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Optimization of a membrane hybrid process for oil-in-water emulsions treatment using Taguchi experimental design

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ABSTRACT

A coagulation–centrifugation/ultrafiltration hybrid process has been performed for the treatment of a commercial oil-in-water emulsion used in metalworking operations. The effect of pre-treatment stage on membrane fouling was evaluated to ascertain the main fouling mechanism and to establish the best membrane cleaning procedure. The hybrid process was optimized in terms of the maximum ultrafiltration (UF) permeate flux. Destabilization temperature, transmembrane pressure, feed flow rate to the UF module, and coagulant salt molar concentration were selected as factors. The experiments were designed using Taguchi method, and the contribution of each factor was determined using a statistical analysis of variance. Experimental results were also discussed in terms of environmental parameters (chemical oxygen demand [COD], conductivity, pH, and turbidity) for the main process streams. The best fit to experimental data of permeate flux decline corresponded to the cake/gel layer formation model. The cleaning procedure combined alkaline and acid washings with mechanical cleaning of membrane surface, and through this combination total permeate flux recovery was achieved. Temperature was the most significant factor affecting permeate flux, followed by coagulant salt concentration, with COD reductions higher than 97.5% for all experimental conditions tested.

Keywords: Oil-in-water emulsion; Hybrid process; Coagulation; Centrifugation; Ultrafiltration; Membrane fouling; Taguchi method

1. Introduction

Oily wastewaters are mainly generated as oil-in-water (O/W) emulsions in many industrial processes, such as petroleum refining, petrochemical, food, leather, and metal finishing. The main source of industrial oily wastewaters is O/W emulsions used as metalworking fluids (MWFs) in cutting, rolling, grinding, finishing, and drawing operations, where produce a substantial impact on tool life and workpiece quality [1].

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These emulsions are complex mixtures of oil, surfactants, additives, and water, which are used as lubricants and cooling agents at the machine–workpiece interface, but they also have other functions, such as chip removal, corrosion protection, and microbial growth control.

Due to thermal and mechanical stresses, these fluids lose their functional properties with use and ultimately need to be replaced, generating large volumes of oily effluents that must be treated before being discharged [2–5]. Furthermore, these exhausted O/W emulsions are considered as hazardous industrial wastewater, and pose a great problem in facilities attempting to comply with discharge limits. Typical limits for oil and grease discharge are in the range of 5–30 mg L^{-1}, depending on the oil source [6].

The treatment process of oily wastewater will depend on physical nature of the oil, total oil content, and chemical nature of other components. The most common treatment methods are settling [7], chemical destabilization (coagulation/flocculation) [8], centrifugation [9], flotation [10], filtration/coalescence [11], electrical methods [3,12], membrane processes [13–20], and vacuum evaporation [21,22]. Often a combination of several techniques is more efficient and cost-effective [23–25].

Membrane processes, mainly microfiltration (MF) and ultrafiltration (UF), have proven to be successful techniques for the treatment of complex oily wastewater, since no chemical additives are required, have low operating and capital costs, and the quality of permeate obtained is rather high (i.e., low chemical oxygen demand (COD)), making it suitable for several applications, such as process water or fresh O/W emulsion reformulation [26]. However, sometimes it is not desirable or even possible to use a membrane system to carry out the entire separation. The major drawback in membrane processes is the permeate flux decline over time, due to concentration polarization and membrane fouling as a result of adsorption and accumulation of rejected oil and other components on the membrane surface [27,28], which leads to high energy consumption and frequent membrane cleaning requirements, and shortens membrane lifetime [14,25,29–33]. Pre-treatment of O/W emulsions prior to membrane filtration is necessary to maintain a high and steady flux through the membrane. In these situations, membrane hybrid processes may be the alternative to obtain a good process performance and to extend membrane life. A membrane hybrid process is the combination of a conventional treatment (mechanical, chemical, or thermal) with a membrane process. These methods have shown successful results for the treatment of MWFs [34–36].

The study and optimization of this kind of hybrid processes generally imply a high number of long-term experiments that can be reduced using Taguchi experimental design, which facilitates the study of a system by a set of independent variables (factors) over a specific region of interest (levels) influencing a process response factor. Taguchi method is applied to fractional factorial design using orthogonal arrays (OA), and it is recommended for long or cumbersome experiments [37]. It has been applied to MF, UF, nanofiltration, and reverse osmosis [38–46].

