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Optimal operating conditions of micro- and ultra-filtration systems for produced-water purification: Taguchi method and economic investigation

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ABSTRACT

In this study, treatment of two real produced waters was studied using microfiltration (MF) and ultrafiltration. In the first step, the effects of operating parameters including temperature, transmembrane pressure (TMP), cross-flow velocity (CFV), and back pulse time (BPT) (in the MF system) at three levels on the amount of permeate flux were investigated using ceramic and polymeric membranes. To design the experiments and optimize the experimental results, the L_9 (3^4) and L_9 (3^3) orthogonal arrays of the Taguchi method and a response category of the-larger-the-better were applied. Analysis of variance was used to determine the most important parameters affecting the permeate flux. The optimum conditions were found at the third level of temperature, TMP, and CFV and at the first level of BPT in the MF system. In the second step, the performance of ceramic and polymeric membranes was studied under the optimum conditions and 99% oil and turbidity and 100% total suspended solids rejection were obtained. Moreover, the range of total organic components removal was 63–77%. Finally, the management cost of produced-water treatment by the MF system was estimated as $0.304/bbl (~$0.002/L).

Keywords: Economic investigation; Membrane; Microfiltration; Produced water; Taguchi; Ultrafiltration

1. Introduction

Oily wastewaters and oil–water emulsions are the most important pollutants in the environment [1]. The increasing production of wastewater, especially oily wastewater, in both residential and industrial areas (such as refinery plants and production units associated with the increasing strictness of environmental regulations) calls for the development of new and more-efficient techniques for disposal or reuse of wastewater [2]. In these industries, the largest single wastewater stream is the produced water [3]. Produced water is the water trapped in underground formations that is brought to the surface along with oil or gas [4]. Traditional techniques used to separate oil-in-water emulsions, such as centrifugation, gravity settlement, and air flotation, have some operational difficulties and do not result in the desired purities [5]. Indeed, none of these conventional methods, which are based on physical and chemical principles,

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can give an absolute guarantee in terms of separation efficiency and effluent quality.

Cheryan and Rajagopalan [6] showed that the conventional treatment methods are inefficient, expensive, and ineffective. Membrane systems can compete with more-complex treatment technologies for treating water with high oil content, low mean particle size, and flow rates greater than 150 m^3/h; consequently, it is appropriate for medium and large offshore platforms [7].

As a result, membrane-based physical separation has in recent years been considered a promising technology [8]. Membrane filtration processes are used in different produced-water treatments because of their high removal efficiency, easy operation, and lower costs [9]. However, they have disadvantages such as fouling and concentration polarization, which result in declines in outflowing permeate flux [10]. Investigation of fouling is worthwhile because fouling causes dramatic flux reductions during operation, hinders selectivity, increases the operational cost, and requires frequent membrane replacement. Therefore, knowledge about the effect of operation conditions on membrane fouling is essential [11]. Decreasing concentration polarization and membrane fouling has become the main issue of many studies. A review of the literature revealed that a number of studies have been conducted to control membrane fouling, including optimization of operating conditions such as oil concentration, temperature, and transmembrane pressure (TMP) [12].

Several researchers have examined the effectiveness of membrane processes in treating oily wastewaters. Microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO) as different aspects of membrane filtration and optimized processes have been studied. Mueller et al. [13] treated oily water synthetic solutions containing different concentrations (250–1,000 ppm) of heavy crude oil droplets of 1–10 mm diameter using polymeric and ceramic membranes. Both membranes produced high-quality permeates (<6 ppm). Salahi and Mohammadi [14] used a polymeric membrane in an UF system for treating real oily wastewater and obtained 98 and 85% removal for oil and grease content and total organic components (TOC), respectively. Ebrahimi et al. [15] reported 95% oil reduction after UF of a metal-industry emulsion.

Studies show that there is still an enormous interest in membrane filtration. Good studies are available, although they are limited in terms of the variability of the factors they examine. Membrane filtration is a complex process, and disregarding the interaction of factors may result in erroneous conclusions. While a reliable solution to this problem would be to consider all the possible combinations of the levels of factors, this wastes time and is expensive. An alternative is to use a design-of-experiment (DOE) method that requires a smaller number of experiments to achieve similar results.

In the current study, the experiments were conducted using the Taguchi experimental design. A commonly applied statistical treatment, analysis of variance (ANOVA), was also used to analyze the results of experiments and to determine how much variation each factor contributes. By studying the main effects of each factor, the general characteristics of the influencing factors can be determined [14]. Table 1 lists some studies that apply the Taguchi method to find optimum conditions and rank the effects of the controlling parameters on the response parameters.

In this study, the influences of some operating conditions on the permeate flux in MF and UF treatment of two real produced waters were studied. For the MF system, three levels for each of four factors—temperature, TMP, cross-flow velocity (CFV), and back pulse time (BPT)—were chosen for the experiments. For the UF system, three levels for each of three controlling parameters—temperature, TMP, and CFV—were selected for the experiments. The objective was to find a combination of levels that resulted in optimum conditions for the highest permeate flux using the Taguchi method. Furthermore, to determine the performance of MF and UF membranes, long-term experiments were done to remove oil and grease, total suspended solids (TSS), TOC, and turbidity under optimum conditions to meet the expectations of two of the desalination units operating in Iran. Finally, a brief review of an economic study of the membrane filtration pilots was presented and the management cost of wastewater treatment by the MF system was estimated.

