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Modeling of Nanofiltration Process for Solvent Recovery from Aqueous Ethanol Solution of Soybean Isoflavones

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Abstract: In this paper, the feasibility of recovering the solvent from the aqueous ethanol solution of soybean isoflavones with nanofiltration (NF) was studied. Five commercially available polymeric NF membranes were employed and STARMEM^{TM} 122 showed acceptable flux and high retention. The central composite design (CCD) of the response surface methodology (RSM) was applied to model the effects of temperature, pressure, and feed concentration on the permeate flux and the total soybean isoflavone retention. The results indicate that the developed models were in good agreement with the experimental results and they can be used to predict this NF process.

Keywords: Central composite design, nanofiltration, response surface methodology, solvent recovery, soybean isoflavone

INTRODUCTION

An increasing number of studies have focused on soybeans and soy-based products in the last decade due to their reported nutritional and health benefits. Soybeans contain many valuable constituents, including isoflavones, proteins, saponins, and phytosterols. Among them, soybean

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isoflavones are reported to play an important role in protecting against several common diseases, including cancer, heart disease, and osteoporosis, as well as easing menopause symptoms for women (1–5).

Currently in industry, the purification processes of soybean isoflavones are commonly carried out by the chromatographic method, especially the macroporous resin method including resin adsorption, the desorption of soybean isoflavones, and the evaporation of the desorption solution. The aqueous ethanol solution with 80% (v/v) ethanol is widely used as the desorption solution and the total solids concentration is in the range of 1–10 g/L. The evaporation process, consuming a lot of steam and energy, is a high-cost process. Furthermore, environmental pollution and other serious problems may result. Consequently, an alterative solvent recovery process is needed.

Nanofiltration (NF), a relatively new pressure-driven membrane process positioned between reverse osmosis (RO) and ultrafiltration (UF), has prospered in many fields over the past few years (6). Generally, the NF membrane has a molecular weight cut-off (MWCO) between 200 and 1000 Da. Thus the feasibility of using NF for solvent recovery from the desorption solution of soybean isoflavones (MW: 250–550), can be highly expected. Nanofiltration has been widely and successfully used in the the food industry, the pharmaceutical industry, and water treatment because of its gentleness, cleanness, low-cost, and easy connection with the other process. A few years ago, most of the established nanofiltration processes treated aqueous systems. But with the development of the solvent-resistant nanofiltration (SRNF) membranes, organic solvent recovery with NF became possible and promising. Meanwhile several applications have already been developed using SRNF membranes (7–11). It has also been observed that the NF performance is much less predictable in organic solvents than in aqueous solution. Yang et al. (12) and Whu et al. (13) observed lower retention values in organic solvents than in aqueous solution. Yanyan Zhao et al. (14) studied the retention of several organic solutes in aqueous and organic solvents through several SRNF membranes and observed similar conclusions but with an uncommon retention performance for MPF-50.

Until now, little research has focused on the application of NF on soybean isoflavones. Among the solutes of Yanyan Zhao's study (14), soybean daidzin, one of the soybean isoflavones, was used and the retention of soybean daidzin in water is acceptable while the retention in methanol is low. In this study,

(a) the purpose is basic theoretical research, not application;
(b) the soybean isoflavone concentration is very low (10 mg/L) and the soybean isoflavone is highly pure;

(c) the solvent used in this study, methanol or water, is quite different from aqueous ethanol solution which is widely used as the desorption solution in the purification process of soybean isoflavones.

Thus, further studies need to be done to demonstrate the feasibility of NF application on the above-mentioned solute (crude soybean isoflavones)–solvent (aqueous ethanol solution) system with high solute concentration.

Furthermore, most of the studies on membrane performance were based on the "one-factor-at-a-time" (OFAT) method, which requires more runs and neglects the effect of factor interactions but also may miss optimal settings of factors (15). The DOE (design of experiment) methodology, a much more efficient method than OFAT, ensures all factors and their interactions systematically investigated. Thus, it has been widely applied in process optimizing or modeling in various fields. Central composite design (CCD), a response surface method (RSM), one of the most popular DOE methodologies, was selected for our study. From our knowledge, no studies have focused on the CCD application for describing the performance of NF membranes in the solvent recovery process.

The objective of this paper was to investigate the effect of process variables on the aqueous ethanol recovering NF process in current industrial practice of soybean isoflavones production, employing a central composite design method.

