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The use of factorial design for modeling membrane distillation

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ABSTRACT

A two-level factorial experimental design was used to investigate the influence of the main operating parameters on evaporation flux and soluble solid content of apple juice during concentration through osmotic distillation (OD) and membrane distillation (MD) processes. The factorial models have been obtained from experimental design to study all interactions among the considered parameters (osmotic agent concentration (0–65% $CaCl_2$), flow rate (10–30 L/h) and temperature difference between feed and osmotic agent (10–30 °C)) and validated statistically by analysis of variance (ANOVA). For both responses, the osmotic agent concentration was the most influential factor. The magnitude of the main influence of $CaCl_2$ concentration was followed by the temperature difference and flow rate, respectively. The analysis of the experimental responses revealed that $CaCl_2$ concentration and temperature difference had significant interactive effects on evaporation flux. All interactions between the studied parameters were significant in the case of soluble solid content at the 99% confidence level. Although the interaction terms have significant effects, their levels were only a small amount compared to linear effects. The predicted responses were compared with the experimental ones. In general, the predicted values were in reasonable agreement with the experimental data, further confirming the very good prediction ability of the models.

1. Introduction

Fruit juices are beverages of high nutritional value since they contain high levels of minerals, vitamins and other beneficial components for human health. In order to obtain chemical and microbiological stability as well as reducing the transport, packaging and storage costs, fruit juices are generally concentrated. During the concentration process, the water should be removed selectively in order to obtain a product with an appearance and taste as close as possible to the original juice. However, multi-stage vacuum evaporation processes are generally used for concentration, resulting significant loss of aroma compounds, irreversible alteration of nutritional value and color changes due to high operation temperatures [1]. One of the solutions to this problem is the use of alternative processes that avoid high temperatures of operation, such as the membrane processes. Membrane distillation (MD) and osmotic distillation (OD) are well known methodologies having great potential as concentration processes carried out at atmospheric pressure and temperatures near the ambient temperature [2–4]. Both operations involve microporous hydrophobic membranes that are in contact with fruit juices at different temperatures and/or compositions. In both processes, a microporous hydrophobic membrane is in contact with feed solution on one side. The driving force for the water transport through the gas phase immobilized within the pores is a water vapor pressure difference related with the water activity differences between the juice and an osmotic agent, in the case of the OD process, or by a temperature difference for the MD process [5,6].

Response surface methodology (RSM), a collection of statistical and mathematical techniques, is a useful tool for development, improvement, and optimization of processes. It is used to examine the relationship between one or more response variables and a set of quantitative experimental variables or factors. Meanwhile, use of RSM has gained prominence in food process design and optimization owing to the ease of operation, reliability and reproducibility of the model parameters. Nowadays, factorial designs have proved their usefulness, and are widely used in the statistical planning of experiments to obtain empirical models relating process response to process factors. Khayet et al. [7] studied RSM in direct contact membrane distillation using salt (NaCl) aqueous solutions and investigated the operating factors, namely, the stirring rate, feed temperature, and solute concentration. However, the effects of main process parameters involved in OD and MD during concentration of fruit juices need to be carried out in detail. The objective of this work is to evaluate the effects of temperature difference between the feed and permeate side of the membrane, concentration of the osmotic agent ($CaCl_2$ solution) and flow rate

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on the evaporation flux (J) and total soluble solid concentration (SSC) of apple juice reached after a predetermined period of membrane/osmotic distillation process using factorial design.

2. Materials and methods

2.1. Experimental set-up

The concentration process was performed using a laboratory-size membrane module (MD 020 CP 2N, Microdyn, Germany) having 40 polypropylene capillaries with 2.8 mm outer and 1.8 mm inner diameter (Table 1).

In the direct contact membrane distillation process, the apple juice, with an initial concentration of 12°Brix, was pumped in the tube side and the deionised water was recirculated in the shell side of the membrane in a countercurrent mode by using peristaltic pumps (Heidolph PD 5001, Germany). The temperature difference imposed between the feed and the permeate side of the membrane at the inlets using two heat exchangers (Lauda E100, Germany) and in every case, the inlet temperature of the juice was maintained constant at 10 ± 1 °C. The temperatures were measured at the inlets and at the outlets of the membrane module using type J thermocouples. Samples of the apple juice were taken over time and the respective concentration was determined using a digital refractometer (Atago PAL-3, Tokyo, Japan).