2. Materials and methods

2.1. Membrane

In this study, the MF module was tubular with stainless steel housing and a ceramic membrane (α-Alumina) with a mean pore size of 0.2 μm (provided by Foshan Co., China). A rectangular flat-sheet polymeric membrane (PAN350, purchased from Sepro Co., USA) was used in the UF system. This membrane is highly hydrophilic. The technical specifications of these membranes are summarized in Table 2. For all experiments in the UF system, new flat-sheet PAN-20 kDa membrane was used. However, in the MF system, the membrane was chemically cleaned after each test.

Table 1
Some studies in which the Taguchi method has been used to find the optimum conditions and relative effect of controlling parameters

Refs.	System/ Membrane type/Feed	Controlling parameters (levels)/Number of levels	Most important factors (in order)	Optimum conditions
[9]	NF/Polymeric/ CIP waste water	pH (4, 7 and 10), temperature (25, 35, and 45°C), and transmembrane pressure (12, 16 and 20 bar)/Three	On flux decline: transmembrane pressure > temperature > pH	Temperature, transmembrane pressure: first level pH: second level
[14]	UF/Polymeric/ Real oily wastewater	Transmembrane pressure (0.5, 1.5, 3 and 4.5 bar), temperature (25, 30, 40, and 50°C), cross-flow velocity (0, 0.5, 1, and 1.5 m/s), pH (4, 6, 8, and 10), and salt concentration (0, 25, 50, and 200 g/L)/Four	Cross-flow velocity > transmembrane pressure > temperature > pH > salt concentration	Transmembrane pressure, temperature, cross-flow velocity, pH: third level Salt concentration: second level
[16]	UF/Polymeric/ Real produced water	Transmembrane pressure (1.5, 3, and 5 bar), temperature (25, 40, and 55°C), and cross-flow velocity (0.5, 0.75, and 1 m/s)/Three	Temperature > transmembrane pressure > cross-flow velocity	Transmembrane pressure: first level Temperature: second level Cross-flow velocity: third level
[17]	MF/Polymeric/ Synthetic oily wastewater	Pressure (0.25, 0.5, and 0.75 bar), temperature (30, 40, and 50°C), feed flow rate (30, 40, and 50 L/h)/Three	Feed flow rate > pressure > temperature	–
[18]	NF/Polymeric/ Yeast industrial wastewater	PVA concentration (5, 10, 15, and 20 g/l), Glutaraldehyde concentration (10, 30, 50, and 70 ml/l), and TiO_2 nanoparticle concentration (0.5, 1, 2, and 5 g/l)/Four	On permeate flux: PVA concentration > TiO_2 concentration > GA concentration On COD rejection: PVA concentration > TiO_2 concentration > GA concentration	For highest permeate flux: GA concentration: third level PVA and TiO_2 concentration: fourth level For COD rejection: PVA, GA, and TiO_2 concentration: first level
[19]	UF/Polymeric/ Refinery oily wastewater	Transmembrane pressure (1.5, 3, and 4.5 bar), cross-flow velocity (0.25, 0.75, and 1.25 m/s), pH (4, 7, and 10), and temperature (25, 37.5, and 50°C)/Three	–	Temperature, pH, cross-flow velocity: third level Transmembrane pressure: second level
[20]	MF/Polymeric/ Synthetic oil–water emulsion	Pressure (0.5, 1.5, 2.5, and 3.5 bar), residence time (30.6, 15.3, 10.2, and 7.6 s), water content (5, 10, 15, and 20 vol.%), temperature (35, 45, 55, and 65°C), and emulsifier content (0.2, 0.4, 0.6, and 0.8 vol.%)/Four	Emulsifier content > temperature > residence time > water content > pressure	Temperature, water content: fourth level Residence time, emulsifier content: first level Pressure: second level
[21]	UF/Cellulosic/ Emulsified oil in water	pH (5, 7, and 11), oil concentration (0.5, 1.5, and 3 (%, v/v)), flow velocity (2.6, 2.9, and 3.2 m/s), temperature (25, 35, and 40°C), pressure (0.5, 1.5, and 3.5 bar), and salt (NaCl and $CaCl_2$) concentrations (in runs 1 and 2: 0, 0.1, and 0.5 g/L for both NaCl and $CaCl_2$; in runs 3 and 4: 1.1, 3.3, and 5.5 g/L for $CaCl_2$ and 0, 5.85, and 11.7 g/L for Nacl)/Three	In Run 1 and 2: pH > flow velocity > pressure > oil concentration > temperature > $CaCl_2$ concentration > NaCl concentration In Run 3 and 4: temperature > oil concentration > flow velocity > pressure > NaCl concentration > pH > $CaCl_2$ concentration	Temperature, pressure, flow velocity: third level Salt and oil concentration: first level

