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Highlights

O/W emulsion was treated by coagulation/centrifugation and ultrafiltration (UF)

The hybrid process was optimized in terms of flux and quality parameters of permeate

Taguchi experimental design was applied with three levels for each factor studied

TMP and temperature were the most significant factors affecting permeate flux

Permeate COD was mainly influenced by UF feed flow rate and temperature

Treatment of oil-in-water emulsions by a destabilization/ultrafiltration hybrid process: statistical analysis of operating parameters

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Abstract

A hybrid process has been studied for an oil-in-water (O/W) emulsion treatment. This process consisted of two stages: emulsion destabilization by coagulation/centrifugation with calcium chloride, and subsequent ultrafiltration (UF) using a 300 kDa tubular multichannel ZrO_2 ceramic membrane. The O/W emulsion was formulated from a commercial oil concentrate (1 wt % in distilled water) used in metalworking processes. The hybrid process was optimized in terms of ultrafiltration permeate flux and permeate quality parameters, such as chemical oxygen demand (COD), pH, conductivity and turbidity. Experiments' planning was designed using Taguchi method to determine the influence of four parameters (transmembrane pressure, feed flow rate to the UF module, destabilization temperature, and coagulant salt molar concentration) on a response factor, with three levels for each of them. The contribution of each factor was determined using a statistical analysis of variance (ANOVA). Transmembrane pressure and temperature were the most significant factors affecting permeate flux, while permeate quality, expressed as COD, was mainly influenced by UF feed flow rate and temperature. This behavior was slightly different when ultrafiltration was performed with 300 kDa flat ZrO_2 ceramic membranes.

Keywords: Oil-in-water emulsion; Taguchi method; Coagulation; Centrifugation; Ultrafiltration

1. Introduction

Metalworking fluids (MWFs) used in machining and rolling operations perform a number of functions, such as lubrication and cooling of workpiece, reduction of tool wear, improvement of surface finishing, and increase of tool life. Typically, they are oil-in-water (O/W) emulsions that become less effective after use, due to thermal degradation and contamination by substances in suspension or dissolved. Once they lose their functionality they have to be replaced, generating large volumes of oily effluents. These waste emulsions should be treated before their discharge in order to comply with environmental policy, due to their high organic content [1].

For each kind of effluent a specific oil/water separation process is used, depending on physical nature of the oil, total oil content, and chemical nature of other components. The most common treatment methods are chemical destabilization (coagulation/flocculation), electrocoagulation [2], centrifugation [3,4], membrane processes [5–10], and vacuum evaporation [11,12]. However, the combination of two or more of these separation techniques is required in many cases to obtain higher separation efficiencies. Methods such as membrane hybrid processes [13–16], membrane biological reactors [17], and destabilization/evaporation integrated processes [18] have shown successful results for the treatment of MWFs. Moreover, hybrid processes also allow obtaining high quality final effluents that could be suitable for several applications, such as process water or O/W emulsion reformulation. It has been reported that ultrafiltration permeates can be used for MWF reformulation, obtaining O/W emulsions with similar interfacial properties than the original one, taking into account their surfactant content [19].

The study and optimization of this kind of hybrid processes generally imply a high number of long-term experiments and a suitable experimental design should be very useful. Therefore, a statistical analysis (*i.e.* ANOVA) is required to find the most influencing factors and the optimum operating conditions. The number of experiments can be reduced by using the 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 factorial fractional design using orthogonal arrays (OA) and it is recommended for long or cumbersome experiments. It has been applied to microfiltration [20,21], ultrafiltration [22–26], nanofiltration [27,28], and reverse osmosis [29,30].

In this work a hybrid process, based in coagulation/centrifugation and ultrafiltration (UF) with tubular ceramic membranes, was studied for the treatment of an O/W emulsion prepared with a commercial MWF. A coagulant salt, $CaCl_2$, was used as destabilization agent in the coagulation/centrifugation stage as reported in previous works [31–33]. The aqueous phase from centrifugation was fed into an UF stage using tubular ceramic membranes. The main objective was to estimate the optimum operating conditions for this hybrid process. Taguchi experimental design was applied and experimental results were analyzed using a statistical analysis of variance (ANOVA). Furthermore, the contribution of different factors was evaluated and compared with a similar hybrid process performed with flat ceramic UF membranes [32].

