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Two-level factorial experiments in the ultrafiltration of oil–water emulsions

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ABSTRACT

The paper evaluates the influence of selected process variables on the separation of oil from an oil-in-saline water emulsion performed using ultrafiltration. The experiments were carried out according to the two-level factorial design with four factors 2^4. The following variables were taken into account: flow velocity (4, 5 m/s), transmembrane pressure (TMP) (0.1, 0.2 MPa), salt concentration (1.0%, 3.5%), and oil concentration (500, 1,500 ppm). The separation of the emulsion was characterized using the initial permeation flux J_0, the steady-state permeation flux J_{SS}, and the oil rejection coefficient *r*. Based on the experimental results, a mathematical description of all the examined variables was proposed in the form of linear and interaction relationships. Such an equation using normalized variables enabled the influence of all process variables on the output quantities, that is, J_0, J_{SS}, and *r*. It was found that TMP had the greatest impact on the efficiency of the process. The presence of salt caused a slight decrease in J_0 and J_{SS}, similar to as the presence of oil.

Keywords: Oil-in-saline water emulsion; Ultrafiltration; Factorial experiment design; Ceramic membrane

1. Introduction

Membrane separation has been widely used in wastewater treatment. It should be noted that mostly waste liquid streams are multicomponent systems and their separation/treatment is a significant technical issue. The presence of inorganic salts in wastewater complicates the removal of contaminants since it adversely influences on the efficiency of membrane separation. The reported researches focus mostly on the treatment of wastewater from the petrochemical industry and refineries as well as oily streams generated by ships (bilge water and ballast water) [1–3]. The main advantage of applying membrane separation techniques directly on ships complies with the environmental standards and significant reduction of the amount of retentate, which must then be utilized on the board of the ship or on a land.

Therefore, extensive researches are carried out on the ultrafiltration (UF) of oil-in-water emulsions using ceramic membranes [4–6] and on the pore blocking mechanism, including fouling [7,8]. The primary objective of the current studies is to achieve possibly high permeation fluxes and possible high oil rejection. On the other hand, the research should be sensibly planned. The accepted scientific method can be evaluated through the following steps: (1) the problem definition; (2) an appropriate theory, idea, or model proposal; (3) a collection of data to test the theory; (4) results analysis; and (5) data interpretation and conclusions.

The aim of the paper is to methodically and clearly explain the method for planning the experiments. Such methods are often used in the form of appropriate software without the need of a specific approach selection.

In the study, the full two-level factorial design of experiments was selected. This method belongs to the category that may be termed the "black box" approach or the response surface methodology. However, the accepted method is simple, does not require a lot of experimental labor and provides valuable additional information on the process.

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After normalizing the variables, it not only helps to estimate the influence of the respective variables on the process but also presents the quantitative relations. Additionally, the interactions between the investigated variables can be determined.

This paper attempts to evaluate the influence of salt present in the oil-in-water emulsion on the efficiency of UF. Several process parameters such as the linear flow velocity (LFV), transmembrane pressure (TMP), concentration of salt (C_{NaCl}) and oil (C_{oil}) in the emulsion were considered as the key factors. The influence of these parameters was evaluated in the experiments planned according to a two-level factorial design.

2. Planning of the experiments

The experimental design can be successfully used in studies related to wastewater treatment using membrane techniques. The Taguchi method is very often used for planning the experiments [9–12]. Matosa et al. [13] planned their experiments using the Taguchi method to determine the influence of four parameters, that is, TMP, feed flow rate, temperature, and salt concentration, each having three levels, on the UF performance. Salahi et al. [14] proposed the Taguchi approach for five controllable factors, each at four levels, to plan the minimum number of experiments. Many Taguchi designs are based on factorial designs (two-level designs and Plackett and Burman designs, as well as factorial designs with more than two levels). Taguchi experimental designs, often called orthogonal arrays, consist of a set of fractional, factorial designs, which ignore interaction and focus on the main effect estimation. However, the Taguchi method is used for specifying, which input quantities are important to the process and which are merely noise. This method does not provide a mathematical description. The factorial and composite designs are used for the experimental optimization.

In case of UF and controllable process variables, the factorial design may be selected and it was applied in this paper. The methodology of planning as well as mathematical description and statistical evaluation used in this work is presented below.

Let us assume the following form of a function describing the experimental data:

$$y = \sum_{k=1}^{M} \alpha_k \Phi_k(x) \tag{1}$$

where α_k – unknown coefficients, $\phi_k(x)$ – a set of assumed functions, and y – results of the process, for example, concentration of the product and yield.

