

MULTIFACTORIAL MODELING OF THE FLUX DURING ULTRAFILTRATION OF STRAWBERRY EXTRACT

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Abstract

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Combined effects of working pressure, volume reduction ratio and feed flow rate on the flux during ultrafiltration of strawberry extract were studied depending on the molecular weight cut-off (1, 10 and 25 kDa) of polyacrylonitrile membranes. The mathematical models and response surfaces obtained demonstrate the highest flux values at high levels of working pressure and feed flow rate and low level of volume reduction ratio.

Key words: ultrafiltration, flux, multifactorial model, strawberry extract

Abbreviations: UF – ultrafiltration ; PAN – polyacrylonitrile ; VRR – volume reduction ratio ; DMSO – dimethyl sulfoxide; DMF – dimethyl formamide; MWCO – molecular weight cut-off

Introduction

Over the last two decades, the worldwide market for membrane technology in the food industry increased with about € 800–850 million and is now the second biggest industrial market for membranes after water and wastewater treatment including desalination (Peinemann et al., 2010). Baromembrane processes – microfiltration, ultrafiltration, nanofiltration and reverse osmosis are the most widely used processes in food technology (Marella et al., 2013). The major applications are in the dairy industry (milk, whey, brine, etc.) followed by other beverage industries (beer, fruit juices, wine, etc.). The success of membrane technology in the food and beverage market is directly related with some of the key advantages of membrane processes over conventional separation technologies (Le et al., 2014). Among these, advantages are:

- gentle product treatment due to moderate temperature changes during processing;
- high selectivity based on unique separation mechanisms, for example sieving;

- compact and modular design for ease of installation and extension;
- low energy consumption compared to condensers and evaporators;
- environmental friendliness;
- increasing the yield and quality of the final product;
- reduction of the production costs.

However, the major disadvantage of these processes is membrane fouling during permeation caused by the retention of some components over the surface of the membrane, leading to a rapid decrease of flux (Cassano et al., 2003; Espamer et al., 2006). The successful application of membrane processes is not possible without knowledge not only of the basic characteristics (flux and selectivity), but the dependence of these characteristics of some operational factors. Some of them such as working pressure, temperature, pH can be adjusted within a certain interval, while the composition and concentration of solutes found difficult to regulate. Each of the factors influences on the efficiency of the membranes.

The working pressure is crucial for realization of baromem-

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brane processes. The increase in working pressure leads to an increase in flux, gradually this increasing slows, then remains constant, regardless of the further increase in pressure. This can be explained by the deposition of gel layer on the membrane surface, such as the increase in the driving force leads to an increase in the thickness of the gel layer and its compacting, and consequently the selectivity of the membrane increases also (Rinaldoni et al., 2009; Espina et al., 2010; Baldasso et al., 2011). Roy and De (2015) established that the increase in pressure from 138 to 551 kPa leads to an increase in flux from 20 to 290 $\text{l.m}^{-2}.\text{h}^{-1}$) during ultrafiltration of Stevia extract using novel CAP-PAN blend membranes. Loginov et al. (2011) investigated the effect of working pressure from 0.05 MPa to 0.37 MPa on the flux during ultrafiltration of sugar beet juices and established an increase with the increase in pressure. The feed flow rate has also positive effect on the flux because the velocity increases which decreases the influence of concentration polarization (Cassano et al., 2007).

The increase in the concentration of solutes in the input stream increases the viscosity, which leads to a reduction of the conduction coefficient and a decrease in flux. Zhang et al. (2015) established that the flux decreases most highly when the volume reduction ratio increased from 1 to 2 than from 2 to 6 during ultrafiltration concentration of alfalfa juice.

The aim of this study was to investigate the combined effects of working pressure, volume reduction ratio and feed flow rate on the flux during ultrafiltration of strawberry extract with UF1-PAN, UF10-PAN and UF25-PAN polyacrylonitrile membranes through multifactorial mathematical model.