Table 2
Characteristics of MF and UF membranes

	MF membrane	UF membrane
Commercial name	α-Alumina	PAN350
Type	Ceramic	Polymeric
Material	Aluminum oxide	Polyacrylonitrile (PAN)
Molecular weight cutoff (MWCO)	Not reported by Foshan Co.	20 kDa
Pore size	0.2 μm	Not reported by Sepro Co.
Water flux (L/m^2 h bar)	>500	1,000
pH range	0–14	3–10
pH range (cleaning)	0–14	3–10
Max. temperature (°C)	800	100
Contact angle	42°	44°
Porosity	30%	35%
Surface area	0.24 m^2	0.006615 m^2

2.2. Process feed

The real produced waters used in the experiments for both the MF and UF systems were generated at the inlet and outlet of skimmer units in two of the desalination units operating in Iran. These feeds were designated feed1 (the inlet of the skimmer unit) and feed2 (the outlet of the skimmer unit). Table 3 gives a detailed analysis of the produced waters. The size distribution of particles ranged from 50 to 720 nm in feed1 and 200 to 800 nm in feed2, which suggests the presence of dissolved oils. Generally, to reach an efficient membrane filtration system for produced-water purification, the oil-droplet size should be larger than the pore size of the membranes. MF employs membranes with a pore size of approximately 0.03–10 microns, an MWCO of greater than 1,000,000 Daltons. UF membranes have a pore size of approximately 2 to 100 nm, an MWCO of approximately 10,000–100,000 Daltons. These pore sizes for MF and UF membranes mean that the studied membranes in this work can effectively separate oil droplets within a range of 20–800 nm from feed flow.

2.3. Filtration section (experimental pilot)

For the experiments with MF membranes, the constructed pilot-scale unit was equipped with three tanks for feed (TK-101), chemical solutions (TK-102) for chemical cleaning, and distilled water (TK-103). Fig. 1 shows the schematic of the filtration unit. As shown in the figure, four valves were used for controlling the streams of feed, permeate, and retentate and for changing the TMP.

The UF experimental setup employed in this study was reported in a previous article [16]. All experiments conducted in cross-flow operation were carried out using concentration mode of filtration (CMF) for 90 min in both systems. During the experiments, the weight of the resulting permeates was measured.

Table 3
Characterization of real produced waters employed in this study

		Values	
Parameter	Unit	Feed1	Feed2
Oil and grease	mg L^{-1}	9	43
TDS	mg L^{-1}	59,628	65,743
COD	mg L^{-1} as O_2	500	450
TSS	mg L^{-1}	108	109
Turbidity	NTU	150	95
Fe	mg L^{-1}	0.26	0.53
TOC	mg L^{-1}	165	110
pH	–	8	8

2.4. Measurements

The values for oil and grease, TSS, turbidity, and TOC of the feed and permeates were measured by special standard methods and devices (Table 4).

The particle-size distribution of emulsified oil droplets in the sample was measured by the light-scattering method using the LLS instrument (SEMA-633), which has a measurement range of 0.4–10,000 nm.

2.5. Experimental design based on the Taguchi method

Disregarding the advantages and drawbacks of Taguchi method, it was selected because of financial matters. We employed Taguchi method to reduce the number of experiments to 9 and economized on use of membranes, fresh water, electricity, feeds, cleaning

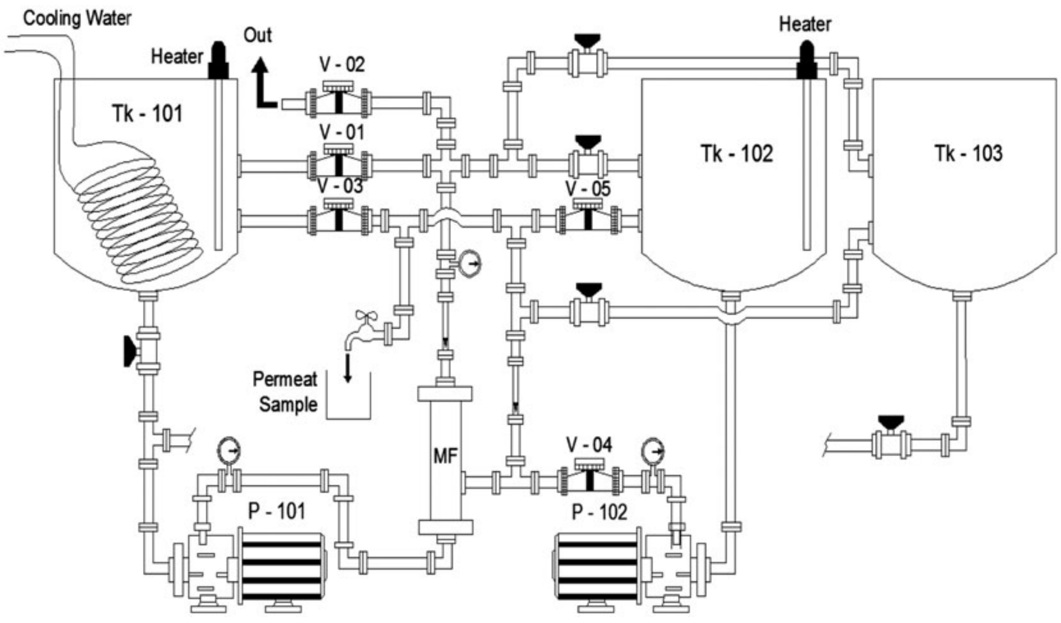

Fig. 1. Schematic diagram of MF experimental pilot.