In this work, a coagulation–centrifugation/ultrafiltration hybrid process was studied for the treatment of an O/W emulsion prepared with a commercial MWF. A coagulant salt, calcium chloride, was used as destabilization agent in the coagulation–centrifugation stage, as reported in previous works [9,30]. The aqueous phase from centrifugation was fed into an UF stage provided with a flat ceramic membrane. The effect of pre-treatment stage on membrane fouling was also evaluated in order to ascertain the main fouling mechanism and to establish the membrane cleaning procedure. The main objective was to determine the best operating conditions for the integrated process to achieve a maximum UF permeate flux. The design of experiments (DOE) using Taguchi approach and subsequent statistical analysis of variance (ANOVA) was applied to evaluate the optimum value for each factor with the minimum number of experimental runs.

2. Materials and methods

2.1. Materials

A fresh, non-used O/W emulsion was prepared in the laboratory from a commercial oil concentrate Besol 5 (Brugarolas Co., Spain). This concentrate was selected because of its wide range of applications in machining processes and its long-term stability [9]. Although its composition is proprietary, it consists of a mixture of mineral oils and several additives, such as emulsifiers, stabilizers, biocides, and corrosion inhibitors. The concentrate was dispersed in tap water by vigorous stirring with a Heidolph Diax 900 homogenizer (Germany), at 15,000 rpm for 5 min, to yield a 1 wt.% emulsion. Characteristics of this emulsion are summarized in Table 1.

Anhydrous calcium chloride ($CaCl_2$, reagent grade), supplied by Panreac Química S.A. (Spain), was used as coagulant salt for O/W emulsion destabilization. It was added to the fresh emulsion at adjusted concentration depending on trial conditions. Then, emulsion destabilization was completed within 30 min in a thermostatic bath.

Table 1
O/W emulsion characteristics

Oil concentration (wt.%)	pH	COD (mg L^{-1})	Turbidity (NTU)	Conductivity (mS cm^{-1})
1	9.2	20,500	>2,000	570.5

Centrifugation of destabilized O/W emulsions was performed using a thermostatic Kubota 6300 (Japan) centrifuge for 15 min at 4,500 rpm. Tangential UF of the resulting aqueous phase was carried out in total recycle mode for 1–1.5 h using 300 kDa cut-off flat disc ceramic membranes (INSIDE DisRAM, TAMI Industries, France), with an active layer of ZrO_2 supported on TiO_2, 2.5 mm thick, 90 mm of diameter, and 56.3 cm^2 of effective area. A schematic diagram of the process is shown in Fig. 1.

$CaCl_2$ aqueous solutions and non-destabilized fresh O/W emulsions were also ultrafiltrated to evaluate membrane fouling and to establish the membrane cleaning procedure. P3-Ultrasil 40 detergent (Ecolab, Spain), citric acid, nitric acid, and acetone (all of them supplied by Panreac Química S.A., Spain) were used as cleaning agents.

COD, conductivity, pH, and turbidity values of the fresh emulsion; aqueous phases after centrifugation (or UF feed); and UF permeates were measured with a HACH DR2010 spectrophotometer (USA), following the reactor digestion method [47], a Mettler Toledo SG3 conductivity meter (Spain), a Crison Basic 20 pH meter (Spain), and a HACH Ratio/XR turbidity meter (USA), respectively.

2.2. Taguchi DOE method

Taguchi method applies fractional factorial designs, called OA, to reduce the number of experiments required to determine their influence over the process output, in this case the UF permeate flux (J) after 1 h of operation. The effects of four factors, *viz.* destabilization temperature (T), transmembrane pressure (TMP), feed flow rate to the UF module (Q), and coagulant salt molar concentration (M) on process performance were analyzed. The experimental design consisted of a set of nine trials corresponding to L_9 orthogonal array under the specific conditions selected for this study: four factors and three levels, summarized in Table 2. At least two runs were performed for each trial to avoid nonlinearity effects.

