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Effect of Operating Parameters on the Separation of Sugars by Nanofiltration

NIHAL AYDOĞAN, TÜRKER GÜRKAN, and LEVENT YILMAZ*
DEPARTMENT OF CHEMICAL ENGINEERING
MIDDLE EAST TECHNICAL UNIVERSITY
06531 ANKARA, TURKEY

ABSTRACT

Due to the complexity of fermentation mixtures, separation of solutes and secondary substrates has a special importance. Membrane processes such as nanofiltration may offer good alternatives for the separation of fermentation products or recovery of substrates. For an efficient separation, the membrane type and operating parameters such as feed flow rate, operating pressure, and feed should be optimized. In this study the separation and recovery of sugars were targeted. It was found that with an increase of the feed flow rate, permeate flux increased since the effect of concentration polarization was minimized. As a result, experiments were carried out at the highest possible flow rate. The effect of pressure was studied at five pressures (10–50 bar). It was found that there is a linear relationship between the pressure and permeate flux up to 30 bars. Beyond 30 bars the effect of pressure became less significant. Thus, 30 bars was chosen as the operating pressure. To investigate the effect of concentration, 1 to 10 weight percentage sucrose and glucose solutions were utilized. It was observed that with an increase in concentration, permeate flux decreased and rejections increased, finally reaching a limiting value. Binary solutions of sucrose and glucose were also studied. It was seen that the separation factor slightly decreased, probably due to a glucose–sucrose interaction. Experimental data were used with a mathematical model to predict the permeate flux and rejection. Good agreement of the predicted results was obtained with the experimental data for a 500 MWCO membrane.

Key Words. Nanofiltration; Sucrose; Glucose; Permeate flux; Rejection

* To whom correspondence should be addressed.

INTRODUCTION

Product recovery from fermentation mixtures is particularly difficult since they contain many types of compounds with different physicochemical properties, further increasing the complexity of the separation. In addition, many products are heat labile and can be adversely affected by evaporation. Thus membrane processes may offer good alternatives for product recovery. Microfiltration and ultrafiltration are becoming well established in many primary downstream operations in the biotechnology and fermentation industries, e.g., to separate cells from fermentation broths (1). Reverse osmosis and nanofiltration are promising separation methods for the recovery of microsolutes. Nanofiltration has some advantages over other methods of removing water and recovering low molecular weight species. In recent years the study of the application of nanofiltration to fermentation broths has increased. In most cases nanofiltration membranes may not allow glucose or sucrose to permeate through, but they permeate most low molecular weight fermentation products. Thus, continuous removal of end products permits continuous fermentation with high substrate levels resulting in an increased production rate (1). To increase the productivity of the fermentation and to recover the sugars from the outlet stream, the use of nanofiltration with fermentation processes is recommended. Moreover, in a newly developed process for the production of ethyl alcohol, withdrawal of the bleed stream is proposed (2). This bleed stream contains unconverted sugars, several organics, and water. By recovering sugars from the bleed stream, not only is the amount of waste minimized but the energy expended for sterilization is also minimized. Before applying nanofiltration to the bleed stream, the separation performance of sucrose and glucose should be studied and the operating parameters should be optimized since real mixtures are quite complicated. There are a number of studies on the separation of sugars in the literature. In a study by Kimura and Sourirajan (3), the separation of sucrose was investigated using a cellulose acetate membrane. It was found that for a given feed solution and feed concentration, an increase of operating pressure increases both rejection (R) and permeate flux (J_v); for a given operating pressure and feed concentration, both R and J_v increase with an increase in feed rate; and, for a given operating pressure and feed rate, J_v decreases and R increases at first and then passes through a slight maximum, with an increase in feed concentration (3). The separation capacity of a binary solution of sucrose and glucose was studied using ZF-99 membranes (4). All these studies focused on predicting the permeate flux during reverse osmosis of highly concentrated multisolute solutions such as fruit juice. In the case of application of nanofiltration to fermentation products, the concentrations of sugars are very low. Therefore, an investigation of the performance of nanofiltration at low sugar concentrations when fermentation

applications are targeted would be useful. Pinho et al. studied the separation of the glucose–ethanol–water system using a cellulose acetate membrane (5). They studied a case in which a reverse osmosis unit was connected to a yeast cell reactor. They observed that rejection of ethanol decreased with increasing glucose concentration. This showed that solutes in the solution may affect the separation performance. The bleed stream of the fermenter contains sucrose and glucose. Interactions of these compounds with each other and with the membrane can affect the separation efficiency and should be investigated.

EXPERIMENTAL

Materials

Two different types of asymmetric polyamide membranes with 500 MWCO (Berghof, BM5) and 200 MWCO (Berghof, BM2) were used during our nanofiltration experiments. For prefiltration purposes, 0.2 μm cellulose acetate membranes were used. Sucrose (technical), glucose (technical), hydrogen peroxide (Merck), sulfuric acid (Merck), phenol (Merck), and distilled water were also utilized.

