

MULTI-FACTORIAL MATHEMATICAL MODEL FOR THE FLUX DURING ULTRAFILTRATION OF COW'S MILK

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Abstract

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Combined effects of working pressure, temperature and fat content on the flux during ultrafiltration of cow's milk with a polyacrylonitrile membrane UF10-PAN through full factorial experimental design were studied. Multi-factorial mathematical model for the influence of working pressure, temperature and fat content of milk on the flux was created. The model and response surfaces show that the highest value of flux is obtained at high level of the working pressure and temperature, and low level of the fat content of milk.

Key words: ultrafiltration, flux, cow milk, pressure, temperature, fat content

List of abbreviations: UF10-PAN – ultrafiltration polyacrylonitrile membrane with 10 kDa molecular weight cut-off; DMSO – dimethyl sulfoxide; VRR – volume reduction ratio

Introduction

During the last few years membrane processes are used for purification, separation and concentration of various food and pharmaceutical products (Wickramasinghe 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 advantages of membrane processes in comparison with the traditional separation methods are: environmental friendliness, lower power consumption, increased yield and quality of the final product, reduction of the production costs, realization of the process at room temperature in order to treat heat-sensitive products and keep their natural properties (Le et al., 2014).

In comparison with traditional filtration, during the membrane filtration the solution is fed in parallel (tangential) on the membrane. The tangential speed captivates the components on the membrane, thereby avoids the formation of large sedi-

ment layer and thus the concentration polarization decreases. The substances retained on the membrane form a concentrated solution (concentrate or retentate), and the substances which pass through, form the filtrate or permeate. Both obtained product can be used in food industry (Saxena et al., 2009).

Ultrafiltration (UF) is a pressure-driven process using a semi-permeable membrane to separate macromolecules or colloids from liquids and it is based on a simple sieving mechanism (De Bruijn et al., 2005; Van Reis et al., 2007).

The successful application of membrane processes is not possible without knowledge not only of the basic characteristics, 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 are difficult to regulate. Each of the factors influences on the efficiency of the membranes.

The working pressure is crucial for realization of baromembrane processes. The increase in working pressure leads

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to an increase in the 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., 2011; Baldasso et al., 2011).

The increase in temperature leads to an increase in the flux and selectivity of the membranes. This is explained by the fact that the increase in temperature leads to lower dynamic viscosity and reduce the negative impact of the concentration polarization, while the productivity increases also (Rinaldoni et al., 2009; Konrad et al., 2012).

The increase in the concentration of solutes in the input stream increases the viscosity, which leads to a reduction of the mass transfer coefficient. For these reasons, the performance of the membranes (flux and selectivity) gets worse (Yorgun et al., 2008).

Shahabadi and Reyhani (2014) studied the combined effects of pressure, temperature and velocity during ultrafiltration of water with a polyacrylonitrile membrane. They found that all factors influence positively on the flux but the more pronounced is pressure followed by temperature and velocity. Nordin and Jönsson (2010) studied the combined effects of velocity and working pressure on the flux during ultrafiltration of wastewater with polymer (6 kDa) and ceramic (5 kDa) mem-

branes. According to them, both investigated factors influence positively on the flux when using the polymer membrane, whereas the increase in velocity leads to an increase in flux of ceramic membrane and the increase of pressure does not have substantial influence, and it remains constant. This signifies that the membrane structure and material have significant effect on the process. Ramachandra Rao (2002) investigates the changes of the flux during ultrafiltration of various dairy products (normalized and skimmed cow's milk, buttermilk, sweet and sour whey). A higher flux is obtained when using the two types of whey in comparison with the other products. The high concentration of protein in buttermilk, skim, and normalized cow milk results in an increase in the concentration polarization and a decrease in the flux.

The aim of this study was to investigate the combined effects of working pressure, temperature and fat content on the flux during ultrafiltration of cow's milk with UF10-PAN polyacrylonitrile membrane through full factorial experimental design.

Materials and Methods

Materials

The experimental investigations were carried out with skimmed cow milk with a fat content of 1 g.kg⁻¹, and normalized cow milk with a fat content of 36 g.kg⁻¹ (provided by company BCC Hendel, Bulgaria).