3. Results and discussion

3.1. Ultrafiltration: membrane fouling and cleaning

Several UF experiments were performed to evaluate flat ceramic membrane fouling. According to Darcy´s law, the permeate flux (J) through the membrane can be expressed as:

$$J = \frac{1}{A_m}\frac{dV}{dt} = \frac{TMP}{\mu R_t} = \frac{TMP}{\mu(R_m + R_f)} \quad (1)$$

where V is the permeate volume, A_m is the membrane area, TMP is the transmembrane pressure, μ is the permeate viscosity, and R_t is the total resistance to permeate flux, which can be expressed, according to the resistance-in-series model, as the sum of the intrinsic membrane resistance, R_m, and the resistance due to fouling, R_f, including concentration polarization.

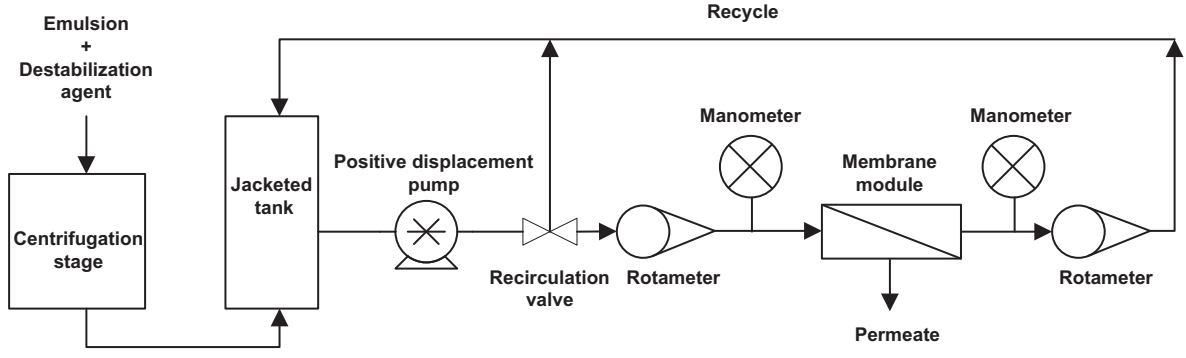

Fig. 1. Schematic diagram of the membrane hybrid process.

Table 2
Factors and levels in the experimental design

Levels	Factors			
	Transmembrane pressure TMP (bar)	Temperature T (°C)	UF feed flow rate Q (L h^{-1})	Coagulant salt concentration M (mol $CaCl_2$ L^{-1})
1	1.0	20	60	0.10
2	1.5	40	90	0.15
3	2.0	60	120	0.20

R_m can be readily determined from pure water UF at different TMP values, since in this case, R_f is equal to zero. These experiments revealed that R_m is equal to 2×10^{12} m^{-1} for the ceramic membrane used.

Concentration polarization is a reversible process because flux reduction is due to an increased diffusional resistance in the boundary layer, which ends spontaneously after the operation. Permeate flux reduction due to concentration polarization can be minimized using a high tangential velocity over the membrane surface [14], as it was the case for the feed flow rates tested in this work.

Fouling causes an irreversible decline in permeate flux, and it may be due to several factors, i.e. surfactant, salt, or oil adsorption on membrane pore walls, membrane pore blocking by oil droplets, or oil cake/gel layer formation on the membrane surface [14,25,28–33,48,49].

Permeate flux reduction for UF of the fresh O/W emulsion and the aqueous phase from destabilization/centrifugation treatment is shown in Fig. 2.

A drastic flux reduction for UF of non-destabilized fresh O/W emulsion is observed because of severe membrane fouling that occurred in the first seconds of operation, reaching a constant permeate flux value of 70 L m^{-2} h^{-1}. However, it can be noticed that the previous demulsification stage reduced membrane fouling, increasing by more than twice the final value of permeate flux (156 L m^{-2} h^{-1}), which remained constant over 20 min of operation. The total resistance for UF of the centrifuged aqueous phase, calculated using Eq. (1), was $R_t = 4.7 \times 10^{12}$ m^{-1}. Thence, the resistance due to membrane fouling was $R_f = 2.7 \times 10^{12}$ m^{-1}.