2. Materials and methods

2.1. Emulsion formulation

An oil-in-water (O/W) emulsion was formulated from Besol 5, a commercial oil concentrate (Brugarolas Co., Spain). The precise formulation of the oil concentrate is proprietary, but it contains a mixture of mineral oils and several additives, such as emulsifiers, stabilizers, biocides, and corrosion inhibitors. This emulsion was selected because of its long-term stability and its common use in Spanish workshops for a wide range of applications, such as cutting, drilling, or grinding processes [31]. This oil was dispersed (1 wt %) in distilled water using a rotor-stator homogenizer Miccra D-9 (ART, Germany), at 16000 rpm for 5 min. 8 L of O/W emulsion were prepared in two batches of 4 L. Formulated O/W emulsion had a pH value of 9.4, with a chemical oxygen demand (COD) of 25745 mg/L, a conductivity of 42.5 mS/cm, and a turbidity higher than 2000 NTU. Furthermore, its mean oil droplet size was 0.26 μm, with a surface tension of 31.2 mN/m, and a zeta potential of –71 mV.

2.2. Destabilization/centrifugation

Anhydrous $CaCl_2$ (reagent grade, Panreac Química S.A., Spain) was used as coagulant salt for emulsion destabilization, with concentrations according to the trial conditions planned in the experimental design. Furthermore the emulsion was heated for 30 min in a thermostatic bath for destabilization enhancement.

The destabilized emulsion was then centrifuged (Kubota 6300, Japan) for 15 min at 4500 rpm. Supernatant was removed and the remaining aqueous phase was further separated in a funnel and sent to the ultrafiltration stage. These operating conditions were selected taking into account previous studies where the same salt was used as demulsifying agent in hybrid membrane processes [31–33].

The zeta or electrokinetic potential is often a useful parameter for studying the emulsion stability since it reflects the electrostatic interactions of moving oil droplets [33]. Zeta potential of the initial emulsion was –71 mV, which indicated high stability due to electrostatic repulsions between oil droplets. Most O/W emulsions, like the one used in this study, are stabilized by surfactants that generate an electrostatic charge on the surface of the oil droplets. A double layer of counter-ions in solution builds up around the droplet and surface charges repel each other, making the emulsion stable. Calcium ions, resulting from $CaCl_2$ addition, compressed the electrical double layer and reduced repulsions among oil droplets, making them to coalesce and thus increasing mean droplet size and decreasing zeta potential [34].

Fig. 1a gives zeta potential and surface tension values as a function of coagulant concentration. Zeta potential measurements were made in a Zetasizer Nano ZS apparatus (Malvern Instruments Ltd., UK), whereas surface tension was determined following the Du Noüy’s platinum ring method using a Sigma 700 tensiometer (KSV Instruments Ltd., Finland). It is observed in Fig. 1a that coagulant addition produced a large reduction of absolute zeta potential values until a plateau around neutrality was reached. Similar results have been reported in the literature [35]. A remarkable decrease in surface tension value was also observed as coagulant concentration increased, likely due to the free oil film formed at the emulsion surface after coalescence took place.

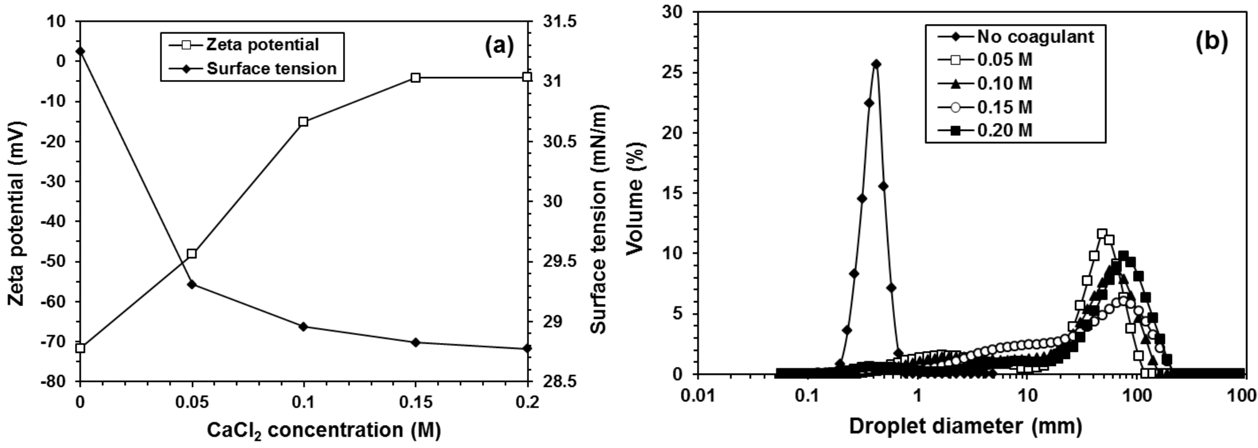

Figure 1. Effect of $CaCl_2$ addition on (a) zeta potential and surface tension, and (b) droplet size distribution of initial O/W emulsion.

