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COUPLING REVERSE OSMOSIS AND OSMOTIC DEHYDRATION: FURTHER INVESTIGATIONS

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ABSTRACT

The recently proposed enricher-stripper configuration for concentrating sucrose solutions from 5 to 60° Brix combining conventional reverse osmosis and osmotic dehydration is further analyzed. A graphical method is presented to estimate limits of operating pressure for the enricher and the stripper. Some area estimates are calculated with hydrophobic high-rejection reverse osmosis membranes in the enricher and high-rejection thin-film composite membranes in the stripper. An alternate configuration is also suggested for the interesting case in which a natural source of brine (seawater) is available.

INTRODUCTION

Concentration of sucrose solutions is a standard unit operation in the agro-alimentary industrial sector; for example, fruit juice is frequently concentrated. Typically, concentration includes an evaporation step, which due to the high latent heat of water is extremely energy intensive. Furthermore, a strict control must be maintained on the process temperature because some of the components (like

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flavor, aroma, etc.) in the solution being concentrated are heat sensitive. Though membranes have the operational advantage of dewatering (concentrating) at ambient temperatures, for the last 10 years, the application of membrane technology in such cases has been considered technology for tomorrow.

The earliest approach in concentrating sucrose solutions from typical feed concentrations of 5° Brix (approximately 5% wt/vol) to 60° Brix (concentrations of sucrose syrups) was made by applying the most basic membrane technology, reverse osmosis (1). This process soon becomes limited due to the extremely high osmotic pressure of the solution being concentrated and hence necessitates the use of high feed-side pressures of 70–100 bar (2,3). The use of such high- pressure feed pumps has the additional disadvantage of high installation and operating costs.

One way of overcoming the problem of osmotic pressure limitations is to use “loose” membranes in which the increase in permeate concentration compensates for the increase in feed osmotic pressure (4). However, an accompanying disadvantage is the loss of important components of the juice. To circumvent this problem, a fairly complex processing scheme must be adopted (5).

Osmotic dehydration is an alternative technique in which the difference in chemical potential of water in two different solutions separated by a membrane (osmotic pressure gradient) is used to dehydrate/concentrate one of the two solutions (6).

Therefore, an attractive proposition is the application of a combination of reverse osmosis and osmotic dehydration. With such a combination, higher concentrations of feed solutions could be achieved at moderate operating pressures that keep high enough transmembrane permeate fluxes if a salt solution (brine) is allowed to flow on the permeate side of the membrane in a countercurrent manner. Such a situation was recently analyzed by Karode, Kulkarni, and Ghorpade (7) in which the feasibility of such a technique was investigated with both theory and experiments. The researchers concluded that the process would be limited by permeate-side concentration polarization and that to reduce contamination of the sucrose solution being concentrated, a very high rejection reverse-osmosis membrane would be required.

Recent advances in membrane distillation in which hydrophobic microfiltration membranes were used (8) demonstrated the efficacy of using such membranes for permeates that have extremely high purity (9). The use of hydrophobic high-rejection membranes for osmotic dehydration would improve the process proposed by Karode, Kulkarni, and Ghorpade (7) in two ways: 1) increased transmembrane flux in the enricher section and 2) elimination of contamination of the sucrose solution being concentrated due to salt leakage from the permeate-side brine solution.

The purpose of this work was to demonstrate the advantages of using hydrophobic microfiltration membranes for concentration of 5–60% sucrose solutions. A simple method of estimating limits to the operating pressure in the enricher and the stripper sections is demonstrated. A scheme to utilize an available natural source of brine, such as seawater, for the above process is outlined.

THEORY

Figure 1 shows the enricher-stripper configuration proposed by Karode, Kulkarni, and Ghorpade (7). The labels of the configuration are analogous with the nomenclature in conventional distillation columns; the sucrose solution is "enriched" in the enricher (water flows from the sucrose solution into the salt solution in the permeate) and the stripper "strips" the dilute salt solution of water for recycling back into the enricher.