Berghof's high-pressure filtration unit, shown in Fig. 1 was used during the nanofiltration experiments. The system is equipped with a high-pressure

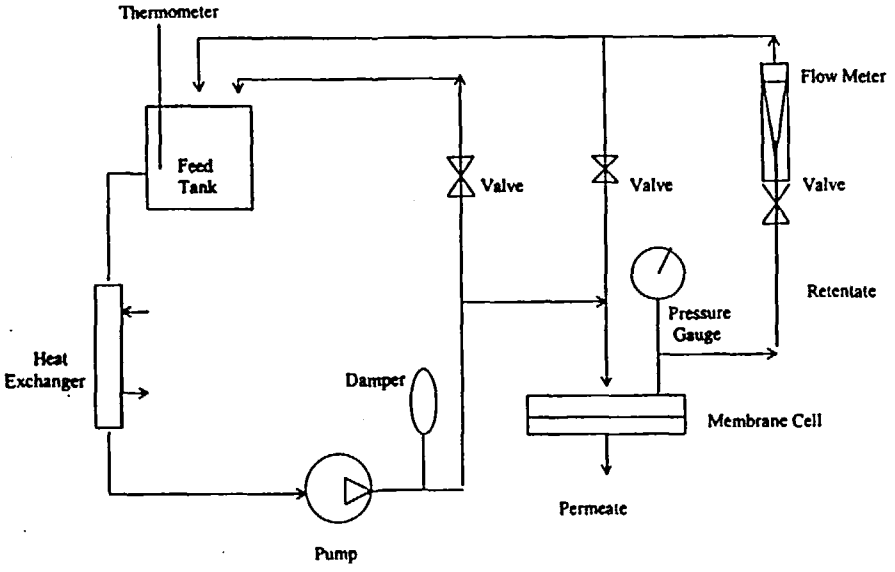

FIG. 1 Experimental setup.

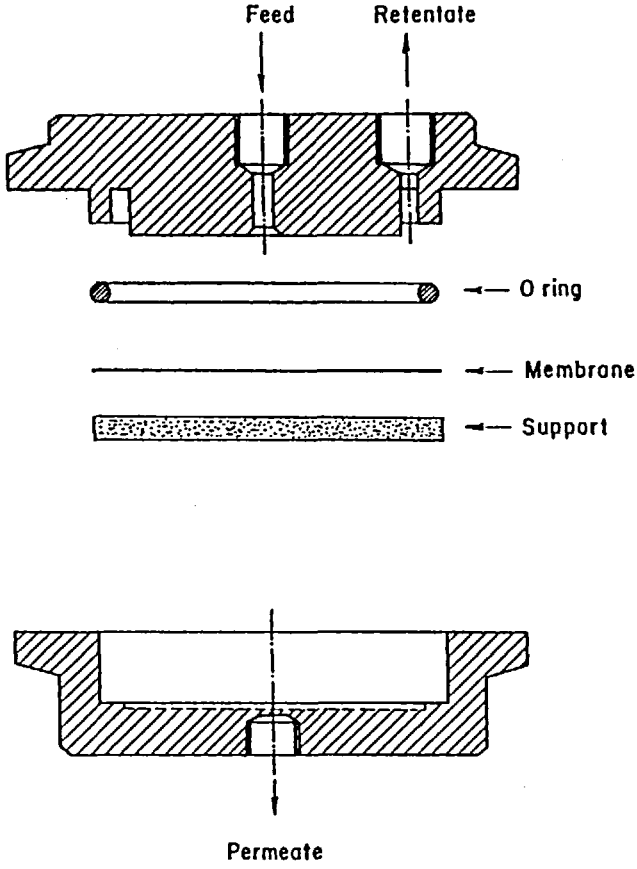

FIG. 2 Schematic representation of membrane module.

pump of three plungers capable of maintaining a liquid system pressure of up to 60 bar. Pulsation in the pressure was prevented by using a 30-bar pulsation damper. The system includes a chamber which holds a flat membrane element of 7.6 cm diameter. The flow geometry of the membrane cell is designed to obtain crossflow to minimize concentration polarization and membrane fouling. A detailed drawing of the membrane module is shown as Fig. 2. Feed solution enters the module from the middle hole of the upper part and is spread over the membrane and leaves the cell from the side hole. As a result of high pressure, the permeate flows through the membrane. The flow rate and operating pressure are manipulated independently by stainless-

steel needle valves located at the outlet of the module. Temperature is controlled with a countercurrent double pipe heat exchanger.

Procedure

Distilled water was further treated by filtering it through a reverse osmosis membrane with 99.2% NaCl rejection. This processed water was accumulated, and 2000–5000 ppm hydrogen peroxide was added to prevent microbiological growth. All test solution were prefiltered through a 0.2 μm cellulose acetate membrane. All reported sugar concentrations are on a weight basis.

Total circulation experiments were performed by returning the permeate and retentate to the feed tank to prevent a concentration change of the feed solution. During experiments, the permeate flux was measured at certain time intervals and samples were taken for concentration measurements. All nanofiltration runs were performed at constant temperature (25°C).

At the beginning of each nanofiltration experiment, distilled water was circulated and the pure water permeate flux of the membrane was measured. The feedwater was then replaced with the test solution, and experiments were continued until steady-state was reached. Using samples taken at the assumed steady state, the concentration of the permeate, c_p, was measured. Samples from the feed solution at the beginning and at the end of experiments were also taken. The average of these two measurements was used as the feed concentration, c_b. Rejection values were calculated by using the equation

$$R = 1 - \frac{c_p}{c_b} \tag{1}$$

Experimental conditions were as follows: feed concentration, 1, 3, 5, 7, 10 wt% sucrose and glucose; applied pressure, 10, 20, 30, 40, 50 bar; feed flow rate, 5, 10.8, 18.9, 29.7 $mL \cdot s^{-1}$; feed temperature, 25°C.

Selectivity, α, is defined by

$$\alpha = \frac{y_{Glu}/y_{Suc}}{x_{Glu}/x_{Suc}} \tag{2}$$

where y and x represent the mole fractions of the permeate and the feed stream, respectively.