Hermia's models [50] for dead-end filtration have been tested to ascertain the fouling mechanisms involved in the crossflow UF of the centrifuged aqueous phase. These models, based on constant pressure filtration laws, can be applied for MF [28,51–53] and UF [54,55]. The general equation is as follows [53,54]:

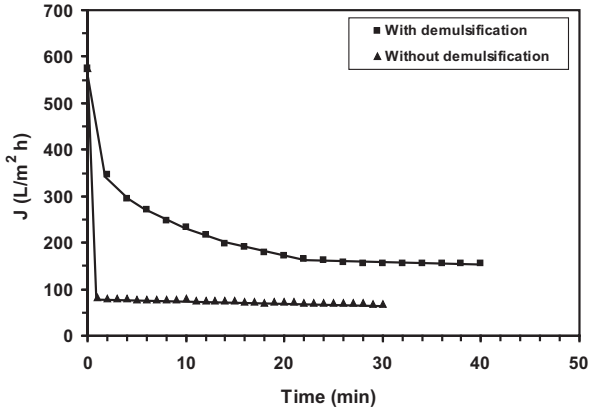

Fig. 2. Permeate flux reduction for UF of the fresh O/W emulsion and the centrifuged aqueous phase obtained after O/W emulsion demulsification with 0.1 mol L^{-1} $CaCl_2$. Operating conditions: $Q = 90$ L h^{-1}, TMP = 2 bar, and $T = 20$°C.

$$\frac{d^2t}{dV^2} = K\left(\frac{dt}{dV}\right)^n \quad (2)$$

where K is a constant, and n is a parameter which takes different values for each fouling model. Table 3 summarizes the fouling models, n values, and Eq. (2) linearized and expressed in terms of permeate flux (J) vs. time.

Complete pore blocking and cake/gel layer formation usually occur when solute molecules (particles or oil droplets) are similar to, or larger than membrane pores, respectively, causing an external fouling on the membrane surface. On the other hand, standard (or pore narrowing) and intermediate pore blocking correspond to fouling inside the membrane pores [51,56]. Pore narrowing by adsorption of dissolved matter as well as pore plugging are considered irreversible fouling [57,58].

In order to ascertain if J decline was controlled by cake/gel layer formation or by pore blocking,

Table 3
Mathematical expressions for membrane fouling models [51,54]

Model	n value [Eq. (2)]	Equation[a]
Complete pore blocking	2	$\ln J = \ln J_0 - K_c t$ (3)
Standard pore blocking	1.5	$\frac{1}{J^{1/2}} = \frac{1}{J_0^{1/2}} + K_s t$ (4)
Intermediate pore blocking	1	$\frac{1}{J} = \frac{1}{J_0} + K_i t$ (5)
Cake/gel layer formation	0	$\frac{1}{J^2} = \frac{1}{J_0^2} + K_g t$ (6)

[a]K_c, K_s, K_i, K_g are constants, and J_0 is the initial permeate flux.

experimental data for the UF of the aqueous phase from Fig. 2 were fitted to these fouling models. The highest value of the coefficient of determination (R^2) was obtained for the cake/gel layer formation model ($R^2 = 0.993$, Fig. 3), followed by the intermediate ($R^2 = 0.941$), standard ($R^2 = 0.902$), and complete ($R^2 = 0.834$) pore blocking models. Therefore, it was concluded that best fit to experimental data, for the experimental conditions tested, corresponded to the cake/gel layer formation model (Fig. 3).

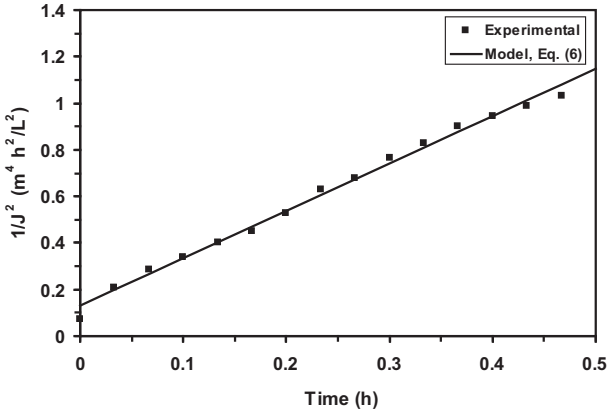

Fig. 3. Permeate flux predicted by the cake/gel layer formation model for the UF of the centrifuged aqueous phase obtained after O/W emulsion demulsification with 0.1 mol L^{-1} $CaCl_2$. Operating conditions: $Q = 90$ L h^{-1}, TMP = 2 bar, and $T = 20$ °C.