Solute and solvent balances on the enricher can be written as:

$$\frac{dQ_\mathrm{f}}{da^\mathrm{E}} = \frac{dQ_\mathrm{p}}{da^\mathrm{E}} = -J_\mathrm{w} \tag{1a}$$

$$\frac{d(Q_\mathrm{f}C_\mathrm{f})}{da^\mathrm{E}} = -J_\mathrm{w}C_\mathrm{f}(1 - R_\mathrm{f}) \tag{1b}$$

$$\frac{d(Q_\mathrm{p}C_\mathrm{p})}{da^\mathrm{E}} = -J_\mathrm{w}C_\mathrm{p}(1 - R_\mathrm{p}) \tag{1c}$$

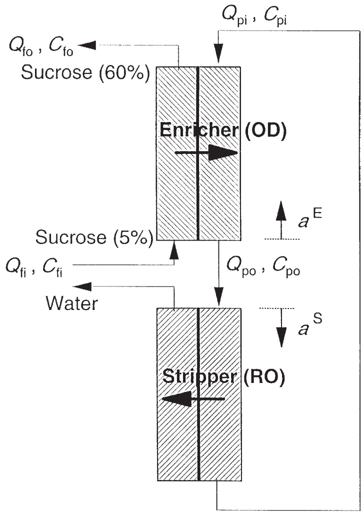

Figure 1. Schematic of the enricher-stripper configuration. OD = Osmotic Dehydration; RO = Reverse Osmosis.

where R_f and R_p are the rejection coefficients for sucrose and salt respectively. The water flux, J_w, is given by

$$J_w = A(\Delta P - \Delta\pi) \tag{2}$$

Here, A is the membrane permeability coefficient for water, ΔP is the transmembrane hydraulic pressure difference, and $\Delta\pi$ is the osmotic pressure difference. The osmotic pressure variation of sucrose and salt solutions as a function of concentration are given by (10):

$$\begin{aligned} \pi_{suc} &= 0.0154C^2 + 0.458C = \alpha_1 C + \alpha_2 C^2 \\ \pi_{sal} &= 0.179C^2 + 6.518C = \beta_1 C + \beta_2 C^2 \end{aligned} \tag{3}$$

where the concentration (C) is in a weight/volume percentage and the osmotic pressure (π) is in bar. α and β are constants.

Because the hydrophobic high-rejection membranes in the enricher would ideally reject both salt and sucrose (8), R_f and R_p in Eqs. (1b) and (1c) can be set to unity. Simplifying the resulting equations and dividing by Eq. (1a), the equation of continuity for the feed and permeate streams can be derived:

$$\begin{aligned} Q_f C_f &= Q_{fi} C_{fi} \\ Q_p C_p &= Q_{po} C_{po} \end{aligned} \tag{4}$$

By dividing Eq. (1b) by Eq. (1c) and integrating from $a^E = 0$ to any a^E, one obtains

$$\frac{C_{po}}{C_p} = \frac{(Q_{po} - Q_{fi})C_f + Q_{fi}C_{fi}}{Q_{po}C_f} \tag{5}$$

Equation (5) relates the sucrose concentration in the feed (C_f) to the salt concentration in the permeate (C_p) at any given area of the enricher (a^E).

By assuming ideal membrane rejections and incorporating the variation of osmotic pressure of salt and sucrose as a function of concentration (Eq. 3), one can rearrange Eq. (1b) to give

$$\frac{dC_f}{da^E} = \left(\frac{A^E}{Q_{fi}C_{fi}}\right) C_f^2[\Delta P^E - \alpha_1 C_f - \alpha_2 C_f^2 + \beta_1 C_p + \beta_2 C_p^2] \tag{6}$$

Eliminating C_p with Eq. (5), one gets

$$\frac{dC_f}{da^E} = \left(\frac{A^E}{Q_{fi}C_{fi}}\right) C_f^2 \left[\begin{aligned} &\Delta P^E - \alpha_1 C_f - \alpha_2 C_f^2 + \beta_1 \frac{Q_{po}C_{po}C_f}{(Q_{po} - Q_{fi})C_f + Q_{fi}C_{fi}} \\ &\qquad + \beta_2 \left(\frac{Q_{po}C_{po}C_f}{(Q_{po} - Q_{fi})C_f + Q_{fi}C_{fi}}\right)^2 \end{aligned} \right] \tag{7}$$

The right hand side of Eq. (7) is a function of C_{f} only and hence can be numerically integrated to calculate the area of the enricher:

$$a^{\mathrm{E}} = \left(\frac{Q_{\mathrm{fi}}C_{\mathrm{fi}}}{A^{\mathrm{E}}}\right)\int_{C_{\mathrm{fi}}}^{C_{\mathrm{fo}}} \frac{dC_{\mathrm{f}}}{f(C_{\mathrm{f}})} \tag{8}$$

where $f(C_{\mathrm{f}})$ is the function defined by the right hand side of Eq. (7).