The same experimental procedure was used in the osmotic membrane distillation process, however, in this case, calcium chloride dihydrate (Merck, Germany) was used as stripping solution. The initial weight of the stripping solution was three times higher compared to that of the juice in order to prevent a significant dilution with consequent decreasing of the driving force during the process. After each experimental run, the membrane module was cleaned by a five step cleaning process. First, both sides of the membrane were rinsed with deionised water. Then 1% (w/w) NaOH solution was circulated for 1 h at 30 °C. After a short rinsing with deionised water, a 1% (w/w) citric acid solution was circulated for 1 h at 30 °C. Finally, the circuit was rinsed with deionised water. The efficiency of the cleaning was ascertained by checking the membrane performance parameters (water flux and solute rejection). The evaporation flux was calculated by measuring the increase in weight of the stripping solution or deionized water with a digital balance (Ohaus AV8101, Germany).

2.2. Factorial design methodology

A total of 13 experiments were performed according to a full factorial design with three factors (8 points of the factorial design and 5 centre points to establish the experimental errors). The variable factors with the coded and actual values are presented in Table 2. The experiments were carried out in randomized run order to determine two characteristic responses: evaporation flux (J) and soluble solid content (SSC). The results were taken after 3 h of concentration process under steady state conditions. The flux was relatively stable throughout the run even when achieving 44°Brix of the feed.

Table 3 shows the experimental matrix design and the results of the response variables studied. The experimental design and analysis of data were done using a commercial statistical package, Design-Expert version 7.1 (State-Ease, Inc., Minneapolis, MN).

The true response surface can often be approximated over a small experimental region by a low-order polynomial. A first-order polynomial model is only able to estimate the main effects of the experimental factors and does not account for either interactions or curvilinear effects. If there is little curvature in the limited region, a first-order model with interaction is appropriate for modeling. Adding interaction terms introduces curvature into the response function [8,9]. The first-order model with interaction terms proposed for each response variable (Y_i) was based on the multiple linear regression method. The empirical model in terms of coded factors was:

$$Y = \beta_0 + \beta_1 x_1 + \beta_2 x_2 + \beta_3 x_3 + \beta_{12} x_1 x_2 + \beta_{13} x_1 x_3 + \beta_{23} x_2 x_3 \qquad (1)$$

where β_i are the values of the regression coefficients. β_0 being the constant term, β_1, β_2 and β_3 the linear effects, β_{12}, β_{13} and β_{23} the interaction effects while the x_1, x_2, x_3 are the independent coded variables (temperature difference, $CaCl_2$ concentration and flow rate of both feed and distillate streams, respectively).

Stepwise deletion of terms was applied to eliminate the statistically non-significant terms. The goodness of fit of the model and significance of each regression coefficient was evaluated by regression analysis and ANOVA. 3D surface plots were generated using Design-Expert software.

3. Results and discussion

The independent and dependent variables were fitted to the first-order model equation with interaction terms (Eq. (1)) and for

Table 1
Data sheet of Microdyn—MD 020 CP 2N filter module.

Configuration	Tubes and shell
Material	
Housing	Polypropylene
Membrane	Polypropylene
Potting	Polyurethane
Membrane and module data	
Number of capillaries	40
Inner diameter	1.8 mm
Pore size	0.2 μm
Membrane area inside	0.1 m^2
Free flow area	1.0 cm^2
Housing dimensions ($D \times L$)	25 mm × 500 mm
Feed flow rate at axial velocity of 1 m/s	360 L/h
Membrane operational data	
Max. transmembrane pressure inside to outside	1.6 bar
Processing temperature	5–40 °C

Table 2
Factors, their coded levels and actual values as used in the design.

Variable	Symbol	Real values of coded levels		
		−1	0	+1
Temperature difference, ΔT (°C)	x_1	10	20	30
$CaCl_2$ concentration, C (% (w/w))	x_2	0	32.5	65
Flow rate, Q (L/h)	x_3	10	20	30

Table 3
Experimental matrix design and results obtained for each of the response variables studied.

Run number	Input variables			Responses	
	ΔT (°C)	C (%)	Q (L/h)	J (kg/m^2 h)	SSC (%)
1	20	32.5	20	0.669	17.76
2	20	32.5	20	0.619	17.37
3	10	65.0	30	1.11	27.00
4	10	65.0	10	0.764	18.00
5	30	65.0	10	1.164	28.69
6	30	65.0	30	1.462	44.11
7	30	0.0	10	0.723	17.44
8	10	0.0	10	0.064	12.80
9	30	0.0	30	1.074	24.97
10	20	32.5	20	0.671	18.06
11	20	32.5	20	0.585	16.85
12	10	0.0	30	0.266	13.84
13	20	32.5	20	0.642	16.96