The effect of coagulant salt addition on O/W emulsion stability has already been studied [9,30], and the minimum $CaCl_2$ concentration needed to destabilize the emulsion known as *critical coagulation concentration* (CCC) was 0.05 mol L^{-1}. $CaCl_2$ concentrations higher than CCC cause emulsion destabilization by coalescence of oil droplets [9], which explained the cake/gel layer formation on the membrane surface.

UF of $CaCl_2$ aqueous solutions was also performed in order to evaluate the effect of coagulant salt concentration on membrane fouling. Experimental results shown in Fig. 4 indicate that low $CaCl_2$ concentrations (0.1 mol L^{-1}) had no effect on permeate flux. However, higher $CaCl_2$ concentrations (0.2 mol L^{-1}) led to membrane fouling, probably by pore blocking.

After each UF experiment of centrifuged aqueous phase from emulsion destabilization, the ceramic membrane was treated with the different cleaning agents. Water flux was measured before and after each cleaning stage, in order to check the permeate flux recovery.

The best results were obtained with the following membrane cleaning procedure: (i) rinsing with water for 10 min at room temperature; (ii) alkaline washing

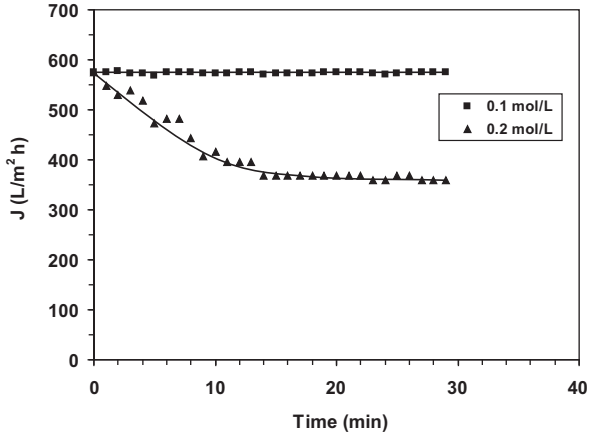

Fig. 4. Effect of coagulant salt concentration on permeate flux reduction for UF of $CaCl_2$ aqueous solutions. Operating conditions: $Q = 90\ L\ h^{-1}$, TMP = 2 bar, and $T = 20$°C.

for 20 min with a solution of 10 g L^{-1} P3-Ultrasil 40 detergent in hot water (50°C); (iii) rinsing with hot water for 20 min; (iv) acid washing for 20 min with a solution containing citric (1.6 g L^{-1}) and nitric (1.8 g L^{-1}) acids in hot water; (v) rinsing with hot water for 20 min; (vi) mechanical cleaning of membrane surface with acetone; and (vii) rinsing with water at room temperature for 10 min. Results indicated that permeate flux recovery after the first rinsing stage was 15%, which corresponded to concentration polarization. This recovery was 39% for alkaline washing and 27% for acid washing. Finally, the mechanical cleaning stage led to a permeate flux recovery of 19%. So, for all trials performed in this study, the total permeate flux recovery after complete membrane cleaning was 99–100% with regard to the initial water permeate flux.

3.2. Coagulation–centrifugation/UF hybrid process

The optimum conditions were those that enabled the system to reach the maximum permeate flux. This was determined by statistical analysis of results from Taguchi method, which used the signal-to-noise (S/N) ratio as statistical indicator. Since high permeate flux was preferred, "the larger the better" criterion was chosen for the S/N ratio [37]. According to this criterion, the S/N ratio was calculated as follows [40]:

$$\frac{S}{N} = -10 \log_{10}\left(\frac{1}{\text{MSD}}\right) \tag{7}$$

$$\text{MSD} = \frac{1}{N}\sum_i Y_i^2 \tag{8}$$

where MSD is the mean standard deviation, Y is the response factor (permeate flux, J), and N is the number of observations (two in this work). Calculated S/N ratios, response factors, and trial conditions are summarized in Table 4.