The coefficients α_k should be calculated from the experimental data. To fulfill this task, the objective function is constructed:

$$Q(\alpha_1 \cdots \alpha_M) = \sum_{i=1}^{M} \left[y_i - \sum_{k=1}^{M} \alpha_k \Phi_K(x_i) \right]^2 \tag{2}$$

The function given by Eq. (2) is convex, which means that to find its global minimum with respect to α_k, the first derivative of Q must be set to zero. This leads to a system of M linear equations with M unknown coefficients α_k. Eqs. (1) and (2) represent one of the possible approaches to the approximation theory.

It is possible to simplify this system of equations by making it orthogonal, which requires the fulfillment of the following condition:

$$\sum_{i=1}^{p} \Phi_k(x_i)\Phi_j(x_i) = \begin{Bmatrix} 0 & for\ k \neq j \\ \neq 0 & for\ k = j \end{Bmatrix} \tag{3}$$

where P – number of experimental points.

This can be done by selecting appropriate reference points x_i (location of experiments). The orthogonal system is usually used in different variants of experimental planning [15–17].

The orthogonal system allows the coefficients α_k to be easily calculated. The system of linear equations is then reduced to the formula given by Eq. (4).

$$\alpha_k = \frac{\sum_{i=1}^{P} y_k \Phi_k(x_i)}{\sum_{i=1}^{P} \Phi_k^2(x_i)} \tag{4}$$

The minimum number of experiments can be achieved if the number of levels of each factor can be reduced to two. The number of experiments in the two-level factorial design is equal to

$$P = 2^n \tag{5}$$

where n – number of factors (independent variables).

Considering that the orthogonal plan gives the opportunity to easily calculate the unknown coefficients, the two-level factorial design is also orthogonal.

To be able to compare the influence of factors on the effect of the process, it is necessary to normalize the factors (independent variables). The simplest way to do it is to use Eq. (6).

$$Z_i = \frac{x_i - x_{0i}}{\Delta x_{0i}} \tag{6}$$

where x_{0i} – central point of the design for variable x_i, Δx_{0i} – initial (assumed) increment of x_i.

According to the two-level factorial design, the experiments should be performed at the following points:

$$x_i = x_{i0} + \Delta x_{0i}; \quad z_i = +1 \tag{7}$$

$$x_i = x_{i0} - \Delta x_{0i}; \quad z_i = -1 \tag{8}$$

The mathematical description of the experimental data in terms of normalized variables z_i and the use of an orthogonal system of functions depends on the number of independent variables. The simplest method of description is provided by the following formula:

$$y = \alpha_0 + \alpha_1 z_1 + \cdots + \alpha_n z_n + \alpha_{1,2} z_1 z_2 + \cdots + \alpha_{n,n} z_n z_n \tag{9}$$

Eq. (9) is a linear combination of single-variable and double-variable functions. Such a formula enables the assessment of the influence of single variables as well as the interactions between any two variables. If necessary, Eq. (9) can be extended by introducing additional combinations of three or more (up to n) variables. It is possible to make the system orthogonal again.

Eq. (9) may also be used for predicting the process results, assuming the values of variables different from the experimental ones.

3. Rejection of oil from saline using UF

3.1. Factorial plan of experiments

The input variables are the process variables that can be manipulated in a controllable manner. In the investigated process of UF separation of oil from the oil-in-saline water emulsion, the input variables undoubtedly include the flow velocity LFV (4, 5 m/s), TMP (0.1, 0.2 MPa), salt concentration C_{NaCl} (1.0, 3.5%), and oil concentration C_{oil} (500, 1,500 ppm). On the other hand, the effects of UF can be assessed by determining the initial permeation flux J_0, steady-state permeation flux J_{SS}, and oil rejection coefficient r.

The two-level factorial design of experiments, in which the central point x_{0i}, the initial increment Δx_{0i}, and real values of process variables given in Table 1, was accepted.

3.2. Experimental

The UF tests were performed using a pilot installation shown in Fig. 1 equipped with a commercial 23-channel TiO_2/ZrO_2 ceramic membrane of a length of 1.178 m, filtration area of 0.35 m^2, and cutoff of 300 kDa, operated at the specified process conditions (temperature 20°C).

The feed solutions used in the tests were mixtures of water, oil, and sodium chloride.

Hydraulic oil HYDROL L-HL 46 (Orlen, Poland) was used in the homogenization process. 10 dm^3 of feed, corresponding to the minimum volume recommended by the manufacturer of the system, was prepared using a VCX-500 ultrasonic processor (Sonics & Materials, Inc., USA) at the following operating parameters: frequency of 20 kHz, vibration amplitude of peak-to-peak resonator 124 µm, resonator diameter 13 mm, temperature 20°C, and scattering time 5 s. The average size of the emulsion drops was 50 µm. The emulsions containing 500 and 1,500 ppm of oil and 0.0%, 1.0%, and 3.5% NaCl were obtained.