For the special case of equal flow rates of the feed and permeate streams at the low-sucrose boundary of the enricher ($Q_{\mathrm{fi}} = Q_{\mathrm{po}}$), Eq. (7) can be simplified and can then be analytically integrated to give

$$\begin{aligned} a^{\mathrm{E}} &= \left(\frac{Q_{\mathrm{fi}}C_{\mathrm{fi}}}{A^{\mathrm{E}}}\right) \\ &\times \left[\begin{aligned} &\frac{x_2^{\mathrm{E}}}{2x_1^{\mathrm{E}^2}} \ln \frac{(x_1^{\mathrm{E}} + x_2^{\mathrm{E}}C_{\mathrm{fo}} + x_3^{\mathrm{E}}C_{\mathrm{fo}}^2)}{(x_1^{\mathrm{E}} + x_2^{\mathrm{E}}C_{\mathrm{fi}} + x_3^{\mathrm{E}}C_{\mathrm{fi}}^2)} \frac{C_{\mathrm{fi}}^2}{C_{\mathrm{fo}}^2} - \frac{1}{x_1^{\mathrm{E}}}\left(\frac{1}{C_{\mathrm{fo}}} - \frac{1}{C_{\mathrm{fi}}}\right) \\ &+ \left(\frac{x_2^{\mathrm{E}2}}{2x_1^{\mathrm{E}^2}} - \frac{x_3^{\mathrm{E}}}{x_1^{\mathrm{E}}}\right)\frac{2}{\sqrt{q^{\mathrm{E}}}}\left(\tan^{-1}\frac{2x_3^{\mathrm{E}}C_{\mathrm{fo}} + x_2^{\mathrm{E}}}{\sqrt{q^{\mathrm{E}}}} - \tan^{-1}\frac{2x_3^{\mathrm{E}}C_{\mathrm{fi}} + x_2^{\mathrm{E}}}{\sqrt{q^{\mathrm{E}}}}\right) \end{aligned}\right] \end{aligned} \tag{9}$$

where

$$\begin{aligned} x_1^{\mathrm{E}} &= \Delta P^{\mathrm{E}} \\ x_2^{\mathrm{E}} &= \beta_1 \frac{C_{\mathrm{po}}}{C_{\mathrm{fi}}} - \alpha_1 \\ x_3^{\mathrm{E}} &= \beta_2 \left(\frac{C_{\mathrm{po}}}{C_{\mathrm{fi}}}\right)^2 - \alpha^2 \\ q^{\mathrm{E}} &= 4x_1^{\mathrm{E}}x_3^{\mathrm{E}} - x_2^{\mathrm{E}^2} \end{aligned} \tag{10}$$

Equation (9) holds for the cases where $q^{\mathrm{E}} > 0$. For cases where $q^{\mathrm{E}} < 0$, to use Eq. (9) for calculating the area of the enricher, q^{E} should be replaced by $-q^{\mathrm{E}}$ and $\tan^{-1}$ by $\tanh^{-1}$ and the "+" in the second line by "$-$". This analytical solution is useful in checking the accuracy of the numerical integration routine.

For the stripper, the area can be calculated by the following expression (which can be derived in an analogous manner described above):

$$\begin{aligned} a^{\mathrm{S}} &= \left(\frac{Q_{\mathrm{po}}C_{\mathrm{po}}}{A^{\mathrm{S}}}\right) \\ &\times \left[\begin{aligned} &\frac{x_2^{\mathrm{S}}}{2x_1^{\mathrm{S}^2}} \ln \frac{(x_1^{\mathrm{S}} + x_2^{\mathrm{S}}C_{\mathrm{pi}} + x_3^{\mathrm{S}}C_{\mathrm{pi}}^2)}{(x_1^{\mathrm{S}} + x_2^{\mathrm{S}}C_{\mathrm{po}} + x_3^{\mathrm{S}}C_{\mathrm{po}}^2)} \frac{C_{\mathrm{po}}^2}{C_{\mathrm{pi}}^2} - \frac{1}{x_1^{\mathrm{S}}}\left(\frac{1}{C_{\mathrm{pi}}} - \frac{1}{C_{\mathrm{po}}}\right) \\ &- \left(\frac{x_2^{\mathrm{S}^2}}{2x_1^{\mathrm{S}^2}} - \frac{x_3^{\mathrm{S}}}{x_1^{\mathrm{S}}}\right)\frac{2}{\sqrt{-q^{\mathrm{S}}}}\left(\tanh^{-1}\frac{2x_3^{\mathrm{S}}C_{\mathrm{pi}} + x_2^{\mathrm{S}}}{\sqrt{-q^{\mathrm{S}}}} - \tanh^{-1}\frac{2x_3^{\mathrm{S}}C_{\mathrm{po}} + x_2^{\mathrm{S}}}{\sqrt{-q^{\mathrm{S}}}}\right) \end{aligned}\right] \end{aligned} \tag{11}$$