Materials and Methods

Strawberry extract

The extract of polyphenols was obtained from strawberry variety Siabella (harvest year 2015). The strawberries were ho-

mogenized after defrosting (2 h, 20°C). The mash (2 kg) was extracted with 2.5 l.kg^{-1} of methanol acidified with hydrochloric acid (1% v/v) during one night at 4°C. After filtration of the extracting blend, the organic solvent was evaporated by rotational vacuum-evaporator (30°C). For purification of the crude extract, a column (465x30 mm internal diameter) with adsorption resin Amberlite® XAD 16 N (Sigma-Aldrich, Steinheim, Germany) was used. Before feeding of the sample, the resin was activated with 500 ml methanol and flushed with 1000 ml distilled water, acidified with trifluoroacetic acid (TFA, pH 2.0). After addition of 250 ml polyphenol extract to the adsorption resin, it was flushed with 1000 ml acidified water (TFA, pH 2.0) and polyphenols were eluted with 500 ml mixture from methanol and acidified water (TFA, pH 2.0) (95:5 v/v). The organic solvent was evaporated from the eluent under vacuum (30°C). The model solution of strawberry extract was prepared with distilled water, acidified (pH 3.0) with 1 M HCl.

Membranes

Polyacrylonitrile (PAN) membranes UF1-PAN, UF10-PAN and UF25-PAN with a molecular weight cut-off of 1, 10 and 25 kDa, respectively, were used. Membranes were obtained by dry-wet phase-inversion method from polymer solutions prepared with the following solvents: UF1-PAN – dimethyl sulfoxide (DMSO) and dimethyl formamide (DMF) at a ratio 1:1; UF10-PAN – DMSO; UF25-PAN – DMF. The membranes were temperature-treated in an aqueous medium for 10 min, as follows: UF1-PAN and UF10-PAN – at 60°C; UF25-PAN – at 80°C.

Experimental system

The membrane filtration experiments were carried out on laboratory equipment with a replaceable plate and frame membrane module shown in Figure 1.

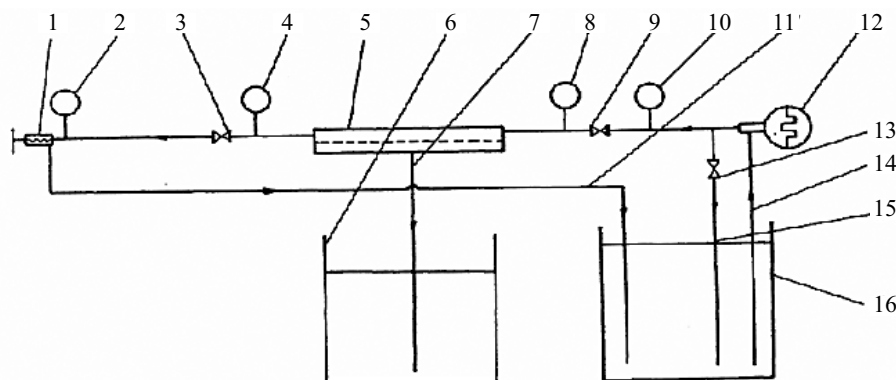


Fig. 1. Scheme of laboratory equipment with a replaceable plate and frame membrane module

1 – valve; 2 – manometer (0–5 MPa); 3 – valve; 4 – manometer (0–0.6 MPa); 5 – replaceable plate and frame membrane module; 6 – tank; 7 – pipeline; 8 – manometer (0–0.8 MPa); 9 – valve; 10 – manometer (0–15 MPa); 11 – pipeline; 12 – pump; 13 – valve; 14 – pipeline; 15 – pipeline; 16 – tank

The experiments were carried out under the following operating conditions: working pressure – from 0.2 to 0.4 MPa, volume reduction ratio (VRR) – from 2 to 6, feed flow rate – from 190 to 330 dm³.h⁻¹. All experiments were carried out at a temperature of 20°C. After experimental measurements of permeate volume (V, cm³) separated from the respective working elements for the time defined (τ , s) under different working conditions, the flux (J , dm³.m⁻².h⁻¹) was calculated using the following formula:

$$J = 3.6V/F \cdot \tau \quad (1),$$

where $F = 0.125$ m² is the membrane surface area in the module.