The UF installation was operated with recirculation of the retentate and the permeate. During each UF run samples of the feed, the permeate and the retentate were collected at specified time intervals. The duration of each experiment was 60 min. The samples were analyzed using the turbidimetric method (TN-100, Eutech Instruments, Netherlands). Each experiment was repeated twice. After each run, the membrane module and the UF unit were chemically cleaned according to the procedure recommended by the manufacturer. The cleaning was continued until the membrane reached the hydraulic permeability characteristic of a clean membrane.

Table 1
Two-level factorial design for membrane ultrafiltration

	Factor name	Unit	x_{0i}	Δx_{0i}	$x_{0i} - \Delta x_{0i}$	$x_{0i} + \Delta x_{0i}$
1.	LFV	m/s	4.5	0.5	4.0	5.0
2.	TMP	MPa	0.15	0.05	0.1	0.2
3.	C_{NaCl}	%	2.25	1.25	1	3.5
4.	C_{oil}	ppm	1,000	500	500	1,500

Fig. 1. Laboratory UF unit with a tubular ceramic membrane: 1 – feed tank, 2 – pump, 3 – tubular ceramic membrane, and 4 – flowmeter.

The described UF experimental procedure helped to determine the permeation flux as a function of time.

The volumetric permeation flux was calculated with Eq. (10):

$$J_V = \frac{V}{A \cdot t} \quad (10)$$

J_V – volumetric permeation flux (m^3/(m^2 s)), V – permeate volume (m^3), A – membrane area (m^2), and t – time (s).

The rejection coefficient r was calculated using Eq. (11).

$$r = \left(1 - \frac{C_P}{C_R}\right) \times 100\% \quad (11)$$

C_R – oil concentration in the retentate (mg/dm^3), C_P – oil concentration in the permeate (mg/dm^3).

The exemplary changes in the volumetric permeation flux with time for three different salt concentrations are shown in Fig. 2. The chart was drawn using the following variables: LFV = 4 m/s, TMP = 0.05 MPa, salt concentration equal to 0.0%, 1.0%, and 3.5%, and C_{oil} = 500 ppm (see Table 4 – data 1, 3, and 5). It was observed that greater concentrations of salt in the feed solution resulted in lower values of the initial J_0 and steady-state J_{SS} permeation fluxes. The change of the flux with time was significantly influenced by the pore blocking mechanism; however, this aspect of the UF process

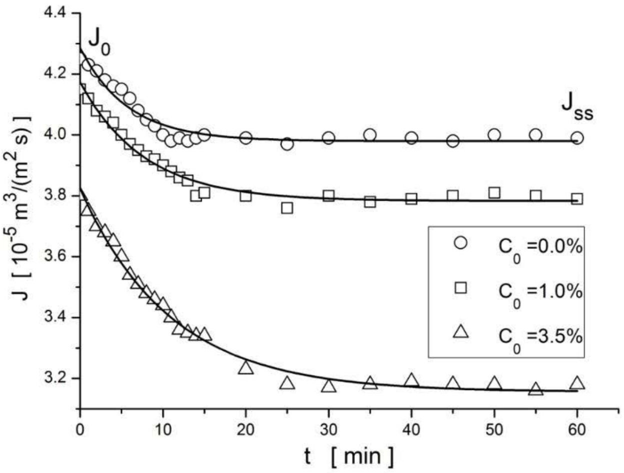

Fig. 2. Volumetric flow rate versus time at different salt concentrations and C_{oil} = 500 ppm in the feed solution.

was outside the scope of this study. Based on the experiments performed in this research, three characteristic quantities of the process, that is, the initial flux, the steady-state flux, and the rejection coefficient were confirmed.

In order to estimate the influence of the process variables on the UF separation, a two-level 2^4 experimental design had to be implemented, in which (*z*1) represented the flow velocity over the membrane LFV, while (*z*2), (*z*3), and (*z*4) stood for the TMP, the concentrations of salt C_{NaCl}, and oil C_{oil} (all variables normalized). The design of experiments ensuring orthogonality and successful evaluation of the influence of the respective process variables and their interactions is given in Table 2. Considering that the number of factors equaled to 4, the number of independent experiments amounted to 16. The second column of Table 2 gives the code that helps to build the two-level full factorial design. All experiments were performed on the previously cleaned ceramic membrane until its high initial permeability was obtained.

