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Investigation of the flux, selectivity and concentration factor during ultrafiltration of goat's milk

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Abstract

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Combined effect of the working pressure, the feed flow rate and the volume reduction ratio on the flux during ultrafiltration of goat's milk with a polyacrylonitrile membrane UF10-PAN through full factorial experimental design was studied. The multi-factorial mathematical model and response surfaces showed that the highest value of the flux was obtained at a high level of the working pressure and the feed flow rate, and low level of the volume reduction ratio. The concentration factors according to the fat content, proteins and mineral substances increased, as the lowest increase was observed with the mineral substances. The selectivity of the membrane according to the proteins achieved 97.93% at volume reduction ratio of 3.

Keywords: ultrafiltration; goat's milk; flux; selectivity; concentration factor
Abbreviations: UF10-PAN – ultrafiltration polyacrylonitrile membrane with 10 kDa molecular weight cut-off; DMSO – dimethyl sulfoxide; VRR – volume reduction ratio; CF – concentration factor

Introduction

Nowadays membrane processes such as microfiltration, ultrafiltration, diafiltration, nanofiltration and reverse osmosis are successfully used for purification, fractionation and concentration of various food components and for this reason they can be defined as a key separation methods in the food industry (Le et al., 2014; Wickramasinghe et al., 2010).

The traditional separation methods (filtration, centrifugation and sedimentation) can be replaced by the membrane processes because they are carried out without phase transition and at a relatively low temperature which preserves the native characteristics of the separated components (Jahadi et al., 2018). Another advantages of the membrane processes are environmental friendliness (Kumar et al., 2013), lower power consumption (Baldasso et al., 2011), increased yield (Macedo et al., 2012; Ong et al., 2013) and quality of the final product (Domagala & Wszolek, 2008; Domagala et al., 2012), reduction of the production costs (Mehaia, 2005), high selectivity based on unique separation mechanism e.g. sieving, solution-diffusion or ion-exchange mechanism, compact and modular design (Mohammad et al., 2012).

Fractionation of particles from suspension through a semipermeable membrane can be achieved by ultrafiltration which is a variety of membrane filtration in which forces like pressure or concentration gradients lead to a separation. A solution flows under pressure over the surface of a suitable membrane and as a result of the applied pressure the solvent and certain dissolved components pass through the membrane. They are collected as permeate or ultrafiltrate while the other components of the solution which are retained by the membrane are well known as concentrate or retentate (Dhineshkumar & Ramasamy, 2017; Jahadi et al., 2018; Van Reis et al., 2007).

The flux and selectivity are the basic characteristics of the ultrafiltration process (Fang et al., 2015; Polyakov & Zydney, 2013). The flux also known as permeation rate is the quantity of permeated liquid (kg or m^3) per membrane area unit (m^2) and time unit (s) while the selectivity is quantitative characteristic for the ability of the membrane to retain components under specific working parameters.

There are many operating factors such as working pressure, temperature, volume reduction ratio, feed flow rate, concentration polarization, composition and concentration of solutes which influences on the flux and selectivity of the membrane during ultrafiltration. The pressure has significant effect on the flux because it is the driving force of the process. The increase in the working pressure led to to an increase in the flux of the membrane (Baldasso et al., 2011; Espina et al., 2010; Rinaldoni et al., 2009). A higher (flux) was obtained when the working temperature is higher during ultarafiltration (Konrad et al., 2012). The increase in the feed flow rate leads to an increase in the flux. Macedo et al. (2011) found that the increase in the velocity from 0.47 m/s to 1.23 m/s during ultrafiltration of whey from ovine cheese led to an increase in the flux of the membranes used. A lot of experimental investigations showed that the increase in the volume reduction ratio during ultrafiltration decreased the flux (Baldasso et al., 2011; Espina et al., 2011; Konrad et al., 2012). The increase in the volume reduction ratio influences on the selectivity. The knowledge of the single and combined effect of the factors during ultrafiltration is a prerequisite for the successful application of membrane processes.

The aim of the present experimental work was to investigate the flux, the selectivity and the concentration factor during ultrafiltration of goat's milk with UF10-PAN membrane.

Materials and Methods

Materials

The experimental investigations were carried out with a whole goat's milk, provided by company „Olympus", Bulgaria.