where

$$\begin{aligned} x_1^S &= \Delta P^S \\ x_2^S &= -\beta_1 \\ x_3^E &= -\beta_2 \\ q^S &= 4x_1^S x_3^S - x_2^{S^2} \end{aligned} \qquad (12)$$

In the present case, for the stripper, q^S is always less than 0.

DISCUSSION

As shown in Fig. 1, the salt stream flowing on the permeate side of the enricher results in a decrease in the effective osmotic pressure of the sucrose solution, which enables the concentration of the sucrose solution at low operating pressures. The osmotic pressure of a 60% sucrose solution is approximately 83 bar (Eq. 3). Hence, to operate the enricher at lower pressures, an appropriate concentration of the salt solution needs to be fed into the enricher on the permeate side. As the concentration of the salt stream is increased, the operating pressure of the enricher progressively reduces. However, at the same time, the stripper must be subjected to higher pressures to concentrate the lean (diluted) salt stream to be fed back into the enricher.

Figure 2 shows the salt concentration needed to be fed into the enricher (Y-axis) as a function of the pressure in the enricher (X-axis). The corresponding osmotic pressure of the salt stream is also plotted on the X-axis. For a specific operating pressure of the enricher (starting point on the X-axis), one can determine the corresponding salt concentration in the figure by moving vertically to the curve intersect that corresponds to the operating enricher pressure. From the point of intersection, travel horizontally until the curve corresponds to the osmotic pressure and read the operating pressure of the stripper. The two points on the X-axis, correspond, respectively, to the minimum operating pressures in the enricher and the stripper for the corresponding salt concentration needed to be fed into the enricher.

A naturally occurring source of salt solution (seawater) is a potentially cheap source of brine to feed into the enricher. Figure 2 shows that if seawater (3.5% salt) is used in the enricher, the minimum operating pressure of the enricher needs to be 59.4 bar (875.25 psi). For most centrifugal pumps, the maximum discharge pressure is limited to 54 bar (800 psi). This analysis indicates that to operate within the limit of 54 bar, a higher concentration of salt must be fed into the enricher. However, we will show that to use seawater, which could be discharged back into the sea, a slightly modified enricher-stripper configuration can be used.

For a specific operating pressure of enricher and stripper, the total area (a^E + a^S) depends on the salt concentration entering the enricher (C_{pi}) and the ratio of flow rates Q_{fi}/Q_{po}. As discussed earlier, variation in C_{pi} in one part leads to a de-

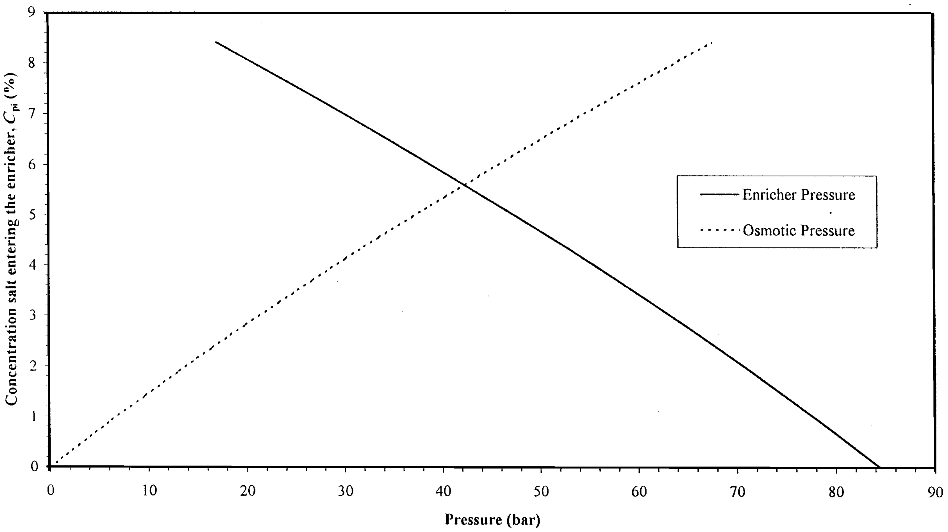

Figure 2. Salt stream concentration needed to be fed to the enricher as a function of enricher pressure.