Table 4
Standard methods [22] and devices used to measure properties of the samples

Parameter	Standard method	Device
Oil and grease	ASTM 5520 B	TOG/TPH analyzer, InfraCal
TSS	ASTM 2540 D	–
Turbidity	ASTM 1889 D	Turbidimeter, HACH, 2100 A
TOC	ASTM 5310 C	TOC analyzer, Dohrmann, DC-190

Table 5
Parameter values and their levels in the both MF and UF systems

		MF system				UF system			
		Levels				Levels			
Parameters	Designation	1	2	3	DOF	1	2	3	DOF
Temperature (°C)	*A*	35	50	60	2	25	40	55	2
Transmembrane pressure (bar)	*B*	1	1.8	2.5	2	1	3	5	2
Cross-flow velocity (m/s)	*C*	0.25	1	3	2	0.5	1	1.5	2
Back pulse time (sec)	*D*	2	4	6	2	–	–	–	–

materials, and so forth. In fact, in the dramatically changing processes, use of Taguchi method is not recommended. The Taguchi method aims at finding optimum process conditions with a minimized sensitivity to noises. It is a type of fractional factorial design that uses orthogonal arrays (OAs) to study the influence of factors using fewer experiments based on the number of control factors and their levels (Table 5). The designed experiments based on the Taguchi method provide a systematic approach to meet optimum conditions [9,21]. Fig. 2 gives a brief overview of the process used in the Taguchi method of factor design.

In this study, four factors for the MF system and three factors for the UF system were chosen based on

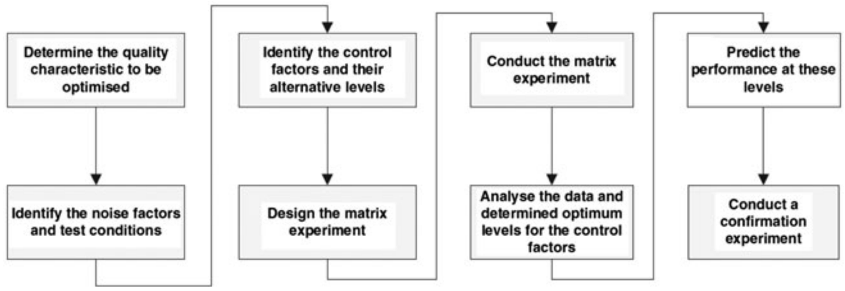

Fig. 2. Flowchart of the Taguchi method [14].

the literature [9,14,16–21,23–26]. Each parameter has two degrees of freedom (DOF); in other words, the minimum number of parameter levels required to determine the levels for the remaining parameters is two. Mathematically, the DOF could be defined as the minimum number of independent coordinates that can specify the position of the system completely. In this method, if k_A is considered the number of parameter levels, the DOF for each factor is $k_A - 1$.

Using the L_9 OA of the Taguchi design [9,16,18], the number of experiments required to investigate the important effects can be reduced to 9, whereas full factorial experimentation requires 3^4 (81) experiments for the MF system and 3^3 (27) for the UF system. The L_9 (3^4) and L_9 (3^3) OAs in the Taguchi design include nine experiments for four factors at three levels for the MF system and three factors at three levels for the UF system. A suggested experimental plan for the L_9s has eight DOF (total DOF). If N is considered as the number of experiments, the total DOF is $N - 1$.

In the Taguchi method, "signal" and "noise" introduce desirable and unacceptable values for outputs, respectively, and their ratio (S/N) is used to transform the response factor to find the optimum conditions, which is obtained using the S/N ratio from the results of experimental data. The equation of the S/N ratio depends on the scale for the quality characteristics to be optimized [16,18,21]. For high permeate flux, a criterion of the-larger-the-better was chosen. The performance characteristic was calculated using the following equation:

$$\text{The-larger-the-better } S/N = -10\log\left(\frac{1}{n}\sum_{i=1}^{n}\frac{1}{Y_i^2}\right) \quad (1)$$

where n is the number of iteration for an experimental combinations and Y_i is the response (here, permeate flux).

ANOVA was applied to determine the significance of the factors [9,16,18,21]. ANOVA's results, including the sum of squares (SS), DOF, mean of square (MS), and error, as well as a statistical parameter called percent contribution (P (%)), are shown in Table 9. The calculation methodology and the equations used are presented in detail in a previous report [16].

3. Results and discussion

3.1. Experimental results

Table 6 gives the values for final permeate flux (J_f), TOC rejection, and turbidity rejection obtained from the experiments for feed1 and feed2 in both the MF and UF systems. The values of TOC and turbidity rejection are almost constant in all the experiments; therefore, just the values of the permeate fluxes at the end of the filtration were considered to find the optimum conditions using the Taguchi approach.

3.2. Taguchi results

Table 7 reports the values of final permeate flux (J_f), and corresponding S/N ratios in the experiments for feed1 and feed2 in the MF and UF systems. To specify the effect of each factor on permeate flux in both systems, the S/N ratio must be calculated for each factor. In the Taguchi method, the S/N ratio is worked out just by averaging the S/N values in different levels of each factor. For instance, the mean S/N ratio values for temperature in the levels 1, 2, 3 could be calculated by averaging the S/N ratios for the trials 1–3, 4–6, and 7–9, respectively [9,16].