Changes in droplet size distribution, determined by laser-light scattering using a Mastersizer S long bench equipment (Malvern Instruments Ltd., UK), are depicted in Fig. 1b. It can be observed that the lack of electrostatic repulsions enhanced oil droplets coalescence, increasing the mean droplet size as $CaCl_2$ was added. However, only slight changes in oil droplet size were noticed for coagulant salt concentrations higher than 0.05 M, which is the critical coagulation concentration (CCC) for this emulsion [3,31].

2.3. Ultrafiltration

A tubular multichannel ceramic membrane (INSIDE CéRAM™, TAMI Industries, France), with an active layer of ZrO_2 supported on TiO_2 (cut-off = 300 kDa), was used for UF experiments. The membrane had three channels with a hydraulic diameter of 3.6 mm, an active area of 90 cm², 25 cm length, and 1 cm external diameter. Crossflow UF experiments were performed for 1 h in total recirculation mode (*i.e.* both permeate and concentrate were returned to the feed tank), using a pilot-scale device described in previous works [7,19]. Briefly, the feed solution (aqueous phase from centrifugation stage) was circulated by a centrifugal pump from a 15 L internally-cooled feed tank towards the membrane module. The circulating flow rate was measured using a flowmeter, while permeate flux was obtained by measuring the time to collect a certain volume of fluid. Feed flow rates and transmembrane pressures were monitored by flow control valves. The operating variables were kept constant during the experiment, and measured by pressure and temperature transducers.

2.4. Membrane cleaning procedure

Membrane cleaning was carried out in several steps. First, the membrane was rinsed with water till no turbidity was observed. In the second step a basic cleaning was performed during 20 min using 10 g/L of P3-Ultrasil 40 detergent (Ecolab, Spain) in hot water (40ºC). Next, it was rinsed with hot water for 20 min. An acid cleaning was then performed during 20 min using a solution containing citric (1.6 g/L) and nitric (1.8 g/L) acids (Panreac Química S.A., Spain). Finally, the membrane was rinsed with water for 10 min. Pure water permeate flux was measured before and after each experiment in order to check the membrane cleaning. A total permeate flux recovery was obtained after membrane cleaning for all trials performed in this study.

2.5. Permeate characterization

UF feed and permeate were characterized for each experiment by measuring the following quality parameters: pH, COD, conductivity, and turbidity. COD was determined using commercial cells (ref. 14541, Merck, Germany) with a similar procedure to EPA 410.4 (US Standard Methods 5220D) and ISO 15705. The final value was obtained with a colorimetric method using a spectrophotometer Spectroquant NOVA 60 (Merck, Germany). Conductivity and pH values were simultaneously determined using a combined measurement device Crison MM40 (Spain). Turbidity was measured with a HACH Ratio/XR turbidimeter (USA).

3. Results and discussion

3.1. Experimental design

Experiments were performed according to a planning based in Taguchi experimental design method. Permeate flux (J) was chosen as response and the influence parameters (factors) were transmembrane pressure (TMP), destabilization temperature (T), feed flow rate to the UF module (Q), and coagulant salt molar concentration (M). These four factors were applied in three levels, and they were combined according to a L_9 orthogonal array indicated in Table 1. Nine trials were carried out and all of them were replicated twice in order to avoid non-linearity effect of any of the factors [32].

Table 1. Experimental design planning for the L_9 array.

Trial	TMP (bar)	T (ºC)	Q (L/h)	M (mol $CaCl_2$/L)
1	1.0	20	400	0.10
2	1.5	40	800	0.10
3	2.0	60	600	0.10
4	1.0	40	600	0.15
5	1.5	60	400	0.15
6	2.0	20	800	0.15
7	2.0	40	400	0.20
8	1.5	20	600	0.20
9	1.0	60	800	0.20

The optimum conditions were those that enabled to reach the maximum permeate flux. However, the conditions for permeate COD minimization were also taken into account. This was determined by a statistical analysis of the results from Taguchi method, which used the signal-to-noise (S/N) ratio as statistical indicator. This parameter was obtained from quadratic mean standard deviation (MSD) of every response factor value (Y_i), defined by Eq. (1), where n is the number of observations (2 in this case). Since high permeate flux was preferred, “the larger the better” optimization criterion was chosen, so that S/N ratio value should be as high as possible [36]. According to this criterion, the statistical parameter S/N, defined through MSD value, was calculated as Eq. (2) shows.

$$\mathrm{MSD} = \frac{1}{n}\sum_{i=1}^{N} Y_i^2 \qquad (1)$$

$$\frac{S}{N} = -10\log_{10}\left(\frac{1}{\mathrm{MSD}}\right) \qquad (2)$$

S/N analysis allowed 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.