MATERIALS AND METHODS

Raw Materials

The mixed solvent, aqueous ethanol with 80% (v/v) ethanol, was prepared from ethanol of analytical grade (purchased from Beijing reagent company, China) and reverse osmosis water.

The solute, crude soybean isoflavone powder, was supplied by North China Pharmaceutical Group Corp. As provided by the supplier, the total soybean isoflavone content (i.e., purity) is 14.63%. Among them, the amount of the three major soybean isoflavones (daidzin, glycitin, and genistin) is close to 100% (53.21%, 10.88%, and 32.04%, respectively). Thus only these three soybean isoflavones are of concern in this study. Their MW values are 418.40, 448.43, and 434.40, respectively, and their chemical structures are shown in Fig. 1.

Known amounts of soybean isoflavone powder were dissolved in aqueous ethanol to prepare feed solutions containing various desired solute concentrations.

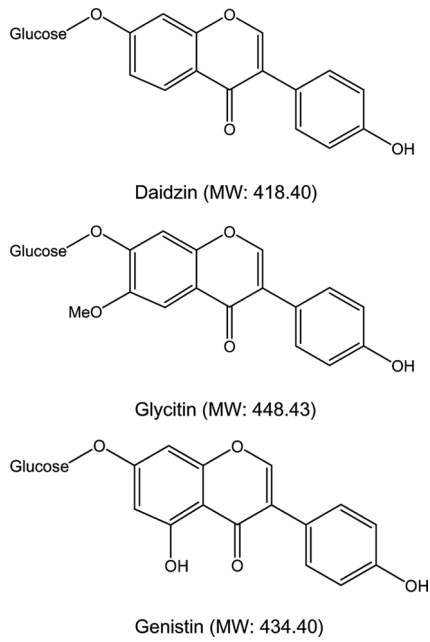

Figure 1. Chemical structures of the three major soybean isoflavones.

Apparatus

All nanofiltration experiments were performed using a C-40B (Nitto Denko, Osaka, Japan) Dead-end test cell, which comprises of a cylindrical stainless steel chamber with removable end plates. A membrane was placed on top of a porous stainless steel support disc (diameter of 75 mm). Buna-N (nitrile rubber) O-rings were used as sealing parts, giving an active membrane area of 32 cm^2. The cell, with a volume of 380 mL, was pressurized with compressed high-purity nitrogen gas and the pressure can be applied up to 4000 kPa. The temperature was controlled by using a water bath and heat exchanger, and stirring was provided by a Teflon-coated magnetic bar.

Membranes and Membrane Pretreatment

Five commercially available polymeric NF-membranes were employed in this study. MPF (MPF-44, MPF-50) membranes were supplied in a "wet" form in a preserving solution: MPF-44 soaked in 0.7% Roccal and MPF-50 in 50% ethanol/water. STARMEM^{TM} (STARMEM^{TM}

122, STARMEM™ 228, and STARMEM™ 240) membranes were supplied in a "dry" form. All the membranes were cut into circular discs (75 mm in diameter) from A4 flat sheets, washed in 80% ethanol/water, and pre-conditioned by saturating them into the mixed solvent for at least 5 days prior to the filtration experiments.

Experimental Procedure

Before each separation experiment, solvent (80% ethanol/water) permeation was carried out until the final stable flux (defined in this study as <2% change at least in half an hour) was reached. Generally, in this study, 1–4 days were needed for stable solvent fluxes. Afterwards, actual separation experiments were conducted. Then, the filtration cell was loaded with 100 mL feed solution for filtration, and all experiments were stopped at $VRF = 2$, which was obtained by (16):

$$VRF = \frac{V_I}{V_R} \tag{1}$$

where VRF is the volume reduction factor, V_I is the volume of initial feed, and V_R is the volume of the retentate. The volume of the permeate was measured with a measuring cylinder. Furthermore, a high stirring rate (300 rpm) was applied. The concentration of soybean isoflavone was measured using a HPLC; employing a phenomenex luna C18 reversed phase column (250 mm × 4.6 mm i.d., 5 μm, USA) on a Waters 2695 high performance liquid chromatography (Waters, USA) which is equipped with 2996 ultraviolet detection set at 254 nm. The column temperature was kept at 50°C and a binary mobile phase consisted of solvent A, 1.8% aqueous glacial acetic acid, and solvent B, methanol at a flow rate of 1.1 mL/min. The standard stock solutions of soybean isoflavone were prepared with HPLC grade methanol. The feed and retentate samples were diluted (if necessary) and filtered through a 0.45 μm filter before analysis. Quantitative data for daidzin, glycitin, and genistin were obtained by comparison with the standards.