The concentration of total sugar was determined by the sulfuric acid–phenol method described by DuBois et al. (6). Concentrations of sucrose and glucose in single component experiments were also determined by this method. The concentration of glucose was also measured using a D-glucose analyzer (YSI 1500 sidekick). In this method, glucose is converted to hydrogen peroxide by special enzymes and its concentration is measured. Due to the presence of hydrogen peroxides in the solution, sugar analyses were car-

ried out 10 days after taking the samples. This time was chosen after measuring the decomposition rate of hydrogen peroxides. The concentrations of glucose and sucrose in binary mixtures were measured by HPLC (Shimadzu CR4A Chromotopac) using a Waters differential refractive index detector and an organic acid column (7).

RESULTS AND DISCUSSION

To characterize the membrane and to provide a frame of reference, data on pure water flux were obtained for each membrane before starting the measurements with sugar solutions. The membrane performances are generally reported at steady-state. Therefore, determination of the approach to steady-state is important. Figure 3 shows an example of the approach to steady-state for permeate flux and rejection. The time required to reach steady-state was in the range of 80–140 minutes, depending on the operating conditions. Both permeate flux and rejection were seen to reach steady-state at approximately

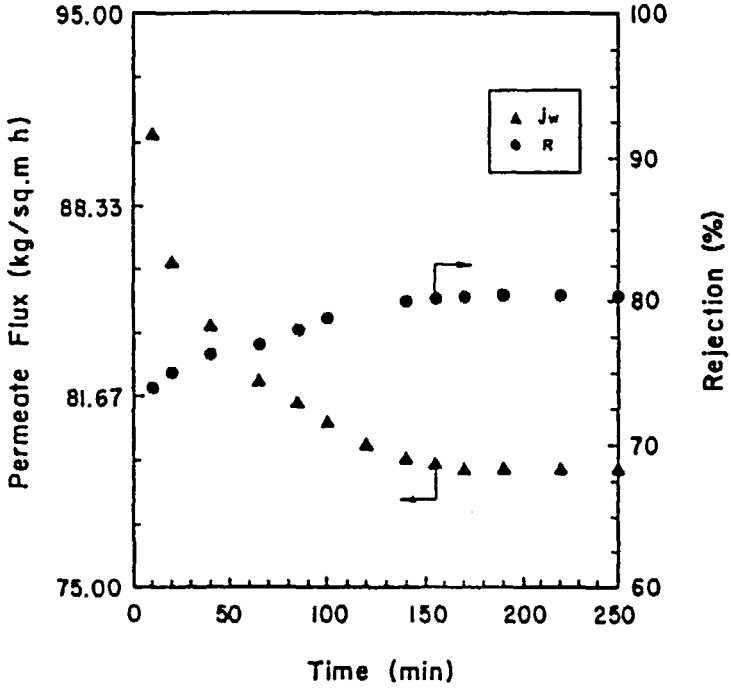

FIG. 3 Variation of permeate flux and rejection with time.

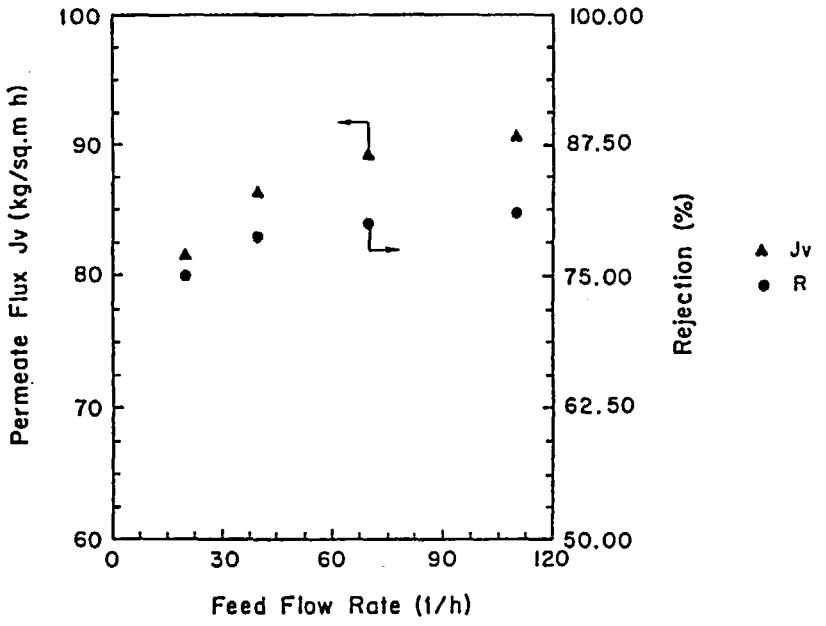

FIG. 4 Effect of feed flow rate on permeate flux and rejection (500 MWCO membrane, 30 bar).

the same time. Therefore, it was concluded that the approach to steady-state could be determined based on permeate flux measurements, which were much easier to carry out. Additionally, to see if there was any fouling on the membrane during the operation, an experiment was continued for 18 hour and no changes in flux and rejection were observed. Based on this observation and by visual inspection of the membrane by microscope, we concluded that fouling was not taking place to a significant extent with our solutions. Since our solutions were dilute, very clean, and without particulate matter, this result was expected.