Fig. 3 illustrates the average S/N ratios for each factor at three levels in the MF and UF systems for feed1 and feed2. The slopes of the trends for different levels are not identical for either the temperature and BPT factors in the MF system or CFV in the UF system. Therefore, they would have different influences

Table 6
Experimental results obtained from feed1 and feed2 in MF and UF systems

	MF system					UF system				
Trial no.	J_f (L/m^2 h)	TOC of permeate (mg/L)	TOC rejection (%)	Turbidity of permeate (NTU)	Turbidity rejection (%)	J_f (L/m^2 h)	TOC of permeate (mg/L)	TOC rejection (%)	Turbidity of permeate (NTU)	Turbidity rejection (%)
Feed1										
1	80	61	45	0.38	99.83	119	47	71	0.32	99.78
2	120	64	42	0.29	99.87	190	48	71	0.15	99.90
3	230	68	38	0.23	99.90	248	48	71	0.27	99.82
4	165	69	37	0.13	99.94	205	53	68	0.24	99.84
5	330	61	45	0.28	99.88	226	43	74	0.29	99.80
6	200	60	46	0.22	99.90	230	54	67	0.20	99.87
7	285	56	49	0.37	99.84	218	46	72	0.45	99.70
8	215	58	47	0.98	99.58	230	58	65	0.14	99.91
9	400	55	50	0.77	99.67	350	50	70	0.10	99.93
Feed2										
1	70	55	67	0.19	99.80	56	37	66	0.24	99.72
2	114	58	65	0.53	99.44	77	36	67	0.32	99.63
3	210	71	57	0.26	99.72	102	35	68	0.48	99.45
4	145	52	68	0.50	99.66	81	36	67	0.15	99.82
5	308	62	62	0.30	99.68	94	35	68	0.41	99.53
6	170	62	62	0.9	99.05	100	38	65	0.15	99.82
7	250	49	70	0.18	99.81	91	35	68	0.33	99.62
8	185	52	68	0.93	99.02	98	40	63	0.45	99.48
9	375	64	61	0.3	99.68	125	39	64	0.37	99.57

Table 7
Final permeate fluxes obtained from feeds and the corresponding S/N ratios in both systems

Trial no.	MF system—feed1 Permeate flux J_f (L/m^2 h)	S/N ratio	UF system—feed1 Permeate flux J_f (L/m^2 h)	S/N ratio	MF system—feed2 Permeate flux J_f (L/m^2 h)	S/N ratio	UF system—feed2 Permeate flux J_f (L/m^2 h)	S/N ratio
1	80	38.06	119	41.51	70	36.90	56	34.96
2	120	41.58	190	45.57	114	41.14	77	37.73
3	230	47.23	248	47.88	210	46.44	102	40.17
4	165	44.35	205	46.23	145	43.23	81	38.17
5	330	50.37	226	47.08	308	49.77	94	39.46
6	200	46.02	230	47.23	170	44.61	100	40.00
7	285	49.09	218	46.77	250	47.96	91	39.18
8	215	46.65	230	47.23	185	45.34	98	39.82
9	400	52.04	350	50.88	375	51.48	125	41.94

