can be predicted from the residual plots of Fig. 5. The ANOVA analysis suggests that the greatest contribution to the flux comes from the pressure and that P-value assumes all the model parameters are significant. The model presented has an R^2 of 99.9%.

TABLE 4 ANOVA OF UF EXPERIMENT AND PREDICTION MODEL FOR FLUX

Source	DF	Seq SS	Contribution	Adj SS	Adj MS	F-Value	P-Value
Temp (°C)	1	17723	1.19%	849.6	849.6	48.76	0.000
P (bar)	1	1465218	98.27%	34784.8	34784.8	1996.42	0.000
Oil (ppm)	1	3839	0.26%	911.5	911.5	52.32	0.000
Temp (°C)*Temp (°C)	1	112	0.01%	111.5	111.5	6.40	0.014
P (bar)*P (bar)	1	76	0.01%	75.7	75.7	4.34	0.042
Temp (°C)*P (bar)	1	645	0.04%	644.9	644.9	37.01	0.000
Temp (°C)*Oil (ppm)	1	1774	0.12%	1774.4	1774.4	101.84	0.000
P (bar)*Oil (ppm)	1	664	0.04%	664.1	664.1	38.11	0.000
Error	55	958	0.06%	958.3	17.4		
Total	63	1491009	100.00%				
Flux (L/m².hr) = -82.71 + 3.342 Temp (°C) + 278.90 P (bar) + 1.239 Oil (ppm) - 0.02033 Temp (°C)*Temp (°C) - 4.35 P (bar)*P (bar) + 0.5914 Temp (°C)*P (bar) - 0.04905 Temp (°C)*Oil (ppm) - 0.5154 P (bar)*Oil (ppm)							

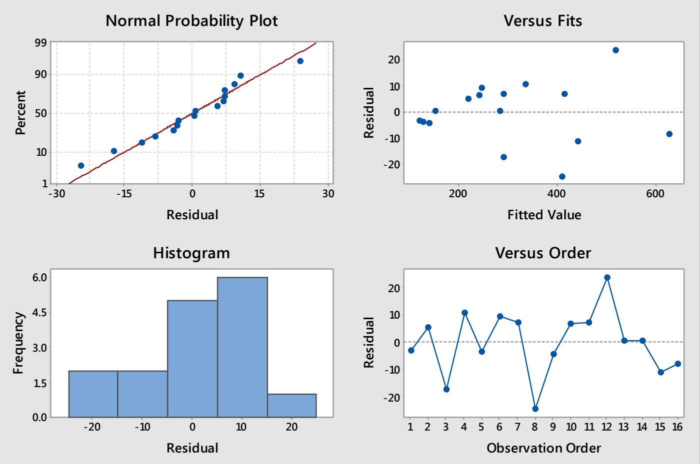

FIG. 5 RESIDUAL PLOTS FOR FLUX OF UF PROCESS

The flux from experimental runs of temperature equal to 30 °C, pressure of 1 bar and oil of 20 ppm were used to evaluate the fouling mechanism. Fig. 6 shows the flux decline with time. Fig. 7 shows different forms of flux with time. The figure indicates that the Cake filtration model is the best fits the experimental runs.

An optimization process was performed utilizing Minitab 17 software on UF process results. The aim of this process was to increase flux and reduce the fouling resistance. The optimum operation conditions are listed in Table 5. A confirmation experiment was conducted and the observed vs. the predicted values are shown in Table 6. The table shows that the deviation from the prediction is less than 2% which reflects a strongly proposed model.

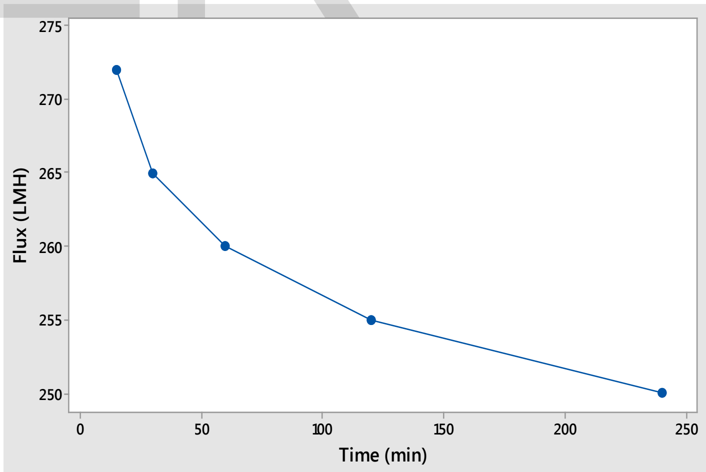

FIG. 6 FLUX DECLINE OF UF PROCESS VS TIME

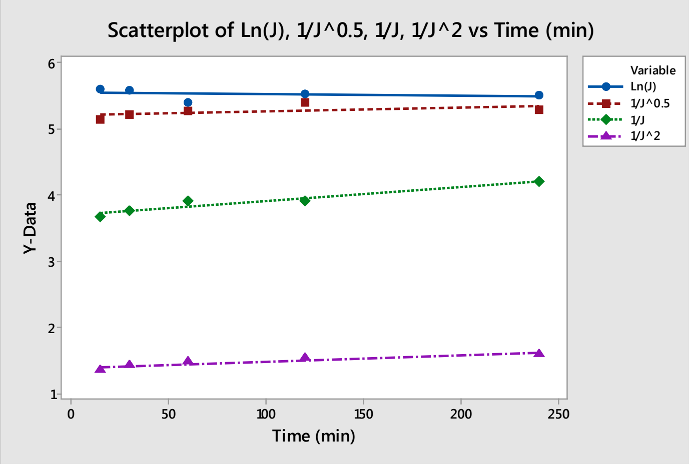

FIG. 7 DIFFERENT FORMS OF FLUX FOR UF PROCESS VS. TIME

TABLE 5 OPTIMUM OPERATION CONDITIONS FOR UF PROCESS

Variable	Setting
Temp (°C)	40
P (bar)	1.97
Oil (ppm)	10
Predicted Flux (L/hr.m2)	618.3

TABLE 6 PREDICTED VS OBSERVED RESULTS FOR UF PROCESS CONFIRMATION TEST

Parameters	Observed value	Deviation
Flux (L/hr.m2)	624.6	1.2%
Oil Removal%	96.6	-

6 CONCLUSIONS

In this study, Taguchi design of experiments (L_{16}) was employed to analyze the different parameters contribution on the simulated oily wastewater treatment using a Hollow fibers UF membrane. According to the ANOVA analysis, the most important parameter for maximum permeates flux for UF process was the pressure. Process optimization was conducted using statistical software. Optimum conditions for UF process were pressure= 2 bar, temperature= 40 °C, and oil =10 ppm. The results showed an oil removal of 96.6% with a flux of 624.6 L/m2.hr can be achieved with a deviation of less than 2% than the theoretical value.

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