Concentration polarization is an important phenomenon affecting membrane performance. In this study, crossflow conditions are dominant within the membrane chamber; thus, concentration polarization is minimized by adjusting the feed flow rate. Four different flow rates were used with a 1% sucrose solution and a 500-MWCO membrane. The results of this study are given in Fig. 4, which shows that the permeate flux increases with increasing feed flow rate, probably because of the decreasing effect of concentration polarization. Permeate flux increases first with the feed flow rate and then levels off. With the increase of

the flow rate from 70 to 110 L/h (18.9–29.7 mL/s), there is only a 2% increase in the observed permeate flux. Beyond this point, concentration polarization has no significant effect. Moreover, rejection is not highly affected by the feed flow rate in this region. Therefore it can be concluded that concentration polarization has no significant effect for the high end of the feed flow rate range in our equipment. Thus, it was decided to carry out further measurements at the highest possible feed flow rate.

Since the hydrostatic pressure difference is the driving force of the separation process and the applied pressure may alter the internal state of the membrane, the effect of pressure on membrane performance was investigated in detail (Figs. 5 and 6). As seen from Fig. 5, there is a linear relationship between pressure and permeate flux up to 30 bar. Beyond this pressure the effect of operating pressure on the permeate flux decreases as the resistance in the boundary layer of the membrane increases to a limiting flux value J_∞. Similar trends were ob-

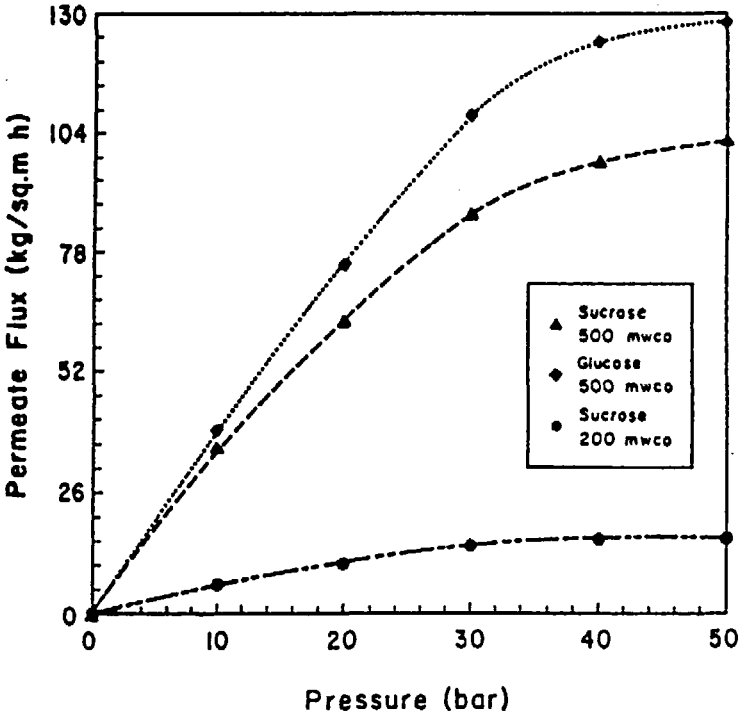

FIG. 5 Variation of permeate flux with pressure (1% sucrose and 1% glucose solutions, 500 and 200 MWCO membranes).

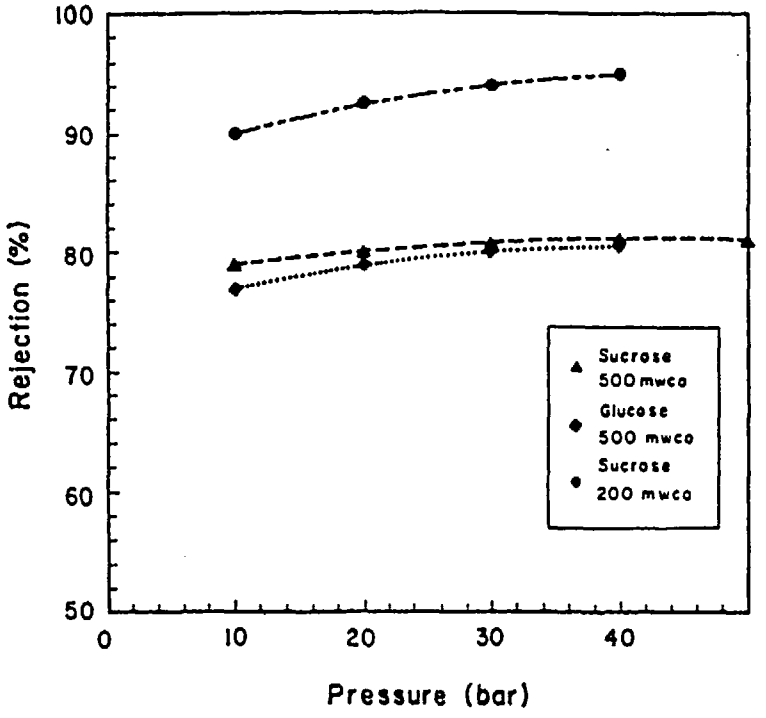

FIG. 6 Variation of rejection with pressure (1% sucrose and 1% glucose solutions, 500 and 200 MWCO membranes).

served when glucose was used as a microsolute instead of sucrose and when a denser 200 MWCO membrane was employed. Slopes of the linear parts (i.e., permeability coefficient) are given in Table 1. When the permeability coefficients are compared, it is seen that the value of the coefficient is the highest for a glucose solution with a 500-MWCO membrane. This is probably because of molecular size difference; due to its small dimensions, permeation of glucose is

TABLE 1
Permeability Coefficients for Different Solutes and Membranes

	1% Sucrose (500 MWCO)	1% Glucose (500 MWCO)	1% Sucrose (200 MWCO)
Permeability coefficient	2.86	3.60	0.49