4. Analysis and interpretation of results

Using the designed matrix and the data given in Table 2, the coefficients of Eq. (9) were calculated, which was easier due to the orthogonality of the design with regard to Eq. (9). This equation, using normalized variables and appropriate coefficients, enabled to assess the impact of all process variables on the examined outputs, that is, J_0, J_{SS}, and *r*.

Consequently, the following coefficients were derived from the calculations:

For the initial flux J_0:

$\alpha_0 = 10.481$;
$\alpha_1 = -0.30625$; $\alpha_2 = 3.2437$; $\alpha_3 = -0.14375$; $\alpha_4 = -0.34375$
$\alpha_{12} = -0.16875$; $\alpha_{13} = 0.11875$; $\alpha_{14} = -0.08125$
$\alpha_{23} = 0.04375$; $\alpha_{24} = -0.03125$; $\alpha_{34} = 0.13125$

For the asymptotic flux J_{SS}:

$\alpha_0 = 9.4865$
$\alpha_1 = -0.18244$; $\alpha_2 = 2.9914$; $\alpha_3 = -0.34757$; $\alpha_4 = -0.56938$
$\alpha_{12} = -0.21396$; $\alpha_{13} = 0.052381$; $\alpha_{14} = 0.013769$
$\alpha_{23} = -0.035181$; $\alpha_{24} = -0.13402$; $\alpha_{34} = 0.13192$

The values of the rejection coefficient *r* in the respective experiments did not differ significantly; therefore, applying the factorial design was not advised. The use of the mean value of *r* calculated from the performed experiments (*r* = 97.1%) was sufficient from the statistical point of view. As the rejection coefficient is influenced mainly by the sort of a membrane, which small changes in its value in relation to the process variables could be explained.

Table 2
Two-level factorial design of experiments for $n = 4$

No.	Code	*z*1	*z*2	*z*3	*z*4	J_0 (10^{-5} m³/m²/s)	J_{SS} (10^{-5} m³/m²/s)	*r* (%)
1.	(1)	–1	–1	–1	–1	7.9	7.52	97.4
2.	a	+1	–1	–1	–1	7.5	7.07	98.0
3.	b	–1	+1	–1	–1	14.8	14.02	97.8
4.	ab	+1	+1	–1	–1	14.2	13.53	98.8
5.	c	–1	–1	+1	–1	7.5	6.69	98.2
6.	ac	+1	–1	+1	–1	7.3	6.44	98.6
7.	bc	–1	+1	+1	–1	14	12.78	98.1
8.	abc	+1	+1	+1	–1	13.4	12.40	96.0
9.	d	–1	–1	–1	+1	7.2	6.04	98.1
10.	ad	+1	–1	–1	+1	7.1	6.60	91.3
11.	bd	–1	+1	–1	+1	14.3	12.70	98.6
12.	abd	+1	+1	–1	+1	12.0	11.20	94.4
13.	cd	–1	–1	+1	+1	6.9	5.60	98.8
14.	acd	+1	–1	+1	+1	6.5	6.00	96.8
15.	bcd	–1	+1	+1	+1	13.7	12.00	95.0
16.	abcd	+1	+1	+1	+1	13.4	11.20	98.1

If the coefficient α_k is positive, an increase in the selected variable causes an increase in the value of J_0 and J_{SS}. If α_k is negative the value of J_0 and J_{SS} decreases. The higher the absolute value of α_k, the greater the influence on J_0 and J_{SS}.

Given the values of all coefficients, the following conclusions can be drawn:

- an increase in the LFV causes a decrease in the flux (J_0 and J_{SS}),
- the TMP has the greatest influence on the efficiency of the process,
- an increase in the salt concentration causes a slight decrease in J_0 and J_{SS}, however, the influence on J_{SS} is greater,
- the earlier relation is also valid for the oil concentration,
- significant interactions between the respective variables take place, when combining the linear velocity with the TMP (negative effect) as well as the salt concentration with the oil concentration (positive effect),
- when J_0 is considered, the positive interaction effect occurs also between the flow velocity and the C_{NaCl}.

The approximation results were statistically evaluated using the determination coefficient R^2 (Eq. (12)) and root mean square error δ (Eq. (13)). The results of evaluation are summarized in Table 3.

$$R^2 = 1 - \frac{\sum_{i=1}^{P}\left(y_{i\exp} - y_{i\text{cal}}\right)^2}{\sum_{i=1}^{P}\left(y_{i\exp} - y_m\right)^2} \tag{12}$$

$$\delta = \sqrt{\frac{\sum_{i=1}^{P}\left(y_{i\exp} - y_{i\text{cal}}\right)^2}{P}} \tag{13}$$

A series of additional experiments was performed under conditions different from those used for the purpose of 24 design. The data is gathered in Table 4.