Membrane

Polyacrylonitrile UF10-PAN membrane with 10 kDa molecular weight cut-off was used in all experiments. Ultrafiltration membrane was prepared by dry-wet phase-inversion method of polymer solutions prepared with a solvent of dimethyl sulfoxide (DMSO). It was heat-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 polyacrylnitrile ultrafiltration membrane with 10 kDa molecular weight cut-off shown in Figure 1.

Fig. 1. Scheme of a laboratory equipment with a replaceable plate and frame membrane module: 1 – valve; 2, 3, 4 – manometers; 5 – replaceable plate and frame membrane module; 6 – pump; 7 - tank for initial solution; 8 – cylinder for permeate

Ultrafiltration experiments

The experiments were carried out under the following operating conditions: transmembrane pressure 0.2 MPa and 0.5 MPa, feed flow rate – 190 dm^3.h^{-1} and 330 dm^3.h^{-1}, volume reduction ratio (VRR) – 2 and 4. All experiments were carried out at a temperature of 20°C.

The volume reduction ratio (VRR) was calculated as follows:

$$VRR = V_O/V_R, \quad (1)$$

where: V_O is the volume of the feed solution, dm^3; V_R is the volume of the retentate obtained during ultrafiltration, dm^3.

After experimental measurements of permeate volume (V, cm^3) for the time defined (τ, s) under different working conditions, the flux (J, dm^3.m^{-2}.h^{-1}) was calculated using the following formula:

$$J = 3.6*V/(F*\tau) , \quad (2)$$

where F = 0.125 m^2 is the membrane surface area in the module.

The selectivity (retention factor R, %) was calculated using the following formula:

$$R = (1 – C_P/C_R)*100, \% \quad (3)$$

where: C_P is the concentration of the component presented in the permeate, %; C_R is the concentration of the component presented in the retentate, %.

For calculating the concentration factor (CF) was used the equation:

$$CF = C_R/C_O, \quad (4)$$

where: C_R is the concentration of the component presented in the retentate, %; C_O is the concentration of the component presented in the initial solution, %.

Determination of the main components of initial goat's milk, retentates and permeate

The experiments for retention and concentration factors were carried out under the following operating conditions: transmembrane pressure of 0.5 MPa, feed flow rate of 330 $dm^3.h^{-1}$, volume reduction ratio VRR = 2 and VRR = 3, temperature of 20°C. In all experiments, samples of the ultrafiltration retentates obtained at VRR = 2 and VRR = 3 and permeate were taken. The initial goat's milk, retentates and permeate obtained were analyzed according to the following indices: dry matter (ISO 6731:2010); fat (ISO 2446: 2008), total protein (EN ISO 8968-1: 2014); mineral substances (BSS 6154:1974).

Statistical analysis

The effect of pressure (p, MPa), feed flow rate (Q, $dm^3.h^{-1}$) and volume reduction ratio (VRR) on the flux during ultrafiltration of whole goat's milk was analyzed by full multi-factorial experimental design (N = 2^3). 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.

Table 1. Experimental design with natural and coded values

№	Natural values			Coded values		
	p, MPa	Q, $dm^3.h^{-1}$	VRR	X_1	X_2	X_3
1	0.2	190	2	-1.0	-1.0	-1.0
2	0.5	190	2	1.0	-1.0	-1.0
3	0.2	330	2	-1.0	1.0	-1.0
4	0.5	330	2	1.0	1.0	-1.0
5	0.2	190	4	-1.0	-1.0	1.0
6	0.5	190	4	1.0	-1.0	1.0
7	0.2	330	4	-1.0	1.0	1.0
8	0.5	330	4	1.0	1.0	1.0

Regression model for the dependent parameters (pressure, feed flow rate and volume reduction ratio) was obtained using StatGraph XIV trial version statistical software.

Fisher's least significant difference test at a significance level of 0.05 was used for the comparison of the experimental values using Excel 2010 by a one-way analysis of variance (one-way ANOVA).

Results and Discussion

Table 2 shows the averages and standard deviations of the flux depending on the three factors tested. The results show that it varies between 6.3 $dm^3.m^{-2}.h^{-1}$ at p = 0.2 MPa, Q 190 $dm^3.h^{-1}$ and VRR = 4 and 15 $dm^3.m^{-2}.h^{-1}$ at p = 0.5 MPa, Q = 330 $dm^3.h^{-1}$ and VRR = 2.