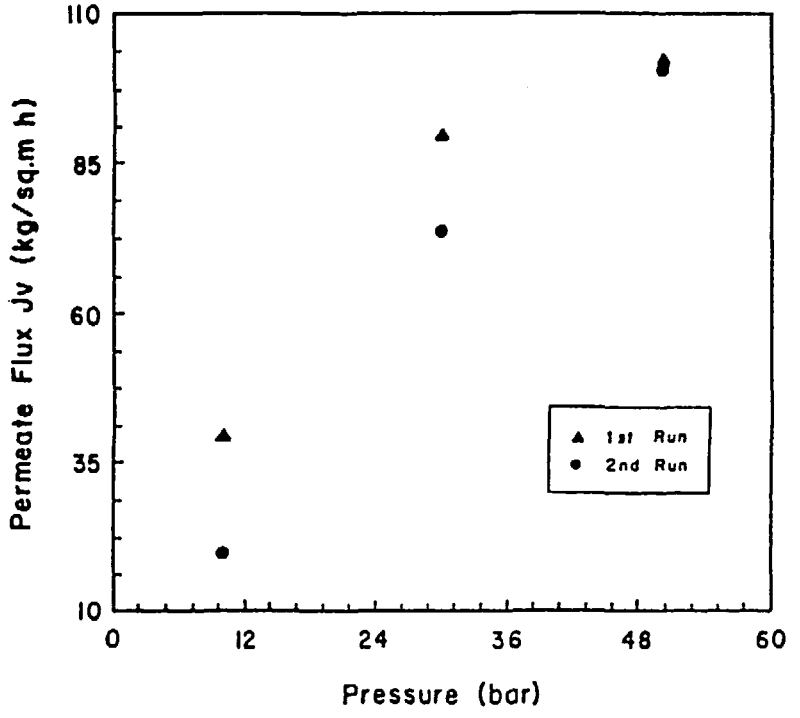

FIG. 7 Compaction of membrane (1% sucrose solution, 500 MWCO membrane, 29.7 mL/s feed flow rate).

easier. As expected, the permeability coefficient is significantly lowered when a 200-MWCO membrane is used.

On the other hand, rejection is not affected to a great extent by pressure, as demonstrated in Fig. 6. Rejections obtained using a 500-MWCO membrane for both sucrose and glucose are approximately 80%. The important observation is that the rejections of sucrose and glucose by the membrane do not vary significantly in spite of the major difference in their molecular sizes. This shows that molecular sieving is not necessarily the only mechanism which controls separation (8). Although rejections obtained with a 1% sucrose solution using a 200-MWCO membrane were the highest (about 90%), it can be concluded that switching from a 500-MWCO membrane to a 200-MWCO membrane does not increase rejections dramatically when it is considered that the molecular weight of sucrose is in between these values, i.e., 360. These observations strongly imply that other factors (such as molecular agglomeration, membrane solute interactions, etc.) also play an important role in the separation mechanism.

Since high operating pressures are applied in nanofiltration processes, membrane structures may be affected. To observe this effect, two sets of experiments were performed using a 1% sucrose solution and a 500-MWCO membrane. The experiments were initially performed at 10, 30, and 50 bars sequentially. After that the same sequence was repeated with the same membrane and solution, from the lowest to the highest pressure. Results of these measurements are shown in Fig. 7. Differences in permeate flux in the two sets are greatest at 10 bar (approximately 50%). This difference becomes smaller at 30 bar, and at 50 bar there is no difference in the permeate flux. When the highest pressure used is 30 bar, permeate fluxes measured at 10 bar before and after compaction remained the same. These findings verify that a membrane is significantly compacted at 50 bar. Similar type of experiments were carried with the 200-MWCO membrane. Compaction was also observed for this type of membrane (Fig. 8); however, because of its denser structure and smaller pore size, membrane performance was affected less by

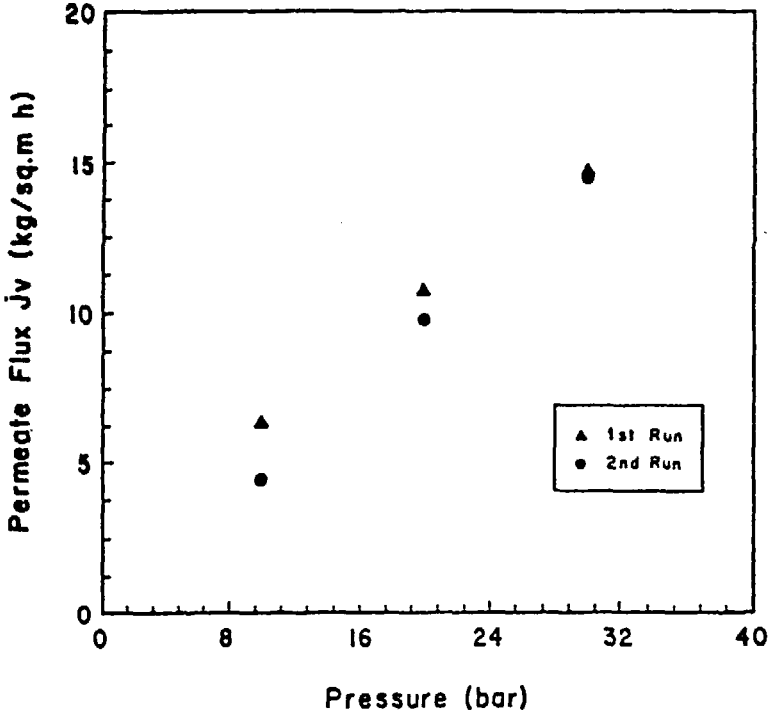

FIG. 8 Compaction of membrane (1% sucrose solution, 200 MWCO membrane, 29.7 mL/s feed flow rate).

compaction. Based on these studies, 30 bar, which also marks the end of the linear region in the pressure vs flux curve, was chosen as the working pressure for further experiments.