The behavior of permeate quality parameters with the different factors was also determined according to S/N ratio. The contribution of each factor was estimated by a statistical analysis of variance (ANOVA).

3.2. Permeate flux

Permeate flux, mean standard deviations and S/N ratios for both replications are shown in Table 2 for each experiment. An average S/N value was calculated for each level in order to compare the influence of the different factors on permeate flux. Their values are shown in Fig. 2.

Table 2. Measured permeate fluxes and calculated S/N ratios.

Trial	**TMP level**	**T level**	**Q level**	**M level**	**Run 1 (L/m²h)**	**Run 2 (L/m²h)**	**MSD**	**S/N**
1	1	1	1	1	68.2	68.5	4669	36.7
2	2	2	3	1	158.3	173.5	27581	44.4
3	3	3	2	1	198.2	195.4	38738	45.9
4	1	2	2	2	118.2	91.2	11142	40.5
5	2	3	1	2	159.9	145.1	23313	43.7
6	3	1	3	2	146.9	139.8	20560	43.1
7	3	2	1	3	242.9	254.9	61976	47.9
8	2	1	2	3	106.3	101.0	10752	40.3
9	1	3	3	3	106.6	149.9	16913	42.3

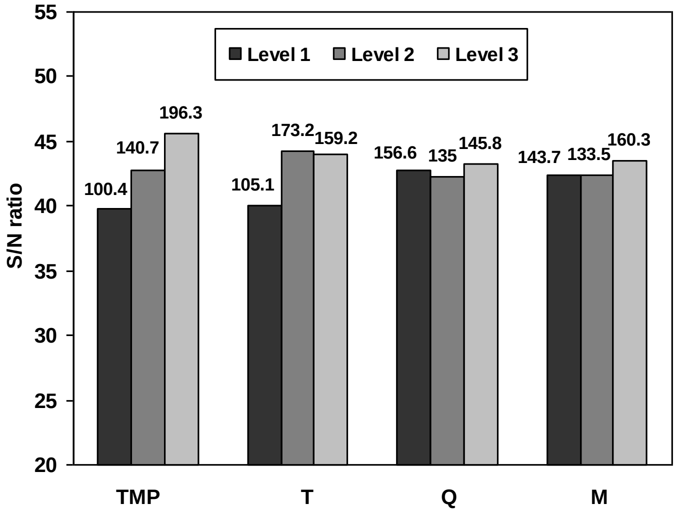

Figure 2. Variation of permeate flux (L/m^{2}h) and S/N ratio for different factors.

Fig. 2 shows that there were significant differences in S/N ratio (from 3 to 6 units) between level 1 and levels 2 and 3 for transmembrane pressure (TMP) and temperature (T). The difference was less significant for UF feed flow rate (Q) and coagulant salt concentration (M), being even negative for the UF feed flow rate. Therefore, TMP was the factor which most enhanced permeate flux, with an increase of 6 S/N ratio units from level 1 to level 3. Temperature was the second factor with higher influence. However, the difference between levels 2 and 3 was lower, in a range from 1 to 3 S/N ratio units for TMP, UF feed flow rate and coagulant salt concentration. This difference was even negative for temperature, being the value –0.4 S/N ratio units. Therefore, permeate flux decreased as temperature increased from level 2 to 3. It can be seen that any value higher than level 2 in any factor resulted in a very low increase in permeate flux from the UF step.

The difference between level 1 and levels 2 and 3 is probably due to the effect of experiment 1 (level 1 for every parameter) on S/N ratio, as the rest of experiments had similar magnitude. The low values for TMP, UF feed flow rate and temperature used in experiment 1 yielded low permeate flux, as shown in Table 2.

Values of the factors higher than level 2 imply low increases in permeate flux but higher energy costs for feed pumping and heating. Thus, level 2 is the most suitable for UF, using tubular ceramic membranes, of the aqueous phase resulting from coagulation and centrifugation of O/W emulsions. So, it enables to set optimum TMP, temperature, UF feed flow rate and coagulant salt concentration values.

In order to determine the relative importance of each factor, experimental results were evaluated using a statistical analysis of variance (ANOVA). The significance of each factor was assessed through the p-values obtained from the ANOVA F statistic. Table 3 shows ANOVA results, where degrees of freedom, sum of squares, mean square (variance), F-ratio, p-value, and the contribution percentage of each factor on response are given. Residual row in Table 3 refers to experimental error and also to error caused by uncontrollable factors (noise); it should be below 50%, otherwise results would not be reliable [32]. The contribution of the residuals in this case was about 3.4%, so the experimental error was not significant.