Statistical analysis

Full multifactorial mathematical model ($N = 2^3$) is used to show the influence of working pressure (p , MPa), volume reduction ratio (VRR) and feed flow rate (Q , dm³.h⁻¹) on the flux during ultrafiltration of strawberry extract. The experimental design with natural and coded values of the factors is presented in Table 1. The experimental data at each point of the design were obtained on the basis of threefold repetition.

Table 1
The experimental design with natural and coded values

№	Natural values			Coded values		
	p, MPa	VRR	Q, dm ³ .h ⁻¹	X ₁	X ₂	X ₃
1	0.2	2	190	-1	-1	-1
2	0.4	2	190	1	-1	-1
3	0.2	2	330	-1	-1	1
4	0.4	2	330	1	-1	1
5	0.2	6	190	-1	1	-1
6	0.4	6	190	1	1	-1
7	0.2	6	330	-1	1	1
8	0.4	6	330	1	1	1

Regression model for the dependent parameter (flux) were obtained using StatGraph XIV trial version statistical software.

Table 3. Regression models for flux depending on the working pressure (X₁), volume reduction ratio (X₂) and feed flow rate (X₃) for the three membranes

Membrane	Regression models
UF1-PAN	$J = 13.3167 + 4.04167X_1 - 1.46667X_2 + 0.408333X_3$ R=0.98; F=0.10 < F _T = 3
UF10-PAN	$J = 19.1133 + 4.91333X_1 - 1.9X_2 + 1.51X_3 + 0.42X_1X_3 - 0.5726667X_2X_3 - 0.41X_1X_2X_3$ R=0.99; F=1.02 < F _T = 4.5
UF25-PAN	$J = 20.78 + 5.43833X_1 - 0.851667X_2 + 0.588333X_3$ R=0.98; F=0.68 < F _T = 3

Results and Discussion

Table 2 shows the averages and standard deviations of the flux depending on the three investigated factors. The lowest results are obtained at $p = 0.2$ MPa, VRR = 6 and $Q = 190$ dm³.h⁻¹, while the highest – at $p = 0.4$ MPa, VRR = 2 and $Q = 330$ dm³.h⁻¹ for all tested membranes. Comparing the flux with the three investigated membranes it can be seen that the highest flux is obtained with UF25-PAN membrane, the lowest – with UF1-PAN membrane. Similar results were obtained from Luo et al. (2013) who established an increase in flux with the increase in molecular weight cut-off.

Table 2
Change of flux (J , dm³.m⁻².h⁻¹) depending on the working pressure, volume reduction ratio and feed flow rate during ultrafiltration of strawberry extract with UF1-PAN, UF10-PAN and UF25-PAN membranes

№	Flux J , dm ³ .m ⁻² .h ⁻¹ UF1-PAN	Flux J , dm ³ .m ⁻² .h ⁻¹ UF10-PAN	Flux J , dm ³ .m ⁻² .h ⁻¹ UF25-PAN
1	10.3±0.8	14.7±0.7	15.1±0.8
2	18.5±0.8	23.2±1.1	26.5±0.5
3	11.1±0.9	17.1±0.7	16.8±0.5
4	19.3±1.1	28.9±0.5	28.2±1.2
5	7.6±1.1	11.5±0.6	14.4±1.9
6	15.3±1.1	21±0.7	24.8±0.7
7	8.1±1	13.4±0.6	15.1±0.4
8	16.4±0.6	22.9±0.6	25.5±1.4

The following adequate models at confidence interval 95 % with significant coefficients were obtained (Table 3).

The standardized diagrams of Pareto for the significance of the investigated factors on the flux with UF1-PAN, UF10-PAN and UF25-PAN membranes are presented in Figures 2, 3 and 4. They show that all factors are significant. The biggest effect on the flux has the working pressure followed by the volume reduction ratio and feed flow rate. The diagrams of Pareto and the models show a positive effect for working pressure and feed flow rate and negative for volume reduction ratio for all samples.