Experiments on the effect of the feed concentration covered the range of 1 to 10% sucrose and glucose solutions with 500 and 200 MWCO membranes. Results shown in Figs. 9 and 10 indicate that there is a sharp decrease in the permeate flux and a sharp increase in rejection with increasing concentration in the low concentration range. After a certain concentration, about 3%, the effect of concentration decreases significantly, most probably because the concentration boundary layer becomes thicker and an increase in concentration does not have a further significant effect on separation performance. Feed and permeate concentrations can also be seen in Table 2, which may help to understand how the quality of the permeate stream changes as the feed concentration increases. Unlike the effect of pressure, rejection is strongly

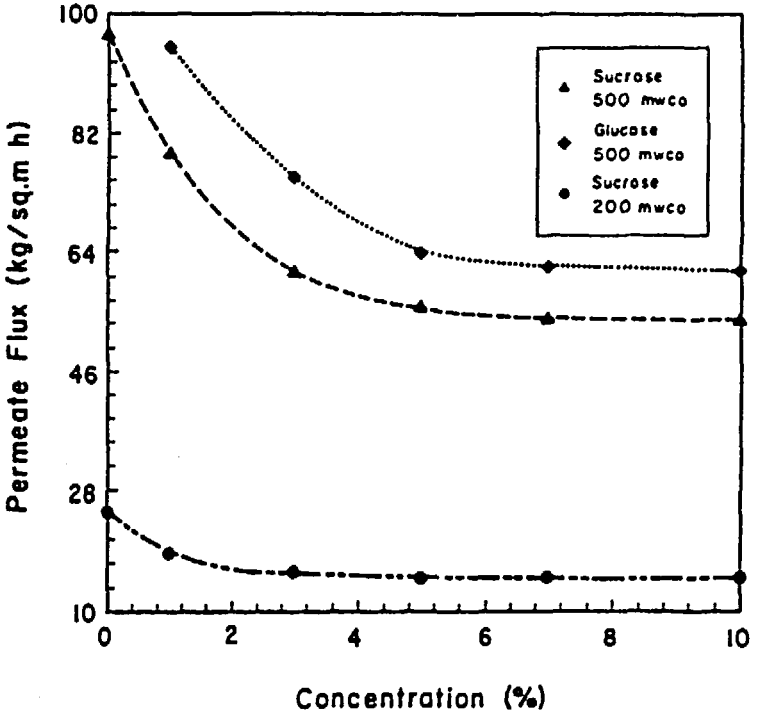

FIG. 9 Variation of permeate flux with concentration (500 and 200 MWCO membranes, 30 bar, 29.7 mL/s feed flow rate).

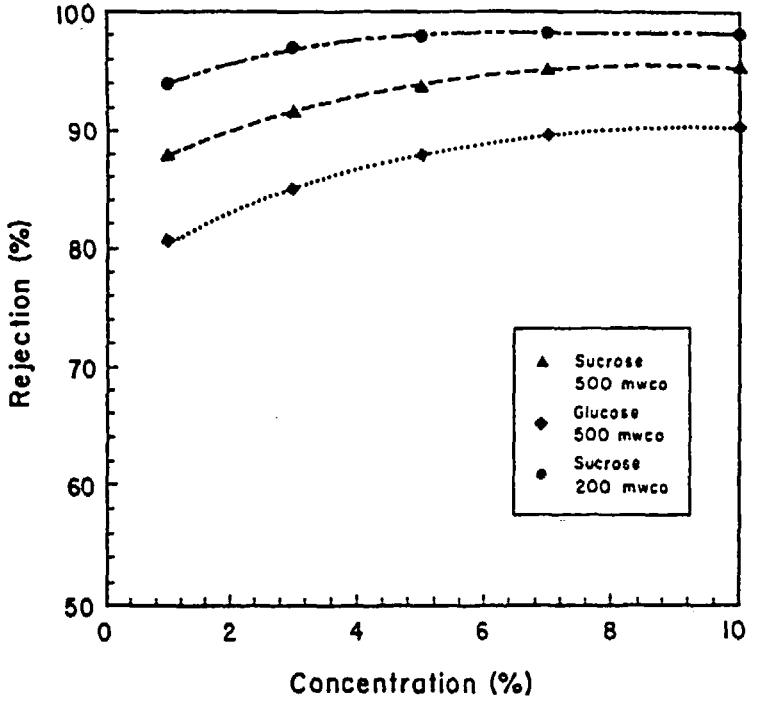

FIG. 10 Variation of rejection with concentration (500 and 200 MWCO membranes, 30 bar, 29.7 mL/s feed flow rate).

TABLE 2
Variation of Permeate Concentration with Respect to Feed Concentration[a]

Feed concentration (%)	Permeate concentration (%)		
	Sucrose (500 MWCO)	Glucose (500 MWCO)	Sucrose (200 MWCO)
1	0.12	0.19	0.06
3	0.25	0.45	0.09
5	0.31	0.60	0.10
7	0.34	0.73	0.12
10	0.46	0.96	0.18

[a] *P*: 30 bar. Feed flow rate: 29.7 mL/s.