crease in the enricher area while correspondingly requiring a larger stripper area and vice versa. The ratio of flow rates Q_{fi}/Q_{po} affects the extent of dilution of the salt stream entering the enricher. The dilution factor of the salt stream in the enricher, F, can be calculated as

$$F = \frac{Q_{po}/Q_{fi}}{Q_{po}/Q_{fi} - (1 - C_{fi}/C_{fo})} \tag{13}$$

Figure 3 shows the dilution factor of the salt stream in the enricher, F, as a function of Q_{fi}/Q_{po} for sucrose concentrations from 5 to 60%. As can be seen, for $Q_{fi}/Q_{po} > 3$, the dilution of the salt stream is not appreciably decreased. Hence, no significant advantage would be gained by reducing the enricher area for higher ratios of flow rate. Such practical limits to the ratio of feed and permeate flow rate at the low sucrose end of the enricher is useful in the actual hydrodynamic design of the membrane contactor. The limits also show that to design systems for $Q_{fi}/Q_{po} \approx 9$, as was done in the preliminary investigation (7), was unnecessary and impractical.

In the optimization of the total area ($a^E + a^S$), the ratio of flow rates Q_{fi}/Q_{po} was varied between 1 and 3. Figure 4 shows the variation of the total area as a function of the salt concentration entering the enricher, C_{pi} for various ratios of flow rate (Q_{fi}/Q_{po}). The value of membrane permeability used for the stripper is that typical of high-rejection, thin-film, composite, reverse-osmosis membranes

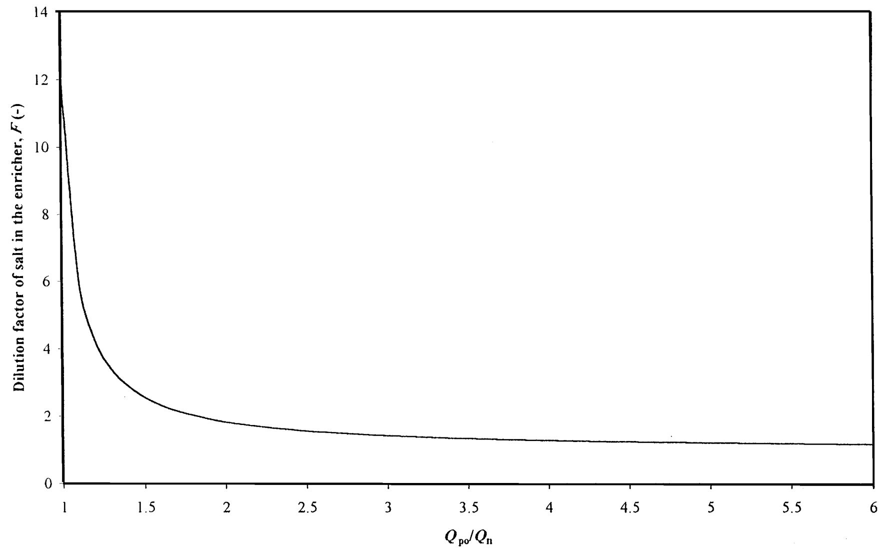

Figure 3. Variation of the dilution factor for salt in the enricher as a function of Q_{po}/Q_{fi}.