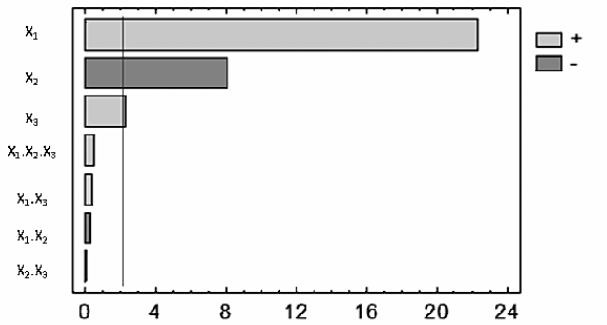


Fig. 2. Standardized diagram of Pareto for UF1-PAN membrane

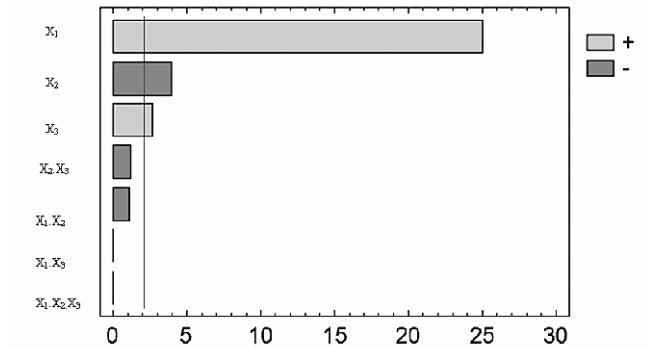


Fig. 4. Standardized diagram for UF25-PAN membrane

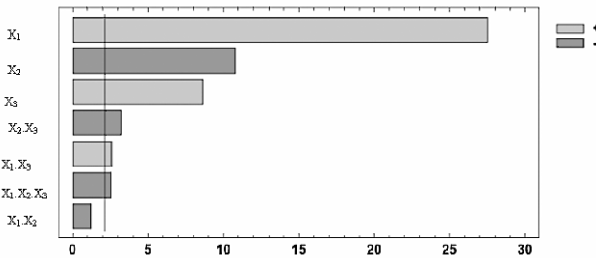


Fig. 3. Standardized diagram of Pareto for UF10-PAN membrane

The surface responses of the flux depending on the working pressure (X_1) and volume reduction ratio (X_2) are presented in Figure 5. It shows that the lowest value of the flux is obtained at high level of volume reduction ratio and at low level of working pressure with all investigated membranes. Increasing the working pressure leads to an increase in flux at two investigated levels of volume reduction ratio. The highest value of the flux is obtained at high level of the working pressure and low level of the volume reduction ratio. Similar results were obtained from Roy and De (2015). They established that the increase in pressure leads to an increase in flux during ultrafiltration of Stevia extract using novel CAP-

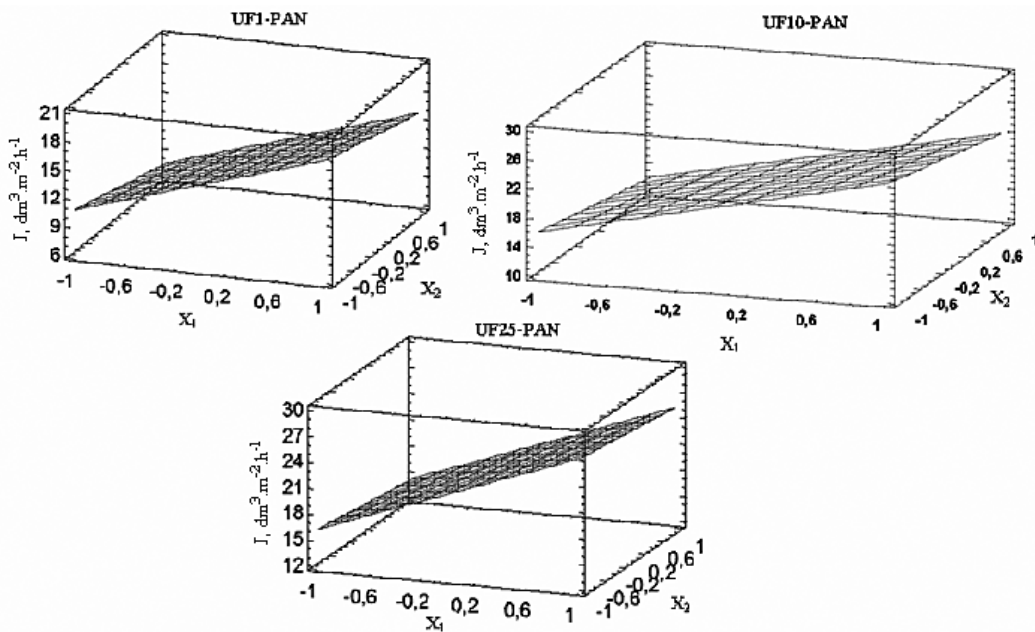


Fig. 5. Surface responses of the flux depending on the working pressure (X_1) and volume reduction ratio (X_2) for the three investigated membranes