Table 3. ANOVA of experimental data for permeate flux as response.

Factor	**Degrees of freedom**	**Sum of squares**	**Variance**	**F-ratio**	**p-value**	**% Contribution**
TMP	2	27844	13922	76.1	0.00	57.3
T	2	15498	7749	42.3	0.00	31.9
Q	2	1390	695	3.80	0.06	2.9
M	2	2186	1093	5.97	0.02	4.5
Residual	9	1648	183			3.4

TMP and temperature were the parameters with the highest influence on permeate flux in the UF stage. An increase in TMP from level 1 (1 bar) to level 2 (1.5 bar) caused a high increase in permeate flux, whereas it was lower for level 3 (2 bar) because of concentration polarization [7]. Moreover, high temperatures promote emulsion destabilization by increasing the kinetic energy of oil droplets, and also influence viscosity and density of emulsion, leading to higher coalescence and centrifugation efficiencies [31]. The low influence of coagulant salt concentration on permeate flux is in good agreement with previous studies where it was reported that only for $CaCl_2$ concentrations around the CCC (0.05 M) there was an important permeate flux increase. This was due to coalescence caused by the lower electrostatic repulsions between oil droplets and also by their size increase [34], as shown in Fig. 1. Furthermore, an excess of $CaCl_2$ over the CCC caused only a reduction of the zeta potential absolute value without increasing the oil droplet size, which favored the deposition of oil droplets on the membrane surface and reduced permeate flux by increasing membrane fouling, as it was previously reported [32,33]. It can also be observed that all factors have a statistically significant effect on J at the 5% significance level, with p-values less than 0.05, except for UF feed flow rate (Q). These results agree with those obtained from S/N ratio analysis.

According to conventional ANOVA criterion, TMP would be the only parameter affecting permeate flux, although the influence of temperature should also be taken into account.

3.3. Permeate quality parameters

Table 4 shows the characterization data of permeates in the UF stage. Quality parameters were studied in terms of S/N ratio, except turbidity because of its high variability. In this case the optimization criterion was the minimization of permeate COD and conductivity, so that S/N ratio value should be as low as possible.

Table 4. Quality parameters of UF permeates.

Trial	COD (mg/L)	UF COD reduction (%)	Total COD reduction (%)	pH	Turbidity (NTU)	Conductivity (mS/cm)
1	667	48.1	97.4	7.4	1.5	13.0
2	798	52.3	96.9	7.9	1.3	16.1
3	874	40.8	96.6	8.3	0.3	16.1
4	1030	18.5	96.0	7.5	1.2	22.9
5	747	26.2	97.1	8.1	0.6	25.1
6	741	33.1	97.1	8.1	1.1	26.8
7	795	29.6	96.9	8.1	0.2	36.6
8	777	23.5	97.0	8.1	0.5	35.8
9	724	27.4	97.2	8.1	0.1	38.7

3.3.1. Chemical oxygen demand (COD)

The behavior of permeate COD values in terms of S/N ratio for the different operating factors is shown in Fig. 3a. A significant increase in S/N ratio was observed when coagulant salt concentration, UF feed flow rate and temperature were increased from level 1 to level 2, especially for those two last factors. However, a decrease in COD was observed increasing till level 3. The influence of TMP was less significant than other parameters, as its variation was lower than 0.5 units.

It should be stated that an important increase in permeate flux took place from level 1 to 2 for all factors. Therefore, the more liquid passing through the membrane, the higher the probability of carrying organic compounds through the membrane pores. However, the increase in permeate flux was lower when level 3 was reached: the higher destabilization efficiency of previous stages became more significant and permeate COD value decreased, as it was previously reported [33].

The highest permeate COD reduction took place at level 1 of coagulant salt concentration, although the lowest permeate COD values resulting from the global hybrid process were found at level 3, as it was seen in Fig. 3a. This was probably due to the low coagulant salt concentration that performed the destabilization far from the best conditions. It can be clearly noticed because a difference up to 6 units was found when comparing with the other two levels.

The increase of COD reduction between levels 2 and 3 was less significant for temperature and coagulant salt concentration, compared with the sharp decrease in S/N ratio for permeate COD in Fig. 3a.