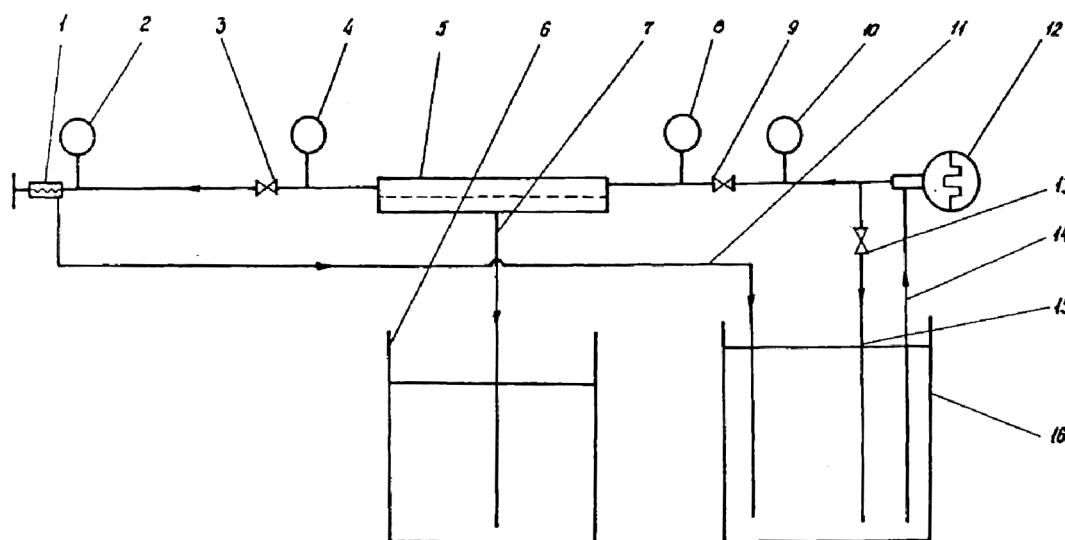


Fig. 1. Scheme of laboratory equipment with a replaceable plate and frame membrane module

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

Membranes

Polyacrylonitrile membrane UF10-PAN with 10 kDa molecular weight cut-off was used in all experiments. Ultrafiltration membranes were prepared by dry-wet phase-inversion method of polymer solutions prepared with a solvent of dimethyl sulfoxide (DMSO). They are temperature-treated in an aqueous medium for 10 min at 60°C.

Experimental system

The membrane filtration experiments were carried out on laboratory equipment with a replaceable plate and frame membrane module fitted with a UF10-PAN polyacrylonitrilic ultrafiltration membrane with 10 kDa molecular weight cut-off shown in Figure 1. This equipment was fitted with a plate and frame membrane module with membrane surface area of 1 250 cm²; a threeplunger high-pressure pump (max 15 MPa) with a capacity of 330 dm³.h⁻¹; a pipeline system with two manometers (0 MPa to 15 MPa) for measuring the inlet and outlet pressure; and a special working pressure regulating valve.

The experiments were carried out under the following operating conditions: transmembrane pressure 0.5 MPa, volume reduction ratio (VRR) – 2 and 3. All experiments were carried out at a temperature of 50°C and input flow of 330 dm³.h⁻¹. After experimental measurements of permeate volume (V, cm³) for the time defined (τ, s) under different working conditions, the flux (J, dm³.m⁻².h⁻¹) was calculated using the following formula:

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

where $F = 0.125 \text{ m}^2$ is the membrane surface area in the module

Statistical analysis

Full multi-factorial experimental design ($N = 2^3$) is used to show the influence of pressure (p, MPa), temperature (T, °C) and fat content (M, g.kg⁻¹) on the flux during ultrafiltra-

tion of skim and normalized cow milk. The experimental design with natural and coded values of the factors is presented in Table 1. Experiments at each point of the design were carried out with three replications.

Regression model for the dependent parameters (pressure, temperature and fat content) was obtained using Stat-Graph v2.0 statistical software.

Results and Discussion

Table 2 shows the averages and standard deviation of the flux depending on the three tested factors. The results show that it varies between 10.67 dm³.m⁻².h⁻¹ at p = 0.2 MPa, T = 20°C and M = 36 g.kg⁻¹ and 16.93 dm³.m⁻².h⁻¹ at p = 0.5 MPa, T = 50°C and M = 1 g.kg⁻¹.