Table 3
Statistical evaluation of the approximation

	R^2	δ (10^{-5} m³/(m² s))
Linear components of J_0	0.985	0.396
Linear components + interactions of J_0	0.992	0.296
Linear components of J_{SS}	0.986	0.367
Linear components + interactions of J_{SS}	0.995	0.222

Table 4
Process variables in the additional experiments

	Factor	Unit	Data 1	Data 2	Data 3	Data 4	Data 5	Data 6
1.	LFV	m/s	4.0	6.0	4.0	6.0	4.0	6.0
2.	TMP	MPa	0.05	0.20	0.05	0.20	0.05	0.2
3.	C_{NaCl}	%	0.0	0.0	1.0	1.0	3.5	3.5
4.	C_{oil}	ppm	500	500	500	500	500	500

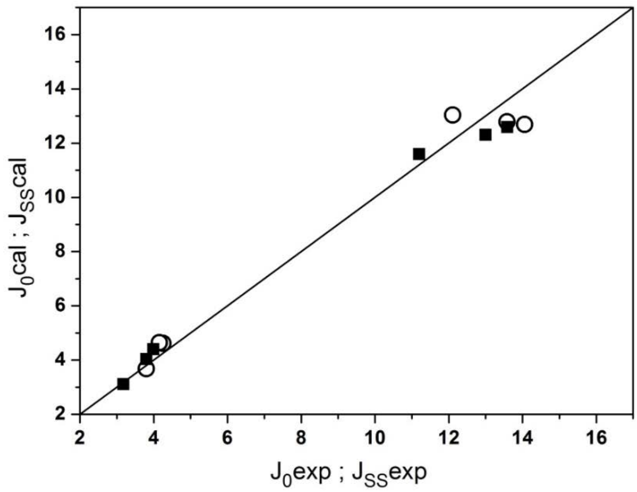

Fig. 3. The comparison of fluxes J_0 and J_{SS}, experimental and calculated with equations derived from the two-level factorial design (J_0 – open circles; J_{SS} – solid squares).

The coefficients α_i and α_{ij} obtained for J_0 and J_{ss} were used in calculations concerning the experimental data given in Table 4. The comparison of the experimental and calculated values of J_0 and J_{SS} is shown in Fig. 3.

Slightly better agreement between the theoretical predictions and the experimental data was obtained for J_{SS} in comparison with J_0. However, in both cases, the results were particularly convergent for small fluxes. The determination coefficient and the root mean square error calculated for J_0 and J_{SS} were R^2 = 0.971, δ = 0.791 and R^2 = 0.985, δ = 0.558, respectively.

5. Summary

In the study, the two-level factorial design of experiments was employed to evaluate the influence of four variables on the permeation flux and the rejection coefficient in the UF process. The selected variables included linear velocity, TMP, C_{NaCl} as well as concentration of oil in an oil-in-water emulsion. The study comprised of 2^4 independent experiments.

Based on the experimental data, a mathematical description of the process was developed giving the initial permeation flux and the asymptotic permeation flux as functions of the assumed process variables after normalization.

The resulting equations helped to evaluate the influence of those variables and their interactions on the investigated output quantities. The mathematical relations obtained in the study can further be used for predicting the effects of UF performed under different conditions.

Symbols

A	—	Filtration area, m²
C_{NaCl}	—	Salt concentration, %
C_{oil}	—	Oil concentration, ppm
C_P	—	Oil concentration in the permeate, ppm
C_R	—	Oil concentration in the retentate, ppm
J_0	—	Initial permeation flux at t = 0, m³/m²/s

J_{SS} — Steady-state permeation flux, $m^3/m^2/s$
J_V — Volumetric permeation flux, $m^3/m^2/s$
LFV — Linear flow velocity, m/s
n — Number of factors (independent variables), –
P — Number of experimental points, –
r — Rejection coefficient, –
t — Time, s
TMP — Transmembrane pressure, MPa
x_{0i} — Central point of factorial design for variable x_i, unit of the variable
Δx_{0i} — Initial (assumed) increment of x_i, unit of the variable
y — Results of, for example, J_0, J_{SS}, and r
y_{ical} — Calculated values of, for example, J_0, J_{SS}, and r
y_{iexp} — Experimental values, for example, J_0, J_{SS}, and r
y_m — Mean value of, for example, J_0, J_{SS}, and r
α_k — Unknown coefficients
$\phi_k(x)$ — Assumed function

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