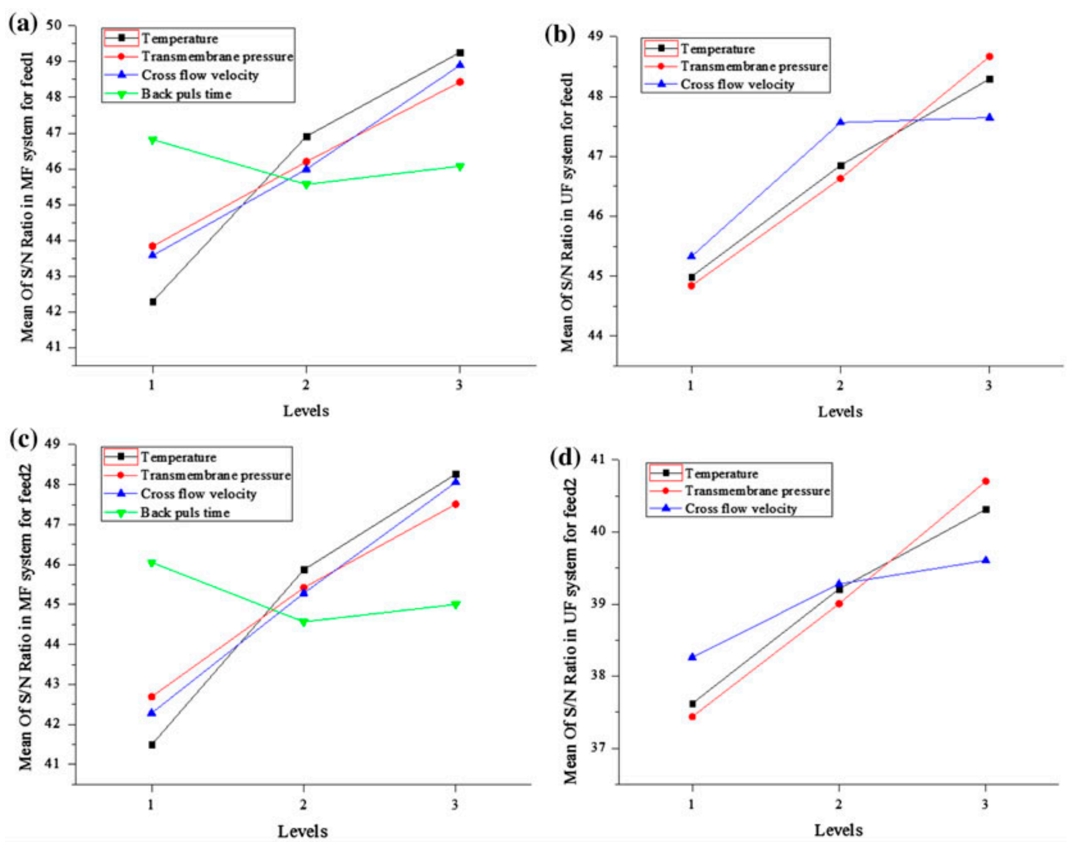

Fig. 3. Main effect curves for S/N ratios of permeate flux of (a) feed1 for four factors in the MF system, (b) feed1 for three factors in the UF system, (c) feed2 for four factors in the MF system, and (d) feed2 for three factors in the UF system, at three levels.

on response factor. Outflowing permeate flux in both the MF and UF systems increased with increases in temperature during the experiments. Many studies show that an increase in temperature increases the amount of permeate flux [24,27]. This increase is usually attributed to the viscosity of solvent, solvent diffusion coefficient in the membrane, and the extent of thermal expansion in the membrane substance. Increasing temperature causes a decrease in the viscosity of solvent and, hence, an increase in the solvent

Table 8
Optimum conditions in the MF and UF systems

Controlling parameters	MF system		UF system	
	Value	Level	Value	Level
Temperature (°C)	60	3	55	3
Transmembrane pressure (bar)	2.5	3	5	3
Cross-flow velocity (m/s)	3	3	1.5	3
Back pulse time (sec)	2	1	–	–

Table 9
ANOVA results for final permeate flux obtained from feed1 in MF and UF systems

MF system					UF system			
Factors	DOF	SS	MS	P (%)	DOF	SS	MS	P (%)
Temperature	2	74,033.333	37,016.667	45.45	2	9,740.667	4,870.333	31.39
TMP	2	30,100	15,050	18.43	2	13,970.667	6,985.333	45.93
CFV	2	40,933.333	20,466.667	25.07	2	4,792.667	2,396.333	14.39
BPT	2	18,233.333	9,116.667	11.05	–	–	–	–
Error	–	–	–	–	2	602	301	8.29
Total	8	163,300		100	8	29,106		100

Table 10
ANOVA results for final permeate flux obtained from feed2 in MF and UF systems

MF system					UF system			
Factors	DOF	SS	MS	P (%)	DOF	SS	MS	P (%)
Temperature	2	57881.333	28940.667	39.51	2	1,040.222	520.111	34.49
TMP	2	28037.333	14018.667	19.14	2	1,649.556	824.778	55.26
CFV	2	39841.333	19920.667	27.19	2	216.222	108.111	6.41
BPT	2	20,748	10,374	14.16	–	–	–	–
Error	–	–	–	–	2	28.222	14.111	3.84
Total	8	146,508		100	8	2,934.222		100

diffusion coefficient. Furthermore, the high temperature may cause the structure of the membrane to expand, facilitating the permeation of solutes [16]. As shown in Fig. 3, the highest permeate flux occurred at the third level of temperature (60°C in the MF system, and 55°C in the UF system) for feed1 and feed2.

In membrane filtration processes, TMP is one of the most significant controlling factors. Fig. 3 illustrates that an increase in TMP in both systems caused an increase in the S/N ratio; the highest level of permeate flux occurred at the highest TMP (2.5 bars in the MF system, and 5 bars in the UF system). Based on Darcy's law, the pressure difference at two membrane sides brings about an increase in permeate flux, although the effects of fouling limit this increase [14]. Previous studies show that at low pressures, the flux increases with pressure, and concentration polarization takes place. This means that permeate flux is directly proportional to TMP. Over this region, higher TMP results in droplets passing rapidly through the membrane pores, so more oil droplets accumulate on the membrane surface and consequently in the membrane pores, leading to membrane fouling [24,28,29].

Also, Fig. 3 shows that as the level of CFV increased, the S/N ratio also rose. Therefore, the highest amount of permeate flux occurred at the highest velocity (3 m/s in the MF system, and 1.5 m/s in the UF system). At a CFV of 0.25 and 0.5 m/s in the MF and UF systems, respectively, because of low levels of turbulence, a cake layer was formed faster. Conversely, as the CFV rose, the permeate flux increased along with the resulting turbulence. Increasing CFV