The permeate flux (J) was obtained by

$$J = \frac{V}{At} \tag{2}$$

where V is the volume of permeate, A is the membrane area, and t is the time. The retention was calculated by

$$R = \left(1 - \frac{C_P}{C_R}\right) \times 100\% \tag{3}$$

where C_P and C_R are the final concentrations in the permeate and retentate, respectively.

For membrane selection, the experiments were performed in triplicate at 25°C and 1500 kPa, with the initial concentration of 10 g/L. According to the permeate flux and the total soybean isoflavone retention, we got our desired membrane, which was used for further experiments.

Experimental Design and Statistical Analysis

Experimental design and statistical analysis were performed using Design Expert software (version 7) from Stat-Ease Inc. (USA). A central composite design for three factors was used, in which the independent variables were converted to dimensionless ones, with the coded values at 5 levels: $-\alpha$, -1, 0, $+1$, $+\alpha$ (the value of α for this CCD was fixed at 1.5). The 5-level-3-factor CCD included a 2^3 design with 6 star points and 6 center points, leading to 20 runs. Replications at the center point were performed in order to estimate the residual error. The temperature, pressure, and the feed concentration were chosen for independent variables. The permeate flux and the total soybean isoflavone retention were selected as the responses to be examined. Experimental runs were randomized to minimize the effects of unexpected variability in the observed responses.

For statistical calculations, the variables X_i were coded as x_i according to the equation

$$x_i = \frac{X_i - X_0}{\Delta X} \tag{4}$$

where x_i is a coded value of variable, X_i is the actual value of variable, X_0 is the actual value of X_i at the center point, and ΔX is the step change value of the variable. Coded and uncoded levels of the three variables are shown in Table 1. The ranges and levels of variables were decided on the basis of the results obtained through our previous work.

Table 1. Coded and real levels of independent variables

		Levels				
Variables	Symbols	$-\alpha$	-1	0	1	α
Temperature (°C)	A	5	10	20	30	35
Pressure (kPa)	B	250	500	1000	1500	1750
Feed concentration (g/L)	C	1.0	2.5	5.5	8.5	10.0

The test for significance of the regression models, the test for significance of the individual model coefficients, and the test for lack-of-fit were performed applying the analysis of variances (ANOVA) to ensure a good model. The lower the *p*-value, the more significant is the corresponding coefficient. When the *p*-value of one term is higher than 0.100, it indicates that it is insignificant at 90% confidence level. In this case, insignificant terms were reduced from the initial model using the ''backward elimination procedure.'' The lack-of-fit test, a measure of the failure of the model to represent data in the experimental domain at which points were not include in the regression, was used to determine whether the model was adequate to describe the observed data.

The nonlinear computer-generated quadratic model was explained by the following equation:

$$Y = \beta_0 + \sum_{i=1}^{k} \beta_i X_i + \sum_{i=1}^{k} \beta_{ii} X_i^2 + \sum_{i=1}^{k-1} \sum_{j=1}^{k} \beta_{ij} X_i X_j + \varepsilon \qquad (5)$$

where Y is the measured response associated with each factor level combination (dependent variables), X_i and X_j the factors (independent variables), β_0 a constant, β_i, β_{ii} and β_{ij} the coefficients for the linear, quadratic, and interaction effect, and ε is a random error. The model evaluated the effect of each independent variable on a response.

R-squared measures how well a regression model approximates real data points, and an *R*-squared of 1.0 (100%) indicates a perfect fit. R^2 always increases as variables are added. Unlike R^2, the adjusted R^2 increases only if the new term improves the model. Thus, the adjusted R^2 is more preferred by model builders. The predicted R-squared statistic is a measure of how much variability in new data that the model is expected to explain. We prefer that the adjusted and predicted *R*-squared values be close to each other. A great difference between them, more than 0.20, indicates a problem with either the data or the model. Further, ''Adeq Precision'' measures the signal-to-noise ratio and a ratio greater than 4 is desirable.