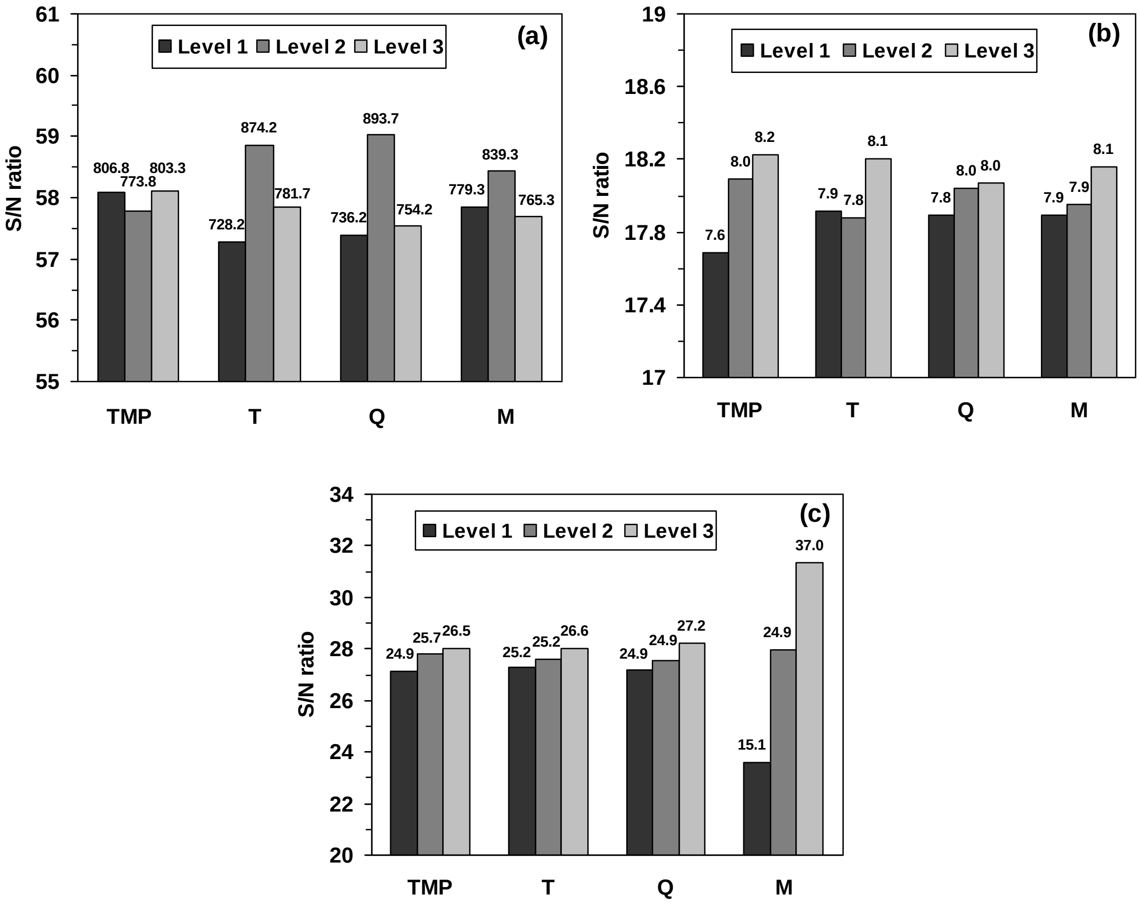

Figure 3. Variation of S/N ratio and (a) permeate COD (mg/L), (b) permeate pH, and (c) permeate conductivity (mS/cm) for different factors.

Oil removal throughout the proposed hybrid process can be seen in Fig. 4, where changes in visual appearance, from white O/W emulsion (feed) to translucent UF permeate, are evident. Thus, COD reduction took place mainly in previous coagulation/centrifugation stages, where an upper layer consisted of destabilized oil was obtained (Fig. 4) and removed prior to UF stage. The COD reduction compared with the initial O/W emulsion ranged between 96.0 and 97.4%, as shown in Table 4.

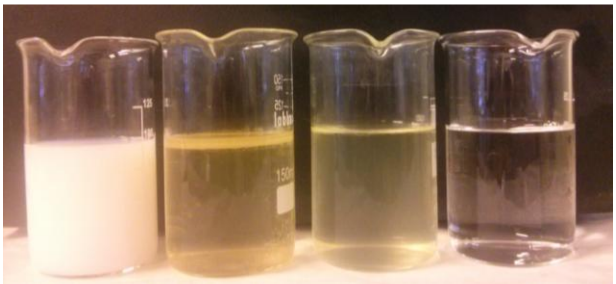

Figure 4. Photograph of O/W emulsion (feed), effluent after coagulation/centrifugation, feed to the UF process, and final UF permeate (from left to right, respectively).

3.3.2. pH and conductivity

Variations of pH in terms of S/N ratio for the different operating factors are shown in Fig. 3b. Values of pH for UF permeates increased from level 1 to level 3 for every factor. This S/N ratio increase ranged between 0.2 units for UF feed flow rate and 0.5 units for TMP.