The following adequate model with significant coefficients at confidence level 0.95 was obtained:

$$J = 9.49708 + 0.794583* X_1 + 0.740417* X_2 - 2.58792*X_3 + 0.592917*X_1*X_2 - 0.290417*X_1*X_3 - 0.274583* X_2*X_3 \quad (5)$$

$R = 0.99 \qquad F = 0.88 < F_T = 4.5$

The standardized diagram of Pareto for the significance of the investigated factors is presented in Figure 2. It shows that the three factors, as well as the factor interactions between them are significant. The regression model obtained and the standardized diagram of Pareto show that the factors pressure and feed flow rate have a positive effect, while the VRR – negative. The biggest effect has the VRR, followed by the pressure and feed flow rate. This is confirmed by the obtained equation coefficients – 2.5872 for VRR, 0.794583 for pressure and 0.740417 for feed flow rate, and by Figure 3 showing a single effect of each of the investigated factors on the flux.

The response surface of the flux depending on the working pressure (X_1) and the feed flow rate (X_2) is presented in Figure 4. It shows that the pressure has a positive effect on

Table 2. Experimental results for the flux depending on the investigated factors

	Natural values			Coded values			Flux J, $dm^3.m^{-2}.h^{-1}$			
	p, MPa	Q, $dm^3.h^{-1}$	VRR	X_1	X_2	X_3	J_1	J_2	J_3	J
1	0.2	190	2	-1.0	-1.0	-1.0	10.70	10.90	10.80	10.8±0.1
2	0.5	190	2	1.0	-1.0	-1.0	11.19	11.49	11.34	11.3±0.15
3	0.2	330	2	-1.0	1.0	-1.0	11.30	11.10	11.20	11.2±0.1
4	0.5	330	2	1.0	1.0	-1.0	14.80	15.20	15.00	15±0.2
5	0.2	190	4	-1.0	-1.0	1.0	6.50	6.18	6.25	6.3±0.17
6	0.5	190	4	1.0	-1.0	1.0	6.20	6.80	6.73	6.6±0.33
7	0.2	330	4	-1.0	1.0	1.0	6.20	6.80	6.50	6.5±0.30
8	0.5	330	4	1.0	1.0	1.0	8.15	8.30	8.30	8.3±0.09

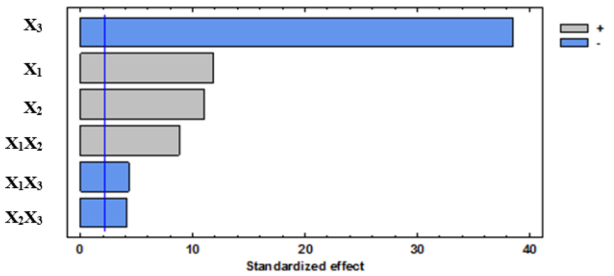

Fig. 2. Diagram of Pareto

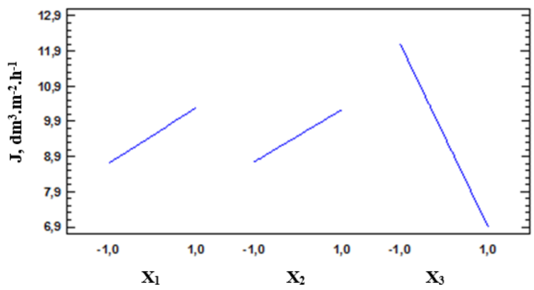

Fig. 3. Single effect of the factors on the flux

the flux, as more pronounced effect is observed at a high level of the feed flow rate than at a low level of the same factor. According to Rinaldoni et al. (2009), the increase in the pressure from 0.5 bar to 1.5 bar leads to an increase in the flux during ultrafiltration of skim milk at 30°C twice. The feed flow rate has also a positive effect on the flux like the working pressure. Increasing the flow rate leads to an increase in the Reynolds number (Re), which increases the turbulence of the flow and leads to a decrease in the concentration polarization and increase in the flux (Cassano & Basile, 2011).