RESULTS AND DISCUSSION

Membrane Selection

For the purpose of membrane selection, the main focus was to find a suitable membrane producing acceptable flux and retention, therefore, a membrane with both a high permeate flux and good soybean isoflavone retention would be desirable. The most important membrane

characteristics and the experiment results for membrane selection are now presented in Table 2.

The fluxes for MPF membranes (MPF-44 and MPF-50) were much lower than the fluxes for STARMEM^{TM} membranes (STARMEM^{TM} 122, STARMEM^{TM} 228, and STARMEM^{TM} 240). This may be due to the different membrane materials. For MPF-44, as a hydrophilic membrane, the pure water flux is certainly high, but the flux of aqueous ethanol solution is usually much lower (17). Thus, in this study, MPF-44 gave the lowest solvent flux although it was the only hydrophilic membrane among the five membranes. Among the three STARMEM^{TM} membranes studied (with similar materials), STARMEM^{TM} 122 gave the highest flux although its MWCO is the lowest. This is because flux does not depend on the MWCO. As shown in the data sheet provided by the supplier, the toluene flux for STARMEM^{TM} 240 is much lower than that for STARMEM^{TM} 122, even though the MWCO of STARMEM^{TM} 240 is much higher. Some researchers also obtained similar results (14). In determining flux, the membrane–solvent interaction properties are more important (10).

STARMEM^{TM} 122 also gave the highest retention. Many studies (12–14,19) have shown that the retention for SRNF was not consistent with the manufacturer-specified MWCO. Table 2 showed similar results. The influences of retention are very complicated for SRNF. The membrane–solvent-solute interaction properties are very important in determining retention.

Being the most effective membrane among the five chosen membranes, taking into account all the above results, we adopted STARMEM^{TM} 122 for further studies in order to evaluate the recovery of 80% (v/v) ethanol from the feed solution.

Central Composite Design

In the present work, the relationship between two criteria of solvent recovery (namely permeate flux and total soybean isoflavone retention) and three controllable factors (namely temperature, pressure, and feed concentration) was studied. According to a CCD, as mentioned earlier, the resulting experimental design and response results are now presented in Table 3. It can be seen that the flux range is 1.29–20.24 $L \cdot m^{-2} \cdot h^{-1}$, and the retention range is 88.10–99.75%. But the highest flux and the highest retention did not occur under the same condition. However, runs 2, 4, 12, 13, and 18 showed acceptable fluxes and retentions. Further, good reproducibility for the central points (runs 15–20) was obtained, due to the homogeneity among the membranes of the same batch.

Table 2. NF membranes data and experiment results for membrane selection

Supplier	Membrane type	Nature[a]	MWCO[a] (Da)	Solvent flux ($L \cdot m^{-2} \cdot h^{-1}$)	Permeate flux ($L \cdot m^{-2} \cdot h^{-1}$)	Total soybean isoflavone retention (%)
Koch	MPF-44	Hydrophilic	200[b]	4.11	0.89	74.39
	MPF-50	Hydrophobic	700[c]	4.13	1.07	67.20
MET	STARMEM™ 122	Hydrophobic	220[d]	42.8	4.88	95.78
	STARMEM™ 228	Hydrophobic	280[d]	22.5	4.18	76.13
	STARMEM™ 240	Hydrophobic	400[d]	23.3	4.38	89.17

[a]Membranes data provided by the suppliers.
[b]Based on rejection of glucose in water.
[c]Based on rejection of Sudan IV (MW 384) dissolved in ethyl acetate.
[d]Based on rejection of linear alkanes dissolved in toluene.

Table 3. Experimental design matrix and the results of CCD[a]

	Variables			Response	
Standard run no.	A: Temperature (°C)	B: Pressure (kPa)	C: Feed concentration (g/L)	Permeate flux ($L \cdot m^{-2} \cdot h^{-1}$)	Total soybean isoflavone retention (%)
1	10	500	2.5	2.16	91.48
2	30	500	2.5	11.93	93.71
3	10	1500	2.5	4.17	94.72
4	30	1500	2.5	20.24	92.32
5	10	500	8.5	1.29	95.74
6	30	500	8.5	4.29	89.82
7	10	1500	8.5	2.75	99.75
8	30	1500	8.5	6.27	91.16
9	5	1000	5.5	4.19	96.01
10	35	1000	5.5	13.86	88.10
11	20	250	5.5	2.43	93.62
12	20	1750	5.5	9.12	96.91
13	20	1000	1	14.12	95.28
14	20	1000	10	6.22	96.94
15	20	1000	5.5	7.34	95.63
16	20	1000	5.5	8.77	95.97
17	20	1000	5.5	6.69	95.61
18	20	1000	5.5	9.51	96.37
19	20	1000	5.5	8.91	95.21
20	20	1000	5.5	7.65	95.83

[a]Membrane used for the study: STARMEM^{TM} 122.