S/N analysis allows determining the most favorable levels for each factor according to permeate flux for emulsion hybrid treatment, and the partial contribution of each of them to the global process. Optimization criterion was the maximization of permeate flux, so that S/N ratio value should be as high as possible [37]. According to this criterion, the best results were obtained for trial six. To evaluate the influence of each factor in permeate flux, S/N ratio was also calculated for each factor at each level by averaging the corresponding S/N for each trial, where the level studied was the same. S/N variation for each factor is shown in Fig. 5.

Table 4
Experimental conditions, measured permeate fluxes (response factor), and calculated S/N ratios for each trial in the L_9 array

	Factors				Response factor (J)			
Trial	TMP (bar)	T (°C)	Q (L h^{-1})	M (mol L^{-1})	Run 1 (L m^{-2} h^{-1})	Run 2 (L m^{-2} h^{-1})	MSD	S/N
1	1.0	20	60	0.10	73.5	81.4	6,011	37.8
2	1.0	40	90	0.15	509.3	583.3	299,851	54.8
3	1.0	60	120	0.20	455.1	550.1	254,912	54.1
4	1.5	20	90	0.20	263.9	373.5	104,603	50.2
5	1.5	40	120	0.10	188.0	193.4	36,385	45.6
6	1.5	60	60	0.15	788.8	804.8	635,034	58.0
7	2.0	20	120	0.15	189.7	133.5	26,905	44.3
8	2.0	40	60	0.20	349.2	315.7	110,780	50.5
9	2.0	60	90	0.10	332.5	315.1	104,918	50.2

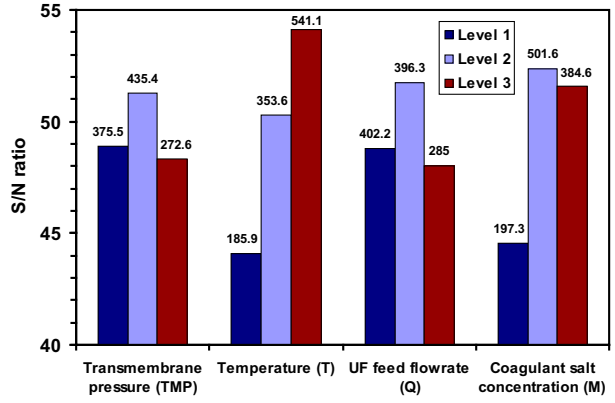

Fig. 5. Variation of permeate flux ($L\,m^{-2}\,h^{-1}$) and S/N ratio for different factors.

ANOVA approach was employed to analyze the results from L_9 OA to determine the relative importance of each factor studied. The significance of each factor was assessed through the p-values obtained from the ANOVA F statistic. Table 5 shows ANOVA results, where degrees of freedom (DOF), sum of squares, mean square (variance), F-ratio, and the contribution percentage of each factor on response are given. Residual row in Table 5 refers to experimental error and also to error caused by uncontrollable factors (noise) which are not included in the experiment: this value should be less than 50%, for the results to be considered reliable. In this study, residuals contribution is about 2%, so experimental error is not significant. All factors have a statistically significant effect on J at the 5% significance level, with p-values less than 0.05.

Quality parameters, measured for each stream in every trial, are reported in Table 6.

Results from Table 5 indicate that temperature is the most important factor affecting permeate flux, with the highest F-ratio value and 47% of total contribution, followed by coagulant salt concentration, whose contribution is 35%. It can also be observed in Fig. 5 that S/N ratio values for temperature and coagulant salt concentration change significantly depending on the level studied.

As it might be expected, permeate flux increased with increasing temperature (Fig. 5), due to the decrease in viscosity and density, thus obtaining high-quality permeates (Table 6). Nevertheless, at level three, improvement in permeate flux was not

Table 5
ANOVA of L_9 orthogonal array

Factor	DOF	Sum of squares	Variance	F-ratio	% Contribution
Transmembrane pressure (TMP)	2	81,384	40,692	23.29	10
Temperature (T)	2	378,638	189,319	108.37	47
UF feed flow rate (Q)	2	52,331	26,166	14.98	6
Coagulant salt concentration (M)	2	282,699	141,350	80.91	35
Residual	9	15,722	1,747		2

Table 6
Quality parameters of aqueous phases from coagulation/centrifugation stage (UF feed) and UF permeates