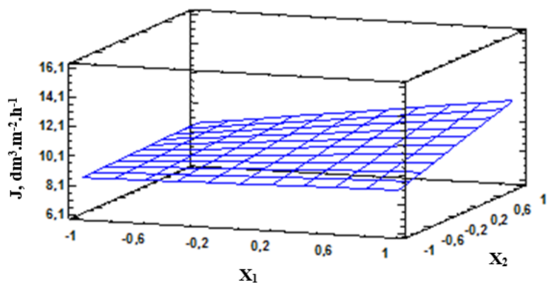

Fig. 4. Response surface of the flux depending on the working pressure (X_1) and the feed flow rate (X_2) at $X_3 = 0$

Figure 5 shows the response surface of the flux depending on the working pressure (X_1) and the volume reduction ratio (X_3). It can be seen that the highest value of the flux is obtained at a high level of the working pressure and a low level of the VRR. The effect of the concentration on the flux during ultrafiltration of whey with polyethersulfone membrane (10 kDa molecular weight cut-off), temperature of 50°C, pressure of 2 bar and inlet flow rate of 840 $dm^3.h^{-1}$ was investigated by Baldasso et al. (2011). The authors show that the increase in VRR from 1 to 7 resulted in a reduction in the flux of the membrane, like this reduction is greatest from the beginning of the process to VRR = 2. This is also confirmed by the experimental work of Bacchin et al. (2006). The authors established that the increase in the concentration of the solution leads to a reduction of the flux. 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) (Shabadi & Reyhani, 2014; Yorgun et al., 2008).

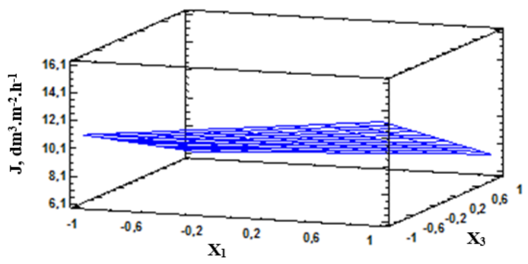

Fig. 5. Response surface of the flux depending on the working pressure (X_1) and the volume reduction ratio (X_3) at $X_2 = 0$

The response surface of the flux depending on the feed flow rate (X_2) and the VRR (X_3) is shown in Figure 6. The highest value of flux is obtained at a high level of the feed flow rate and a low level of the VRR. The lowest flux is obtained at a low level of the feed flow rate and a high level of the VRR. This trend is similar to the influence of the pressure and the VRR on the flux (Figure 5).

The principal components of initial goat's milk, retentates and permeate obtained by ultrafiltration at VRR =

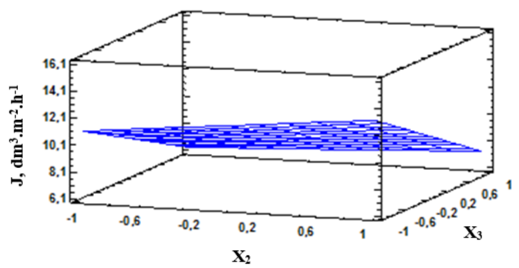

Fig. 6. Response surface of the flux depending on the feed flow rate (X_2) and the volume reduction ratio (X_3) at X_1=0

2 and VRR = 3, as well as the retention (selectivity) and concentration factors are presented in Table 3. It can be seen that the dry matter content changes from 12.30% to 23.86%, the total proteins – from 3.53% to 10.09%, the fat content – from 3.5% to 10.1%, the mineral substances – from 0.87% to 1.51%. Table 3 shows that more significant

Table 3. Main components of goat's milk, retentates and permeate, concentration (CF) and retention (R) factors at VRR = 2 and VRR = 3