ANOVA Analysis and Model Development

The fit summary revealed that the quadratic models are statistically significant for both the permeate flux and the total soybean isoflavone retention because they exhibited low standard deviations and high "Adjusted *R*-Squared" values. Therefore, the two quadratic models were used for further analysis. The results of ANOVA for the permeate flux and the total soybean isoflavone retention are shown in Table 4.

For the flux model, the *p*-value (<0.0001) implied that the model was statistically significant at 99.99% confidence level, which is desirable. A and C are the two most significant model terms on the response as compared to the other model terms studied. As all the concerned terms considered, all the three operating parameters (temperature, pressure

Table 4. ANOVA table for the regression models

Source	*p*-value for *J* (flux)		*p*-value for *R* (retention)	
	All terms included	Insignificant terms excluded	All terms included	Insignificant terms excluded
Model	<0.0001	<0.0001	<0.0001	<0.0001
A	<0.0001	<0.0001	<0.0001	<0.0001
B	0.0008	0.0003	0.0002	<0.0001
C	<0.0001	<0.0001	0.0106	0.0072
AB	0.1191	0.0990	0.0017	0.0010
AC	0.0007	0.0003	<0.0001	<0.0001
BC	0.1162	0.0964	0.0688	0.0559
A^2	0.8271	–	<0.0001	<0.0001
B^2	0.0057	0.0033	0.0914	0.0764
C^2	0.3739	–	0.9075	–
Lack of Fit	0.1771	0.2397	0.0834	0.1114

and concentration) were important for flux. The backward elimination procedure reduced A^2 and C^2 (insignificant at 90% confidence level) from the initial response surface model automatically. The *p*-values for lack-of-fit before and after excluding insignificant terms were both higher than 0.10, which shows that there was no indication of significant lack-of-fit observed at 90% confidence level. The *p*-value for lack-of-fit became higher after excluding the insignificant terms.

For the retention model, A, AC, and A^2 are the three most significant model terms on the response as compared to other model terms. The backward elimination procedure reduced C^2 from the initial quadratic model. The *p*-value for lack-of-fit before excluding insignificant terms was between 0.05 and 0.10. It means that the lack-of-fit is not significant at 95% confidence level but significant at 90% confidence level, which is not good. After excluding insignificant terms (at 90% confidence level), the *p*-value for lack-of-fit became higher and the lack-of-fit can be said to be insignificant at the 90% confidence level, which is desirable.

Table 5 presents the *R*-squared values for flux and retention models before and after excluding insignificant terms. It can be seen that, for each model, the R^2-value became slightly smaller, the adjusted R^2-value became slightly higher, and the predicted R^2-value increased relatively greater after excluding insignificant terms. For the initial flux model, the value of (predicted R^2-adjusted R^2) is greater than 0.20, which indicates the predicted R^2 was not in reasonable agreement with the adjusted

Table 5. R-Squared statistics for the regression models

	R^2		Adjusted R^2		Predicted R^2	
Model	All terms included	Insignificant terms excluded	All terms included	Insignificant terms excluded	All terms included	Insignificant terms excluded
Flux	0.9532	0.9490	0.9112	0.9192	0.7030	0.7915
Retention	0.9743	0.9743	0.9512	0.9556	0.8279	0.8498

R^2 and there was a problem with either the data or the model. After excluding insignificant terms, the predicted R^2 became in reasonable agreement with the adjusted R^2. The predicted R^2-values were both in reasonable agreement with the adjusted R^2-values for the initial and reduced retention models. After excluding insignificant terms, the predicted R^2-values became closer to the adjusted R^2-values. On the whole, these R-squared values look very good. In addition, for the two initial models, the values of "Adeq Precision" were 17.85 and 27.62, respectively. After excluding insignificant terms, the values of "Adeq Precision" became higher, 20.93 and 30.46 respectively. They were both greater than 4 and indicate adequate signals.