Table 2

Experimental results for the flux depending on the investigated factors

| № | Flux, dm ³ /(m ² .h) |
|---|--|
| 1 | 11.47±0.53 |
| 2 | 13.42±0.12 |
| 3 | 15.00±0.22 |
| 4 | 16.93±0.67 |
| 5 | 10.67±0.32 |
| 6 | 11.85±0.22 |
| 7 | 12.60±0.12 |
| 8 | 13.44±0.43 |

The follow adequate model at confidence interval 95% with significant coefficients was obtained:

$$J = 13.3171 + 0.704583 * X_1 + 1.28708 * X_2 - 0.999583 * X_3 + 0.377083 * X_1.X_3 - 0.584583 * X_2.X_3 \quad (2)$$

R = 0.96; F = 0.57

The standardized diagram of Pareto for the significance of the investigated factors is presented in Figure 2. It shows

Table 1
The experimental design with natural and coded values

| № | Natural values | | | Coded values | | |
|---|----------------|-------|------|----------------|----------------|----------------|
| | P, MPa | T, °C | M, % | X ₁ | X ₂ | X ₃ |
| 1 | 0.2 | 20 | 0.1 | -1.0 | -1.0 | -1.0 |
| 2 | 0.5 | 20 | 0.1 | 1.0 | -1.0 | -1.0 |
| 3 | 0.2 | 50 | 0.1 | -1.0 | 1.0 | -1.0 |
| 4 | 0.5 | 50 | 0.1 | 1.0 | 1.0 | -1.0 |
| 5 | 0.2 | 20 | 3.6 | -1.0 | -1.0 | 1.0 |
| 6 | 0.5 | 20 | 3.6 | 1.0 | -1.0 | 1.0 |
| 7 | 0.2 | 50 | 3.6 | -1.0 | 1.0 | 1.0 |
| 8 | 0.5 | 50 | 3.6 | 1.0 | 1.0 | 1.0 |

that the three factors, as well as the factor interactions between them are significant, except the interaction $X_1 \cdot X_2$. The regression model obtained and standardized diagram of Pareto show that the factors pressure and temperature have a positive effect, the fat content – negative. The biggest effect has the temperature, followed by fat and pressure. This is confirmed by the obtained equation coefficients, respectively – 1.28708 for temperature, 0.999583 for fat content and 0.704583 for pressure and by Figure 3 showing a single effect of each of the investigated factors on the flux.

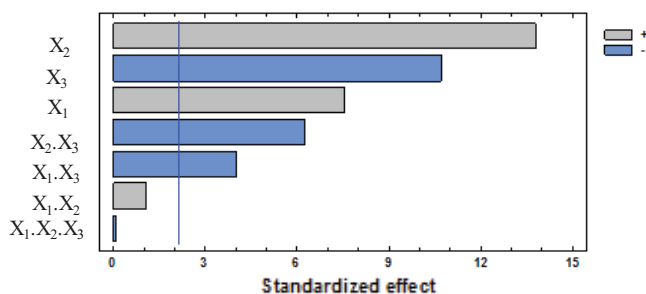


Fig. 2. Diagram of Pareto

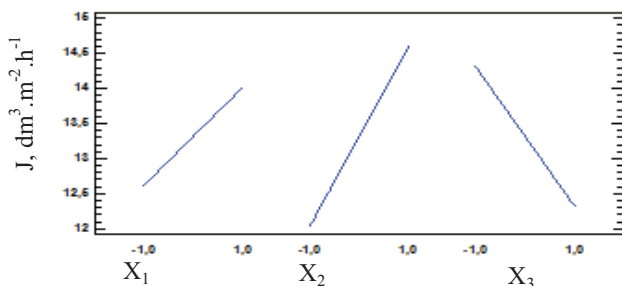


Fig. 3. Single influence of the studied factors on flux

The response surface of the flux depending on the working pressure (X_1) and temperature (X_2) is presented in Figure 4. It shows that the lowest value of the flux is obtained at low level of the two investigated factors, and the highest – at high level. Increasing the working pressure leads to an increase in the flux at two investigated levels of temperature. The same trend is observed at the factor temperature. Similar results were obtained from Shahabadi and Reyhani (2014) for a polyacrylonitrile membrane with a 20 kDa molecular weight cut-off. They found that the flux increases with the increase in temperature in the range of 30°C to 50°C and with the increase in pressure from 0.2 MPa to 0.5 MPa, but the more pronounced is the effect of pressure. This could be explained by the more narrow range of temperature.