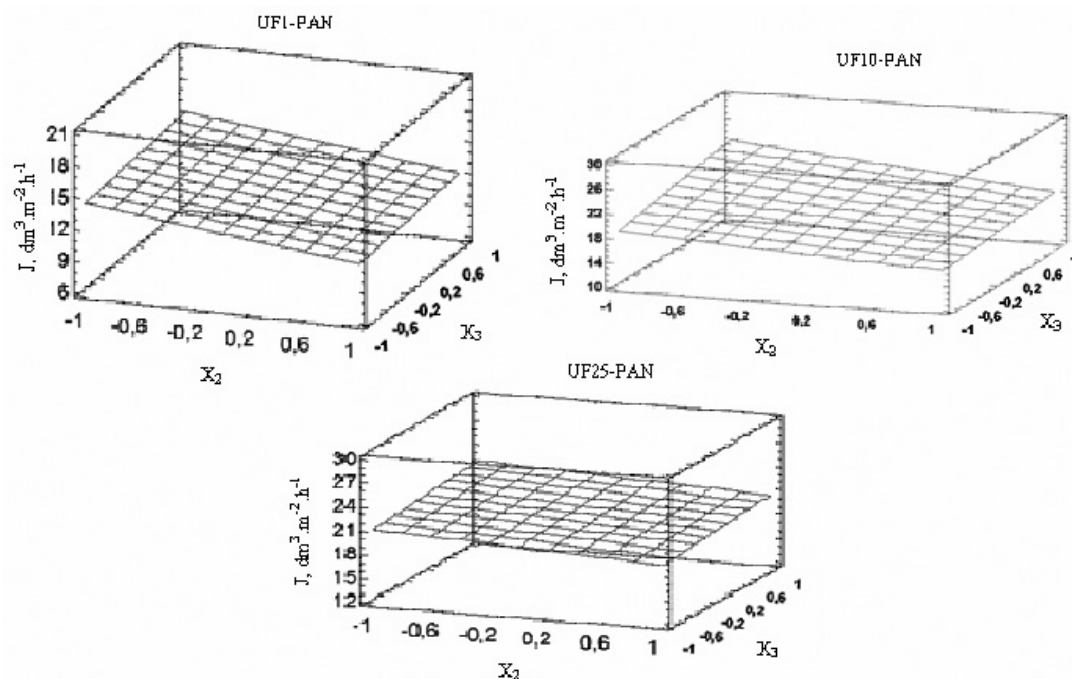


Fig. 6. Surface responses of the flux depending on the volume reduction ratio (X_2) and feed flow rate (X_3)

PAN blend membranes. Zhang et al. (2015) investigated the effect of volume reduction ratio on the flux during ultrafiltration of alfalfa juice and established that the more pronounced decreasing of flux is in the beginning of the process.

The surface responses of the flux depending on the volume reduction ratio (X_2) and the feed flow rate (X_3) are shown in Figure 6. It can be seen that the highest value of the flux is obtained at high level of feed flow rate and low level of volume reduction ratio. The lowest flux is obtained at low level of feed flow rate and high level of volume reduction ratio. The increase in feed flow rate leads to an increase in the velocity of solution. This contributes to the increase in Reynolds number which causes the decrease of the effect of concentration polarization and an increase in flux (Tasselli et al., 2007).

Conclusion

Multi-factorial mathematical models were created describing the effects of working pressure, volume reduction ratio and feed flow rate on the flux during ultrafiltration of strawberry extract using polyacrylonitrile membranes (1, 10 and 25 kDa MWCO). The models and response surfaces obtained demonstrate the highest flux values at high levels of

working pressure and feed flow rate and low level of volume reduction ratio.

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