It can be said that the models became better after excluding insignificant terms at 90% confidence level. The finial predictive equations obtained (after excluding insignificant terms) for flux (J) and retention (R) in terms of the coded factors are as given below:

$$J = 8.56 + 3.75\mathrm{A} + 1.90\mathrm{B} - 2.86\mathrm{C} + 0.85\mathrm{AB} - 2.42\mathrm{AC} - 0.86\mathrm{BC} - 1.54\mathrm{B}^2 \quad (6)$$

$$R = 95.84 - 2.12\mathrm{A} + 0.97\mathrm{B} + 0.54\mathrm{C} - 0.91\mathrm{AB} - 1.79\mathrm{AC} + 0.44\mathrm{BC} - 1.78\mathrm{A}^2 - 0.35\mathrm{B}^2 \quad (7)$$

The response equations above permitted the evaluation of the factor effects. The positive sign in front of the terms indicates a synergistic effect, while the negative sign indicates an antagonistic effect. Figures 2 and 3 show the experimental values versus the predicted values using the two model equations developed. The R^2-values of the two plots are 0.9490 and 0.9743, respectively. The results indicate that the two models were both in good agreement with the experimental results and they can be used to predict the permeate flux and total soybean isoflavone retention within the limits of the experiment.

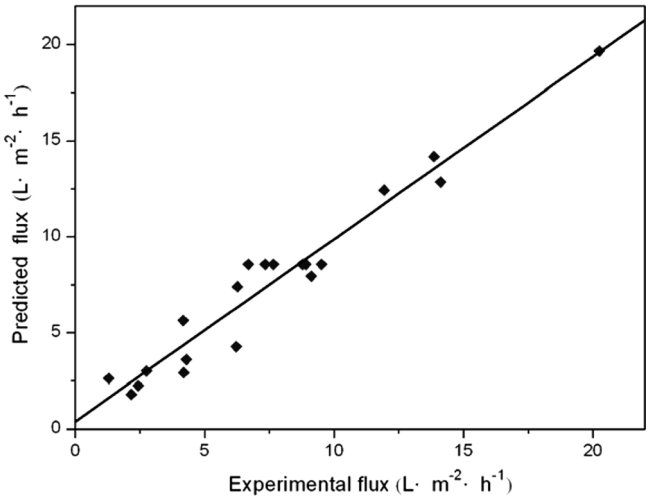

Figure 2. Experimental values vs. predicted values for permeate flux.

Effect of Process Variables on Permeate Flux

In order to gain a better understanding of the results, the three-dimensional (3D) response surface plots were used to predict the relationships between the independent variables and the dependent variables.

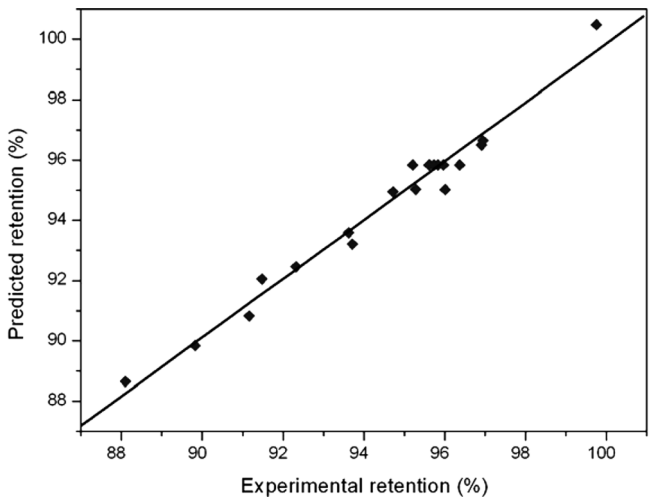

Figure 3. Experimental values vs. predicted values for total soybean isoflavone retention.