Values of pH increased in a similar way as permeate fluxes, being these increases more significant for TMP and temperature. It agreed with the results of permeate flux in Fig. 2. Furthermore, the feed pH was more alkaline than pH values of permeates. Changes in permeate flux were also observed by other authors at pH higher than 10 for UF of O/W emulsions using modified polymeric membranes [26].

Coagulant salt concentration was the factor with the highest influence on permeate conductivity, as it can be seen in Fig. 3c. This can be explained because of the ions present in the permeate, that have passed through the membrane. S/N ratio increased in a similar order of magnitude as the salt concentration for the three levels.

An increase in conductivity was also observed increasing the level in other parameters, like it happened with pH.

3.3.3. Contribution of each factor

The contribution of each factor to quality parameters of permeate can be obtained using ANOVA as it was applied to permeate flux. Results are presented in Table 5.

Conductivity variations were exclusively caused by $CaCl_2$ concentration. Temperature and UF feed flow rate were the main parameters affecting permeate COD. Meanwhile, pH was mainly influenced by TMP and temperature, as permeate flux. These results can be considered statistically representative, as residuals were lower than 50%. However, the high contribution of the experimental error did not allow rejecting the null hypothesis (response factor unaffected by the other factors) for COD and pH. The influence of all factors followed the same trend as in S/N ratio analysis.

Table 5. Contribution (%) of each factor to quality parameters of permeate.

Factor	pH	Conductivity	COD
TMP	30.4	0.5	1.2
T	12.1	0.5	19.6
Q	3.2	1.4	26.7
M	7.2	92.5	5.5
Residual	47.1	5.1	47.0

3.4. Influence of membrane geometry in ultrafiltration stage

The contribution of the different factors was compared with previous results obtained in a hybrid coagulation-centrifugation/ultrafiltration process using flat ceramic membranes for the UF stage [32]. ANOVA was applied to the data and the relative contributions of each parameter to different responses were estimated. Flat ZrO_2 membranes with 90 mm of diameter and 56.3 cm^2 of area had the same cut-off (300 kDa) as tubular membranes. The comparison between both geometries is shown in Fig. 5.

Significant differences in the most influencing factors were observed between flat and tubular ceramic membranes. Permeate flux was affected in a similar way by temperature and UF feed flow rate for both geometries, as shown in Fig. 5a. However, the influence of transmembrane pressure was very high for tubular membranes, being less important for flat membranes. A similar behavior was observed for coagulant salt concentration, which had high influence for flat membranes, but low for tubular ones. Anyway, temperature was a key parameter in both cases and the effect of UF feed flow rate was not significant.

Fig. 5b shows that pH value of permeate from flat membranes was mostly affected by temperature instead of TMP, which was the most influencing parameter for tubular membranes. In both cases UF feed flow rate and coagulant salt concentration had low influence (<20%). However, Fig. 5c shows that TMP was the most important parameter affecting permeate conductivity for flat membranes, with a contribution higher than 80%. On the contrary, it had almost no influence for tubular membranes, where coagulant salt concentration determined permeate conductivity. This can be explained by the fact that experiments reported by Allende *et al.* [32] were performed using tap water for the emulsion preparation. Therefore, an increase in coagulant salt concentration would be buffered by the background conductivity.