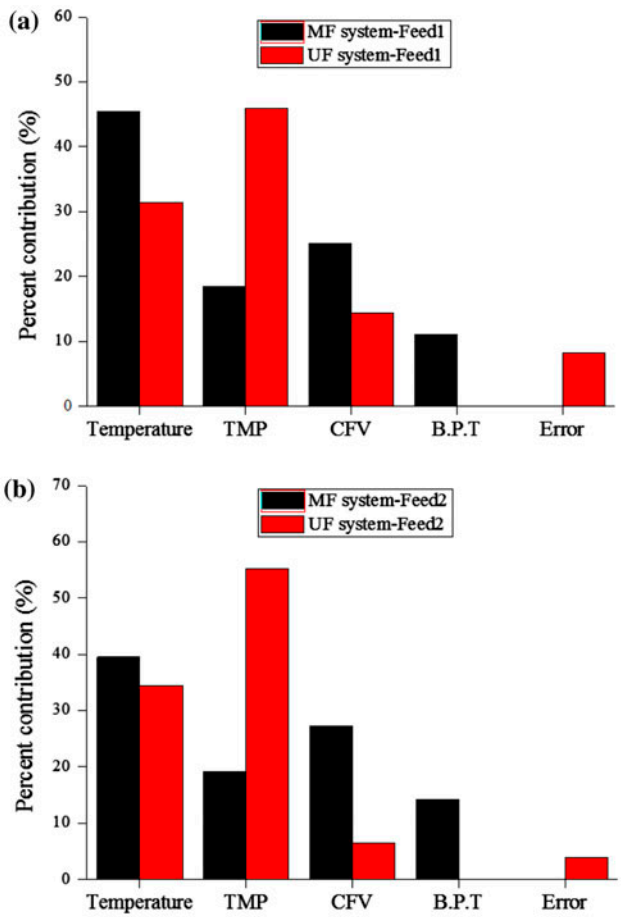

Fig. 4. Percent contribution of each factor on the performance statistics for: (a) feed1 and (b) feed2.

increases the mass-transfer coefficient in the concentration boundary layer and the extent of mixing over the membrane surface [16,24]. At higher velocities, some of the created layer detaches from the membrane surface and returns to the liquid; this phenomenon weakens the influence of concentration polarization and increases permeate flux [19,24,30,31].

In the MF system, BPT was used for decreasing the fouling resistance, which minimizes the TMP on the membrane foulants and lets them go back to the fluid bulk. In this method, the permeate stream valve is closed for several seconds and the TMP decreases until it reaches zero. As Fig. 3 shows, the amount of permeate flux decreased with an increase in BPT. Therefore, the highest amount of permeate flux occurred at the lowest BPT (3 s).

The optimum conditions for the highest permeate flux were chosen based on the levels that gave the highest S/N ratios for the factors. The optimum operating conditions in the MF and UF systems are listed in Table 8. As in many previous studies, the optimum temperature and CFV were the highest. For instance, Salahi et al. [19] in a UF treatment of oily wastewater found the optimal temperature, CFV, and pH at their third, or highest, level. Hesampour et al. [21] purified emulsified oil in water using cellulosic UF membrane and obtained the optimum temperature and flow velocity at their third, or highest, level.

3.3. ANOVA results

ANOVA was employed to see whether or not the controlling parameters of the processes were statistically significant. Results of ANOVA for the MF and UF systems are listed in Tables 9 and 10 for feed1 and feed2, respectively. The percentage contributions of all factors, obtained from Eq. (6) of [16], to permeate flux in both systems are shown in Fig. 4. In the MF system, temperature has the most effect, followed by CFV, TMP, and BPT. In the UF system, TMP has the most effect, followed by temperature and CFV. With regard to diverse parameters in membrane filtration systems, different results could be obtained. For example, the factor with the most effect on the treatment of a real oily wastewater using an MF polymeric membrane was CFV, followed by TMP, temperature, pH, and salt concentration [14]. Gorouhi et al. [17] identified feed flow velocity as the factor with the most effect, followed by TMP and temperature, in a MF system used for purification of a synthetic oily wastewater.

3.4. Long-term experiments

To conduct the second stage, which was to study the performance of α-Alumina and PAN350 membranes under optimum conditions, the processes of prolonged MF and UF were performed on the feeds for 4 h. The amount of permeate flux decreased with filtration time, as a result of pore blocking, concentration polarization, and cake-layer formation [23,32–34], which occurred in the first one hour of the process; thereafter, permeate flux from membranes decreased gradually to the point that the system reached a steady state. By the end, the level of flux was almost constant. Table 11 reports the final amounts of flux obtained from filtration of feed1 and feed2 during the four-hour experiments in both systems. As noted, flux decline is related to membrane fouling. Almost all factors of a feed, to a certain extent, cause fouling of membrane. The nature and amount of the sediments depend on several things, such as the range of pore size, concentration of solute material, flow hydrodynamics, properties of the membrane surface, and the

Table 11
Process performance of α-Alumina and PAN350 membranes at optimum conditions and the values of final permeate flux in long-term experiments in the MF and UF systems

		Parameter	Produced-water quality	Permeate quality	Percent removal-[standard deviation]	Final flux (L/m^2 h)
Feed1	MF system	Oil and grease	9	<2 ppm	99-[5.90]	312
		TSS	108	Trace	100-[0.48]	
		Turbidity	150	0.28	99-[0.37]	
		TOC	165	62	63-[1.99]	
	UF system	Oil and grease	9	<2 ppm	99-[5.53]	280
		TSS	108	Trace	100-[0.39]	
		Turbidity	150	0.6	99-[0.31]	
		TOC	165	40	76-[1.72]	
Feed2	MF system	Oil and grease	43	<2 ppm	99-[7.11]	295
		TSS	109	Trace	100-[0.54]	
		Turbidity	95	0.3	99-[0.25]	
		TOC	110	25	77-[2.26]	
	UF system	Oil and grease	43	<2 ppm	99-[6.95]	104
		TSS	109	Trace	100-[0.44]	
		Turbidity	95	0.4	99-[0.17]	
		TOC	110	32	71-[2.13]	

Table 12
MF module cost.