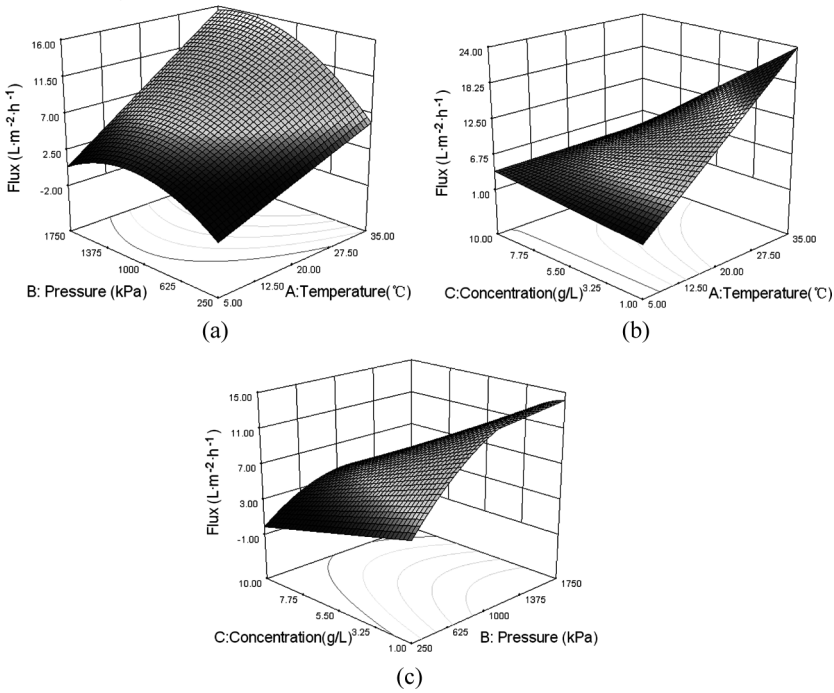

Figure 4. Response surface graph (3D) showing the effect of process variables on permeate flux.

Figure 4 shows the 3D response surfaces of the permeate flux. The plots show the effects of two factors on the response at a time and the other variable condition was maintained at zero level in all the presented figures.

As can be seen from Figs. 4a and 4b, temperature had a positive effect on the permeate flux. It is usually hypothesized that this trend is due to the following probable reasons. In the first place, the viscosity of the feed solution reduces with increasing temperature, which enhances the mass transfer of each component. Further, the diffusion coefficient increases when the operating temperature goes up. In addition, the average pore size of the membrane increases slightly with increasing temperature.

Generally, high operating pressure results in high permeate flux, which has been observed in many reports. This phenomenon can be explained on the basis of the solution-diffusion mechanism (18). In our work, as can be seen from Figs. 4a and 4c, the permeate flux increased with pressure at low level. However, the flux declined when the pressure

was higher, which was more obvious at low temperature or high concentration. A similar phenomenon was also observed by other researchers (19). This is probably due to the membrane blocking or fouling resulting from the concentration polarization. After one batch nanofiltration at high pressure in our study, especially at low temperature and high concentration, visible agglomerates were observed on the membrane surface. Because of concentration polarization, the total solids concentration on the membrane surface will be higher than the feed concentration. Further, if the surface concentration is higher than the solubility, surface precipitation may form and cause the blocking of the membrane.

Figures 4b and 4c indicate that feed concentration had a negative effect on the permeate flux. This can be explained by the concentration polarization phenomenon. Concentration polarization problems often exist in many nanofiltration processes when solute concentrations are high. As a result, many researchers chose a dilute solution to minimize the concentration polarization for the purpose of the basic theoretical research. However, in actual applications, solutes will be more concentrated. The feed concentrations in our experiments were in the range of 1.0–10.0 g/L, therefore, concentration polarization phenomena were observed.

Effect of Process Variables on Total Soybean Isoflavone Retention

Figure 5 presents the 3D response surfaces of total soybean isoflavone retention. As before, the figures are based on the retention model with one variable kept constant at its zero level and varying the other two variables within the experimental range.

Several researchers have focused on the effect of process variables on the performance of NF-membranes. However, the results for retention were not exactly the same in different solvent-solute-membrane systems. Based on the results of analysis and plots presented in Fig. 4, the total soybean isoflavone retention increased with pressure (Figs. 5a and 5c), probably due to the relative increase in the diffusivity of solvent through the NF-membrane. As can be seen from Figs. 5b and 5c, the feed concentration also had a positive effect on the total soybean isoflavone retention, probably due to the concentration polarization. As far as the temperature was concerned, the plots (Figs. 5a and 5b) indicate that retention decreased with increasing temperature when the pressure and concentration were at high levels. However, when the pressure and concentration were low, the retention increased at low temperature and then declined when the temperature went beyond a certain limit.