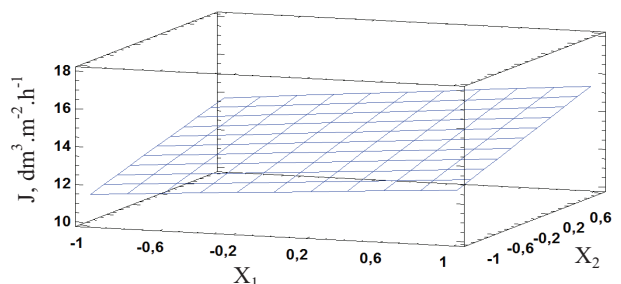


Fig. 4. Response surface of the flux depending on the working pressure (X_1) and the temperature (X_2)

The response surface of the flux depending on the working pressure (X_1) and fat content of milk (X_3) is presented in Figure 5. It shows that the highest value of the flux is obtained at high level of working pressure and low level of milk fat content. The lowest value of the flux is obtained at low level of working pressure and high level of milk fat content. The increase in milk fat content leads to an increase in the dry matter of the product. This causes an increase in dynamic viscosity of the solution which leads to a reduction of mass transfer coefficient. All this reasons get worse the performance of the membranes (flux and selectivity) (Shahabadi and Reyhani, 2014; Yorgun et al., 2008, Baldasso et al., 2011), which is confirmed by this experimental study.

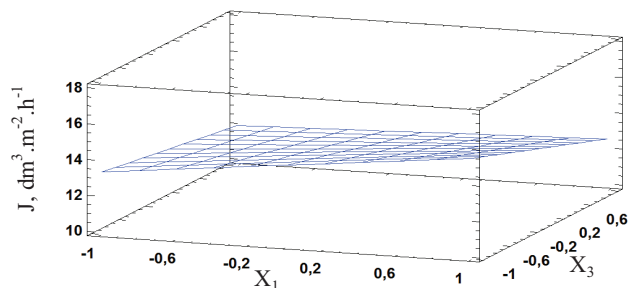


Fig. 5. Response surface of the flux depending on the working pressure (X_1) and fat content of milk (X_3)

In the process of increased pressure centrifugal pumps can act as destroyers of milk fat globules. This results in the formation of free fat, which reduces flux through the membrane and the retention of the fat decreases. This process takes place mostly at a high concentration. For this reason, for each product the ultrafiltration is stopped at a certain difference between inlet and outlet pressure over membranes.

The response surface of the flux depending on the temperature (X_2) and the fat content of milk (X_3) are shown in Figure 6. It shows that the highest value of flux is obtained at high level of temperature and low level of milk fat content. The highest flux is obtained at low level of temperature and high level of fat content. This trend is similar to the influence of pressure and fat content on the flux (Figure 5). The increase in temperature leads to a reduction of the dynamic viscosity of the solution, as the negative influence of the concentration polarization, while the flux increases simultaneously (Konrad et al., 2012; Karnataka 2003).

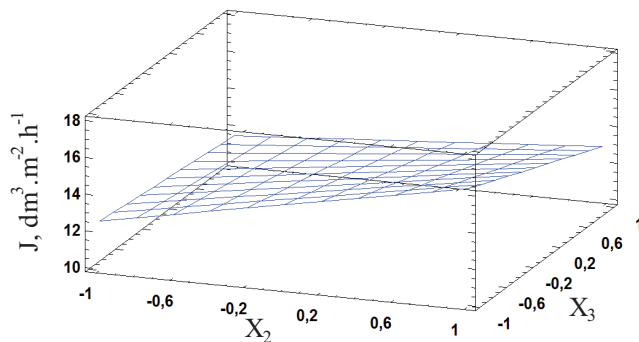


Fig. 6. Response surface of the flux depending on the temperature (X_2) and fat content of milk (X_3)

Conclusion

A multi-factorial mathematical model for the effect of the working pressure, temperature and milk fat content on the flux during ultrafiltration of cow milk with a polyacrylonitrile membrane with 10 kDa molecular weight cut-off is created. The model and response surface show that the highest value of the flux is obtained at high level of working pressure and temperature, and low level of milk fat content.

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