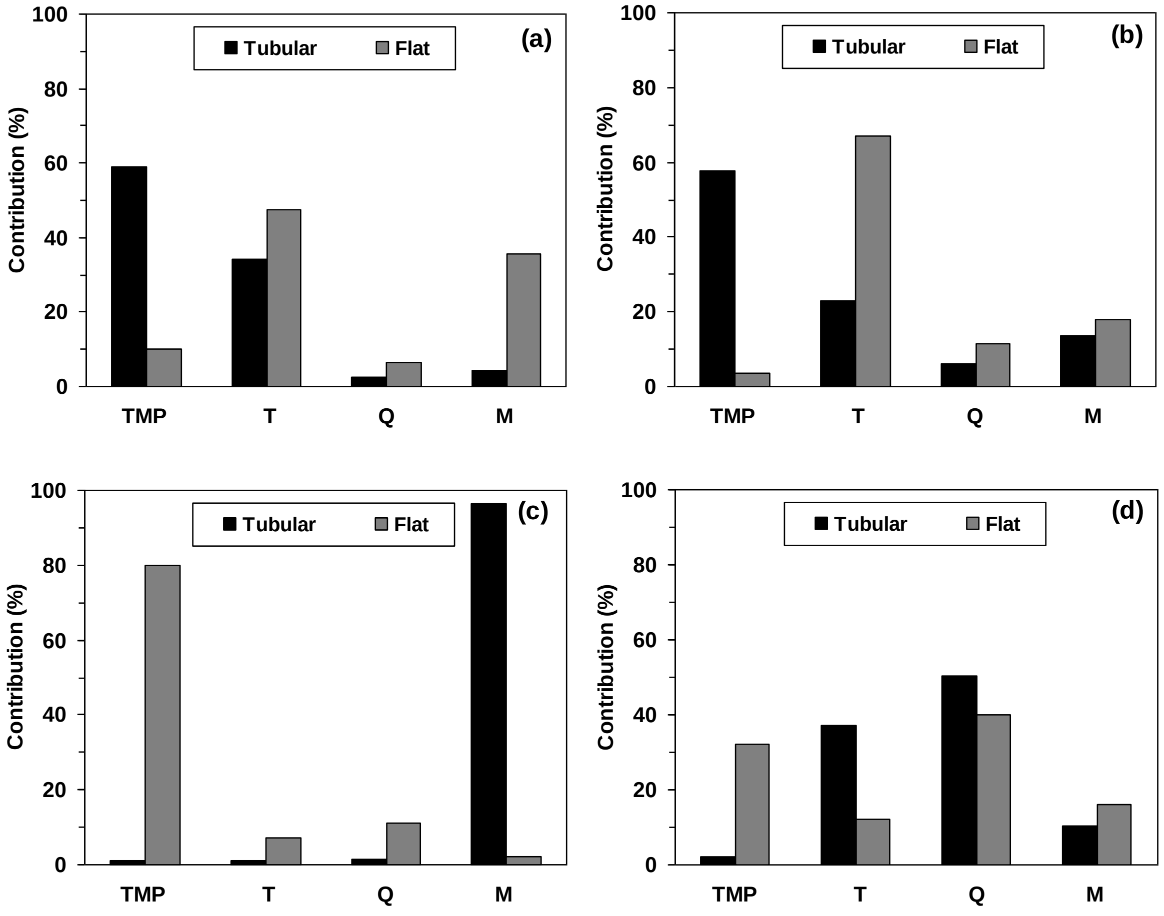

Figure 5. Contribution of each factor to different responses for tubular and flat ceramic UF membranes: (a) permeate flux, (b) permeate pH, (c) permeate conductivity, and (d) permeate COD.

Data shown in Fig. 5d agree with the fact that UF feed flow rate was the most influencing parameter on permeate COD for both flat and tubular membranes. A similar influence was found for coagulant salt concentration. However, the second parameter was different for both membranes. The COD value of permeate from flat membranes was mainly affected by TMP, while temperature had a similar effect when tubular membranes were used.

The different effect of TMP for both membranes might be related to the different geometry and values of the three UF flow rate levels (60, 90 and 120 L/h for flat membranes), and to the smaller filtration area. Furthermore, temperature control was more difficult for tubular membranes because of the larger volume of feed treated.

4. Conclusions

Taguchi method provides an excellent experimental design for time-consuming and cumbersome procedures with good statistical tools for solving multivariable processes. Therefore, several conclusions can be obtained from this work:

- A hybrid process of coagulation/centrifugation and further ultrafiltration with ceramic membranes for treating O/W emulsions from MWF yields COD reductions up to 97.4%. These UF permeates of high quality could be suitable for several applications, such as process water or O/W emulsion reformulation.
- Following the criterion of permeate flux, the most favorable conditions for ultrafiltration stage corresponded to level 2 (TMP = 1.5 bar, T = 40ºC, Q = 600 L/h, M = 0.15 mol/L). However, level 2 is not the optimum for other quality parameters, being level 3 (TMP = 2 bar, T = 60ºC, Q = 800 L/h, M = 0.20 mol/L) the most suitable if the criterion is permeate COD. Therefore, selection criteria should be previously established in order to find the optimum conditions as a balance solution between UF feed flow rate and permeate quality expressed as COD.
- Contribution of influence factors changes according to membrane geometry, being the effects of transmembrane pressure and temperature significantly different between flat and tubular ceramic membranes. This fact should be taken into account for subsequent process scale-up.

Nomenclature

ANOVA	analysis of variance
CCC	critical coagulation concentration (mol/L)
COD	chemical oxygen demand (mg/L)
J	permeate flux (L/m^{2}h)
M	coagulant salt concentration (mol/L)
MSD	mean standard deviation
n	number of observations
NTU	nephelometric turbidity units
Q	feed flow rate to ultrafiltration stage (L/h)
S/N	signal-to-noise ratio
T	destabilization temperature (ºC)
TMP	transmembrane pressure (bar)
Y	response factor value

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*Graphical Abstract