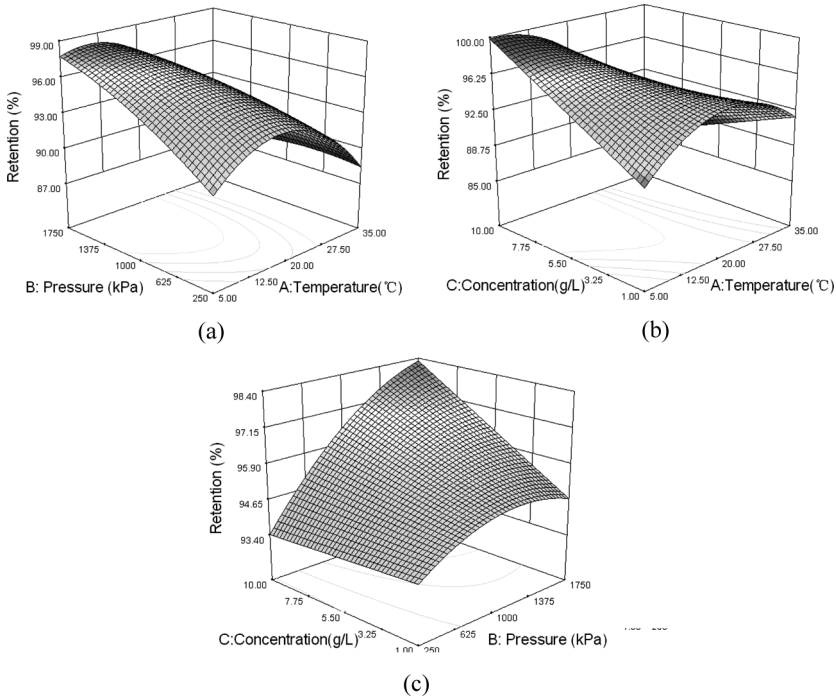

Figure 5. Response surface graph (3D) showing the effect of process variables on total soybean isoflavone retention.

Confirmation Tests

In order to check the model adequacy, four confirmation experiments were performed for the permeate flux and total soybean isoflavone retention. The results of the confirmation tests and the comparison of experimental and predicted values are now listed in Table 6. As can be seen from Table 6, the experimental response values were close to the predicted ones and the percentage errors calculated were small. The percentage error ranges between the experimental and the predicted value of permeate flux and total soybean isoflavone retention lie within 3.77% to 14.76% and 0.15% to 2.77%, respectively. It demonstrates that the obtained Eqs. (6) and (7) are both accurate models.

It can be said that the final reduced models developed through CCD are reasonably accurate and can be used to successfully predict the permeate flux and the total soybean isoflavone retention for any combination of the temperature, pressure, and feed concentration within the range of the experimentation conducted.

Table 6. The results of confirmation tests[a]

	Parameters			Permeate flux ($L \cdot m^{-2} \cdot h^{-1}$)			Soybean isoflavone retention (%)		
Run	Temperature (°C)	Pressure (kPa)	Concentration (g/L)	Experimental value	Predicted value	Error (%)	Experimental value	Predicted value	Error (%)
1	25	1000	10	5.08	4.33	14.76	92.78	93.80	1.10
2	35	1500	5	16.43	17.05	3.77	90.7	88.19	2.77
3	10	500	1	1.84	1.60	13.04	92.83	91.11	1.85
4	15	1750	7.5	3.79	3.48	8.18	99.35	99.20	0.15

[a]Membrane used for the study: STARMEM™ 122.

CONCLUSIONS

The present study has demonstrated the feasibility of using nanofiltration for the solvent recovery from aqueous ethanol solution of soybean isoflavones and the feasibility of CCD application for studying the effect of process variables on this nanofiltration process. Among the five chosen membranes, STARMEM^{TM} 122, selected for further experiments, has shown higher permeate flux and retention of total soybean isoflavones. In this CCD, temperature, pressure, and feed concentration were chosen as independent variables, and the responses were the permeate flux and total soybean isoflavone retention. Within the limits of the conditions studied, the flux increased with the increasing temperature and decreasing feed concentration. Pressure had a positive effect at low pressure, but flux declined when the pressure became higher, due to surface precipitation resulting from concentration polarization. In the case of retention, the pressure and feed concentration both had positive effects. While the influence of temperature was a bit complicated. Finally, acceptable flux and high retention were obtained and the CCD could be useful to model this nanofiltration process.

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