Indices	Samples			Average values ± SD
	1	2	3	
Goat's milk				
Dry matter, %	12.56	12.3	12.04	12.30 ± 0.26[a]
Total protein, %	3.63	3.53	3.43	3.53 ± 0.10[a]
Total protein in % of dry matter	28.9	28.7	28.49	28.70 ± 0.21[a]
Milk fat, %	3.6	3.5	3.4	3.5 ± 0.10[a]
Milk fat in % of dry matter	28.66	28.46	28.24	28.45 ± 0.21[a]
Mineral substances, %	0.89	0.87	0.85	0.87 ± 0.02[a]
Mineral substances in % of dry matter	7.09	7.07	7.06	7.07 ± 0.02[a]
UF retentate at VRR = 2				
Dry matter, %	17.69	17.47	17.25	17.47 ± 0.22[b]
Concentration factor (CF) of dry matter	1.41	1.42	1.43	1.42 ± 0.01[a]
Total protein, %	6.82	6.74	6.66	6.74 ± 0.08[b]
Total protein in % of dry matter	38.55	38.58	38.61	38.58 ± 0.03[b]
Concentration factor (CF) of total protein	1.88	1.91	1.94	1.91 ± 0.03[a]
Retention factor (R) of total protein, %	95.75	96.88	98.05	96.89 ± 1.15[a]
Milk fat, %	6.42	6.3	6.18	6.30 ± 0.12[b]
Milk fat in % of dry matter	36.29	36.06	35.83	36.06 ± 0.23[b]
Concentration factor (CF) of milk fat	1.78	1.8	1.82	1.80 ± 0.02[a]
Mineral substances, %	1.19	1.18	1.17	1.18 ± 0.01[b]
Mineral substances in % of dry matter	6.73	6.75	6.78	6.75 ± 0.03[a]
Concentration factor (CF) of mineral substances	1.34	1.35	1.38	1.36 ± 0.02[a]
Retention factor (R) of mineral substances, %	53.78	54.24	54.7	54.24 ± 0.46[a]
UF retentate at VRR = 3				
Dry matter, %	24.33	23.86	23.39	23.86 ± 0.47[c]
Concentration factor (CF) of dry matter	1.94	1.94	1.94	1.94 ± 0.00[b]
Total protein, %	10.24	10.09	9.94	10.09 ± 0.15[c]
Total protein in % of dry matter	42.09	42.29	42.5	42.29 ± 0.21[c]
Concentration factor (CF) of total protein	2.82	2.86	2.9	2.86 ± 0.04[b]
Retention factor (R) of total protein, %	97.17	97.92	98.69	97.93 ± 0.76[a]
Milk fat, %	10.3	10.1	9.9	10.10 ± 0.20[c]
Milk fat in % of dry matter	42.33	42.33	42.33	42.33 ± 0.00[c]
Concentration factor (CF) of milk fat	2.86	2.89	2.91	2.89 ± 0.03[b]
Mineral substances, %	1.66	1.51	1.36	1.51 ±0.15[c]
Mineral substances in % of dry matter	6.82	6.33	5.81	6.32 ± 0.51[b]
Concentration factor (CF) of mineral substances	1.87	1.74	1.6	1.74 ± 0.14[b]
Retention factor (R) of mineral substances, %	66.87	64.24	61.03	64.05 ± 2.92[b]
Permeate				
Dry matter, %	5	4.92	4.84	4.92 ± 0.08[d]
Total protein, %	0.29	0.21	0.13	0.21 ± 0.08[d]
Total protein in % of dry matter	5.8	4.27	2.69	4.25 ± 0.21[d]
Mineral substances, %	0.55	0.54	0.53	0.54 ± 0.01[d]
Mineral substances in % of dry matter	11	10.98	10.95	10.98 ± 0.03[c]

change was in the protein and fat contents. This is due to the molecular weight cut-off of the membrane used in this research and its capacity to retain the high molecular-weight substances such as whey proteins and fat, and to permit the passage of the low-molecular substances such as minerals and lactose.

The results show that the concentration factor of the dry matter increase 1.42 times at VRR = 2 and 1.94 times at VRR = 3 ($p < 0.05$). Under the same operating conditions the protein concentration factors increase 1.91 and 2.86 times ($p < 0.05$) respectively, fat concentration factors – 1.80 and 2.89 times ($p < 0.05$), mineral substances – 1.36 and 1.74 times ($p < 0.05$). It can be seen that the concentration factors of high molecular-weight substances is more pronounced than the low molecular-weight ones. The data are in agreement with these reported in the literature (Baldasso et al., 2011; Macedo et al., 2012).

The results for the retention factor (R, %) show that it increases from 96.89% (VRR = 2) to 97.93% (VRR = 3) according to the proteins ($p < 0.05$), from 54.24% (VRR = 2) to 64.05% (VRR = 3) according to the mineral substances ($p < 0.05$). This results confirmed the suitability of the choice of the membrane with satisfactory selectivity.

Conclusion

A multi-factorial mathematical model for the effect of the working pressure, the feed flow rate and the volume reduction ratio on the flux during ultrafiltration of goat's milk with a polyacrylonitrile membrane with 10 kDa molecular weight cut-off was created. The model and response surfaces showed that the highest value of the flux was obtained at a high level of the working pressure and the feed flow rate, and a low level of the volume reduction ratio. The concentration factors according to the fat content, proteins and mineral substances increased, as the lowest increase was observed with the mineral substances. The selectivity of the membrane according to the proteins achieved 97.93% at VRR = 3.

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