	UF feed				UF permeate			
Trial	COD reduction[a] (%)	pH	Turbidity (NTU)	Conductivity ($mS\,cm^{-1}$)	Total COD reduction[a] (%)	pH	Turbidity (NTU)	Conductivity ($mS\,cm^{-1}$)
1	76.8	7.1	225	19.7	97.6	7.7	0.18	11.4
2	77.9	8.2	185	16.1	98.0	8.1	0.07	11.9
3	80.0	8.2	165	15.6	98.2	8.3	0.03	14.8
4	76.9	7.9	205	13.7	98.5	7.9	0.16	12.1
5	77.6	7.8	192	11.2	98.3	7.6	0.07	10.1
6	80.2	8.4	176	13.1	97.8	8.3	0.05	11.7
7	76.8	7.2	216	10.7	97.9	7.1	0.12	12.3
8	78.0	8.6	198	11.8	97.7	8.4	0.16	12.9
9	79.4	8.3	171	11.2	97.9	8.1	0.09	11.0

[a]Related to initial O/W emulsion.

observed as coagulant salt concentration increased. It could be explained by the large excess of $CaCl_2$ over the CCC, which reduces permeate flux by increasing membrane fouling [30], as it was previously observed (Fig. 4).

TMP and feed flow rate had less influence on permeate flux, with total contributions of 10 and 6%, respectively (Table 5). A combination of higher temperature, lower feed flow rate, and moderate TMP caused a lower decline in permeate flux. As shown in Fig. 5, increasing TMP from 1 to 1.5 bar led to an increase in permeate flux, and also in oil rejection, thus improving permeate quality (Table 6). At higher TMP (2 bar), the oil layer over the membrane surface became more compact and pore blocking took place, with severe permeate flux reduction and slight decrease in permeate quality due to the passage of more pollutants through the UF membrane pores. However, COD reductions higher than 97.5% were obtained in all cases.

4. Conclusions

A hybrid coagulation–centrifugation/ultrafiltration process has been optimized for the treatment of a commercial O/W emulsion using flat disc ceramic membranes. The previous demulsification stage reduced membrane fouling and increased the permeate flux by more than 200%. A membrane cleaning procedure was developed allowing complete membrane regeneration after emulsion treatment.

The hybrid process was optimized using Taguchi methodology and ANOVA analysis. Among the factors analyzed, temperature seemed to have more influence on permeate flux than the other ones, followed by coagulant salt concentration. The optimum operating conditions for maximizing permeate flux were: TMP = 1.5 bar, $T = 60$ ℃, $Q = 60$ L h^{-1}, and $M = 0.15$ mol $CaCl_2$ L^{-1}, with COD reduction of 97.8 %.

Treatment of a commercial MWF with the proposed hybrid process yielded COD reductions higher than 97.5% for all experimental conditions tested, making these high-quality UF permeates suitable for several applications, such as process water or O/W emulsion reformulation.

Nomenclature

A_m — membrane area (m^2)
ANOVA — analysis of variance
COD — chemical oxygen demand (mg L^{-1})
DOF — degrees of freedom
J — permeate flux (L m^{-2} h^{-1})
J_0 — initial permeate flux (L m^{-2} h^{-1})
K — constant in Eq. (2) (units depend on the parameter n)
K_c — complete pore blocking model constant (s^{-1})
K_g — cake/gel layer formation model constant (s m^{-2})
K_i — intermediate pore blocking model constant (m^{-1})
K_s — standard pore blocking model constant ($m^{-0.5}$ $s^{-0.5}$)
M — coagulant salt concentration (mol L^{-1})
MSD — mean standard deviation
n — constant in Eq. (2) that depends on the fouling mechanism (dimensionless)
N — number of observations
NTU — nephelometric turbidity units
Q — feed flow rate to ultrafiltration stage (L h^{-1})
R^2 — coefficient of determination
R_f — resistance due to fouling (m^{-1})
R_m — intrinsic membrane resistance (m^{-1})
R_t — total resistance to permeate flux (m^{-1})
S/N — signal-to-noise ratio
t — time (s)
T — destabilization temperature (°C)
TMP — transmembrane pressure (bar)
V — permeate volume (L)
Y — response factor
μ — permeate viscosity (kg m^{-1} s^{-1})

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