TABLE 3
Results of Experiments with Binary Mixtures[a]

System	J_v (kg/m^2·h)	R (%)
2% Sucrose	68.4	94.0
2% Glucose	70.4	81.2
1% Sucrose + glucose (1:1)	72.0	91.3
10% Sucrose	50.4	86.4
10% Glucose	54.2	95.0
5% Sucrose + glucose (1:1)	56.5	95.0

[a] Membrane MWCO: 500. P: 30 bar. Feed flow rate: 29.7 mL/s.

affected by concentration. Variation in concentration affects permeate fluxes and rejections in the same manner for all of the solute and membrane types used, indicating a similarity in the mechanism. The effect of concentration on membrane performance is very strong up to 5% for glucose but thereafter the effect of concentration is much smaller. Due to its molecular size, rejection of glucose is lower than that of sucrose. Permeate fluxes obtained with the 200-MWCO membrane were smaller and rejections were higher because of the denser structure of the membrane. Observed trends of permeate flux and rejection with a concentration change are similar to those reported in the literature for various solutes and membranes (3, 9).

To investigate the effect of solute–solute interaction on membrane performance, binary mixtures of sucrose and glucose were employed. In this part of the study, 500 MWCO membranes and 30 bar upstream pressure were used. Two and 10% total sugar solutions, containing equal amounts of sucrose and glucose by weight, were used as feed solutions. Flux rejection and selectivity values obtained by using these solutions were compared with values obtained from single component experiments. The results are presented in Tables 3 and 4. Mixture selectivities were calculated by using Eq. (2), and permeate concentrations were measured directly for mixtures. Ideal selectivi-

TABLE 4
Separation Factor of Glucose for Ideal and Binary Systems[a]

	Ideal	Mixture
$\alpha_{Glu/Suc}$ (2%)	3.13	2.43
$\alpha_{Glu/Suc}$ (10%)	2.72	2.33

[a] Membrane MWCO: 500. P: 30 bar. Feed flow rate: 29.7 mL/s.

ties were calculated by using the rejection values reported in Table 4 for single-component-containing solutions by using

$$\alpha_{ideal} = \frac{1 - R_{Glucose}}{1 - R_{Sucrose}} \quad (3)$$

Selectivities obtained by using multicomponent mixtures are slightly smaller than ideal ones. The results indicate that some glucose is retained together with sucrose, therefore glucose rejections are increased relative to single-component experiments. Solute–solute interaction and changes in the structure of the layers formed on the top of the membrane and within the pores are the probable reasons for this difference. This reduction in selectivities is less significant at higher total sugar concentrations due to high rejection of both solutes. Our low selectivity values indicate that nanofiltration is not a very suitable method for fractionation of sugars, as also reported in the literature (10). But in the application of nanofiltration to real fermentation systems, the main aim is to recover the total sugars. The results of multicomponent experiments show that besides glucose, rejection of total sugars increases without any decrease in flux values. Therefore, the performance for total sugar recovery increases, encouraging nanofiltration studies with bleed streams.

Modeling Studies

To predict permeate flux and rejection by using a minimum amount of experimental data, the mechanism of nanofiltration must be understood and a mathematical model should be developed. As already discussed, permeate flux and rejection depend on three operating parameters: pressure, concentration, and feed flow rate. As a preliminary step to process simulation, the mathematical relationship of permeate flux and rejection as a function of process conditions has to be established. Traditionally, correlations of the type $J_v = f(p, c_b)$ and $R = f(p, c_b)$ have been established as polynomial functions by parameter fitting of the experimental data (11). Since the polynomial functions have no physical basis, a large number of experimental data are needed for determination of the permeate flux and rejection under different process conditions. Therefore, mass transfer models should be used for the calculation of permeate flux and rejection.

Several models for mass transport in membranes have been developed in the literature. A theoretical description was given for the following models (11):

- Irreversible thermodynamics
- Frictional model
- Solution diffusion model
- Solution diffusion imperfection model
- Preferential adsorption capillary model
- Diffusion viscous flow model
- Finely porous model

It was claimed that all the models mentioned are special cases of the mathematical model described as a "statistical-mechanical theory" by Mason and Lonsdale (11). These models are derived for reverse osmosis membranes and may not be directly applicable for nanofiltration membranes in which the transport mechanism may differ. Most of these studies used NaCl solution data for comparison. In the case of organic solutions, interaction of the membrane with the solutes may alter the situation.

In this study the mathematical model of Mason and Lonsdale (11) was used to calculate the permeate flux and rejection in the desired pressure and concentration ranges of the solute. Flux and rejection are described by four parameters values which depend on solute, membrane, temperature, and flow velocity across the membrane. Our aim is to calculate these parameters by using the minimum amount of experimental data. A similar investigation using an ethanol–water solution was carried by Niemi et al. (12).