	Number of elements	Element price (€)	Number of modules
Alumina membrane	61	180	96
Module housing	–	50.75	–

Table 13
Raw material cost for the MF system in this study

Material	Annual consumption	Unit price ($)	Total cost (million $)
Citric acid	14,600 kg	1.4682	0.02137
NaOH	8,760 kg	0.81566	0.00714
Microfiltration filter	–	–	0.027
Total raw material	23,360	–	0.05551

interactions between the membrane and the dissolved material [16].

The performances of the α-Alumina and PAN350 membranes for MF and UF at optimum conditions are given in Table 11. All rejections of oil and grease, TSS, turbidity, and TOC were calculated according to feed concentrations, and composite permeate concentrations were obtained at the end of the long-term experiments in both systems to evaluate the overall performance of each system. The final permeation flux after 4 h was completely free of oil and grease, turbidity, and TSS. However, TOC could not be entirely removed. Although these membranes are a good selection to treat the produced water, the ceramic membrane (α-Alumina) performed much better than the polymeric membrane (PAN350) because of its higher outflowing permeate flux.

Therefore, to design a new filtration pilot for the feed of a skimmer unit outlet (feed2), based on the long-term permeate flux in the MF system, the required number of membrane modules was calculated. All the necessary equipment and materials, including tanks, heat exchangers, cleaning materials (acids and bases), and filters, were listed and their prices were calculated. It was determined that 96 ceramic membrane modules were needed to provide the required area for filtration. The cost of vessels, tanks, and heat exchangers was calculated based on their materials, thicknesses, and shape. In the long-term experiments, the slope of permeate flux was used to predict the time at which a 10% decrease in permeate flux would occur; this time was taken as the start of the cleaning process. Based on this, the amount of cleaning agents required annually to operate the filtration pilot was calculated. The price of other required items, such as land, utilities, contingencies, insurance, wiring, and a building, was estimated from the engineering estimations in chemical-engineering handbooks [35]. The calculated cost of produced-water management using membrane filtration was $0.304/bbl (~$0.002/L); a comparison of this value with reported values from other studies ($0.25–2/bbl) or ($0.0016–0.0126/L) [36] shows the high potential of this process as an alternative for produced-water management.

3.5. Economic investigation of the MF process

Economic investigation of a process has two main concepts: fixed investment and production cost. Fixed investment includes direct costs (such as equipment costs, instrumentation, and control and piping costs) and indirect costs (such as engineering and construction costs). In the case of fixed investment, equipment cost was estimated by calling producers for their prices. The total fixed investment required for designed plant is $5.51673 million.

Production cost relates to the production process and includes factors such as raw material cost, maintenance, insurance, general plant overhead, and depreciation. Tables 12 and 13 represent the MF module and the required treating process costs, respectively.

Likewise, total production cost—insurance, wage, overhead, etc.—for this plant is $0.60168 million, and the calculated profit for the last year of operation is $1.98304 million. The calculated internal rate of return (IRR) is 25.83%, and the period of investment return is 4.67 years. Given the annual purification capacity of the designed plant, which is 3,197,400 m^3 or 2,275 m^3/m^2, the management cost of wastewater is $0.304/bbl (~$0.002/L).

4. Conclusions

This study describes the purification of two real produced waters in two-stage MF and UF processes. Firstly, the effects of operating parameters—temperature, TMP, and CFV—in both systems and BPT (in the MF system alone) at three levels on the amount of permeate flux was studied using ceramic and polymeric membranes. The Taguchi method was used to find the optimum conditions:

(1) Third level of temperature (60°C in the MF system and 55°C in the UF system).
(2) Third level of TMP (2.5 bars in the MF system and 5 bars in the UF system).
(3) Third level of CFV (3 m/s in the MF system and 1.5 m/s in the UF system).
(4) First level of BPT (2 s) in the MF system.

ANOVA proved that in the MF system, temperature was the factor with the most influence on the permeate flux, followed by CFV, TMP, and BPT. In the UF system, TMP had the most effect, followed by temperature and CFV. In the second step of this study, the performance of ceramic and polymeric membranes was studied under the optimum conditions, with the following results:

(1) The rejection values of oil and grease, TSS, and turbidity were 99, 100, and 99%, respectively, for feed1 and feed2, for both systems. (2) In the MF system, TOC-removal values were obtained as 63% and 77% for feed1 and feed2, respectively. (3) In the UF system, TOC-rejection values were obtained as 76 and 71% for feed1 and feed2, respectively.

Finally, the management cost of produced-water treatment by MF system was estimated as $0.304/bbl (~$0.002/L), indicating the high potential of this process as an alternative for produced-water management. This study suggests that α-Alumina and PAN350 membranes are good choices for MF and UF in produced-water treatment.

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