Permeate flux and rejection are described by the following equations:

$$J_v = (D_1 c_w + D_2)\Delta p_m \tag{4}$$

$$\frac{1}{R} = \frac{P}{\sigma}\frac{1}{J_v} + \frac{1}{\sigma} \tag{5}$$

Concentration at the membrane surface was calculated using Eq. (6) (13):

$$c_w = c_b + (c_b - c_p)(e^{J_v/k} - 1) \tag{6}$$

As already discussed in the Experimental part of the study, separation performance strongly depends on the boundary layer of the membrane. Thus, the effect of this layer must be considered. The effective pressure difference is expressed by

$$\Delta p_m = \Delta p - \sigma \Delta \Pi \tag{7}$$

where Δp is the pressure difference and $\Delta \Pi$ is the osmotic pressure difference across the membrane. In the study of Niemi, $\Delta \Pi$ was calculated by Eq. (8). This equation does not give good results for sucrose. Therefore, literature values of sucrose solutions were used in our calculations.

$$\Delta \Pi = (c_b - c_p)RT \tag{8}$$

The mass transfer coefficient k may be calculated from available correlations (14). The following correlation was used in this study because it produced good results in a study which employed a membrane unit with a similar type of flow pattern (15):

$$N_{Sh} = 0.046(N_{Re})^{0.75}(N_{Sc})^{0.33} \tag{9}$$

Experimental data of a sucrose solution with a 500-MWCO membrane at the lowest and highest concentrations were used in the calculation of parameters. Characterization of parameters can be performed as follows: 1) Calculate $1/R$, $1/J_v$, Δp_m, and c_w using the experimental data; 2) fit this data to

TABLE 5
Parameters of Statistical Mechanical Model Calculated from Minimum Amount of Experimental Data

Parameters (defined in text)	500 MWCO membrane	200 MWCO membrane
C_1 ($m \cdot s^{-1}$)	−1.0E-2	3.8E-7
C_2	1.2	1.1
D_1 ($m \cdot Pa^{-1} \cdot s^{-1}$)	−6.2E-11	−6.2E-12
D_2 ($kg \cdot m^{-2} \cdot Pa^{-1} \cdot s^{-1}$)	1.0E-9	1.1E-9

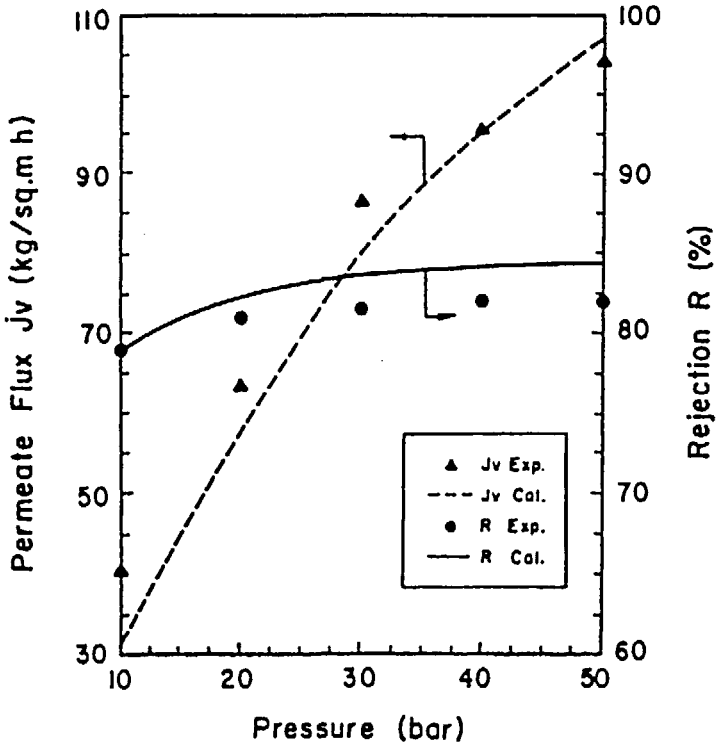

FIG. 11 Results of modeling study with 500 MWCO membrane and 1% sucrose solution.

Eqs. (2) and (4). Table 5 shows these calculated parameters, where C_1, C_2, D_1, and D_2 represent diffusion, selectivity, flow factor, and membrane constant, respectively (12). Figure 11 shows both calculated and experimental values of the permeate flux and rejection. The relative error between measured and calculated values for rejection ranged from 1.7 to 3.0% and for permeate flux the deviation ranged from 2.7 to 23.0%. Since the aim of the original model (12) was to predict the model parameters by using a minimum amount of data, the same strategy was followed in this work. As the amount of experimental data used to predict the coefficients increases, this error becomes smaller. Moreover, the model we used was derived for a different system, and our aim is not to produce a new model which gives the best fit for our data but to show the applicability of general models. Therefore, the fit between experimental and modeling results, shown in Fig. 11, is quite satisfactory for our purposes.

CONCLUSIONS

It this study the effect of operating parameters on the performance of nanofiltration for the separation of sugars was determined. It was observed that increasing the feed flow rate increases the permeate flux and rejection by minimizing the effect of concentration polarization. Since the effect of the flow rate is diminished at the high end of the feed flow rate range, it can be concluded that for this flow geometry, mixing provided by the highest flow rate, 29.7 mL/s, provides satisfactory mixing. Permeate fluxes increased with increasing upstream pressures linearly up to 30 bar. Membranes severely compacted at higher pressures. Increasing the pressure did not affect rejection significantly. Only in the low range did an increase in solute concentration cause important decreases in permeate flux and increases in rejection. After a critical concentration level (which is higher for glucose than for sucrose), permeate fluxes and rejection values remained relatively constant. Our results indicate that a mechanism for the separation of organics by nanofiltration can be based solely on molecular size.

Studies with multicomponent solutions demonstrated that solute–solute interactions affect the performance of nanofiltration membranes. Rejection of a smaller solute (e.g., glucose) was improved by the presence of a large one (e.g., sucrose). These results showed that studies with real products and bleed streams are necessary for coupling nanofiltration to fermentation processes.

ACKNOWLEDGMENT

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