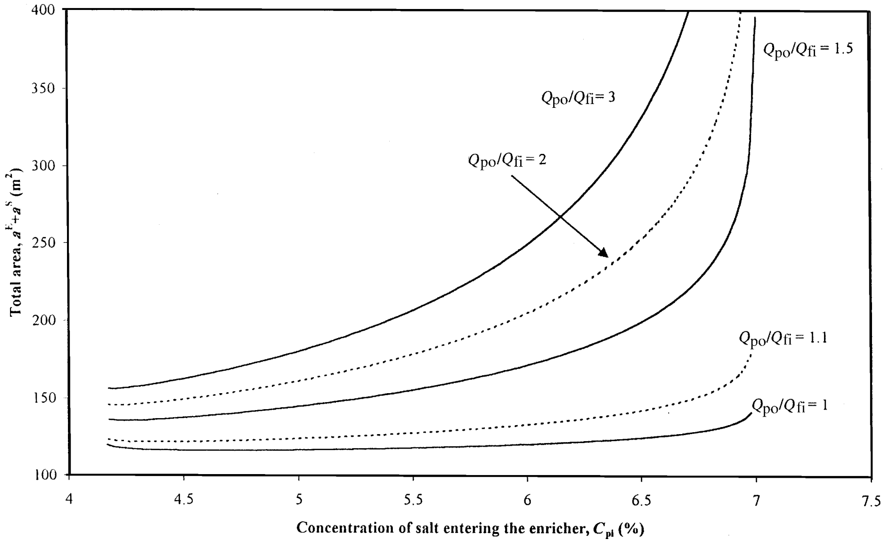

Figure 4. Variation of total area as a function of salt concentration entering the enricher for various values of Q_{po}/Q_{fi}. Other parameters include $Q_{fi} = 100$ lpm; $\Delta P^E = 54.4$ bar; $A^E = 9.1 \times 10^{-7}$ m^3/m^2·s·bar; $A^S = 4.1 \times 10^{-7}$ m^3/m^2·s·bar.

(7). For the enricher, the permeability value for hydrophobic polyvinyldifluoridine (PVDF) membranes, which are used for membrane distillation, were used (8). Figure 4 shows that the optimum concentration, C_{pi}, decreases as the ratio of the flow rates increases. The increase in flow-rate ratios leads to a reduction in the dilution factor for the salt. This, in effect, means that the stripper becomes limited due to the high osmotic pressure of the salt solution. Therefore, the optimum concentration is progressively lower as the ratio of flow rates increases.

Figure 5 shows the variation of enricher/stripper and the total area; both the enricher and stripper operate at 47.6 bar (700 psi) as a function of C_{pi} for $Q_{fi}/Q_{po} = 1$. The optimum concentration is 5.2%. The figure shows that for low operating pressures, that the stripper area controls the total area because the stripper becomes quickly limited due to the high osmotic pressure of the salt stream.

A further analysis made through the use of Fig. 2 shows that if the enricher is operated at 54.4 bar (approximately 800 psi), the minimum operating pressure in the stripper should be kept at approximately 30 bar. In such an operation, installation and operating costs for the enricher could be substantially reduced compared to the case discussed above (stripper also operating at 54.4 bar).

Figure 6 shows the area requirements for a configuration where $Q_{fi}/Q_{po} = 1$ at the optimum salt concentration at the enricher inlet. The operating pressure is indicated on the figure, and the other model parameters are the same as for Fig. 4.

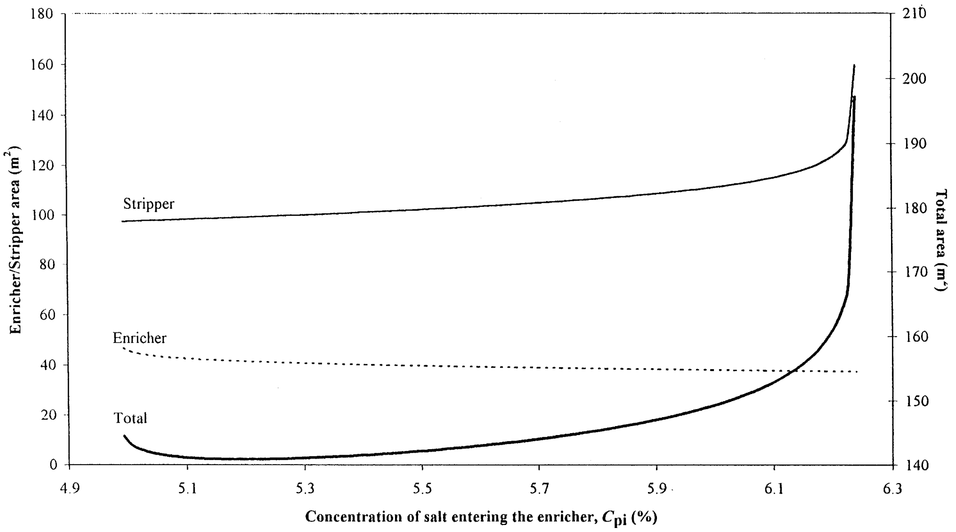

Figure 5. Variation of enricher, stripper, and total area as a function of salt concentration entering the enricher for $\Delta P^E = 47.6$ bar and $Q_{po}/Q_{fi} = 1$. Other parameters are the same as those in Fig. 4.

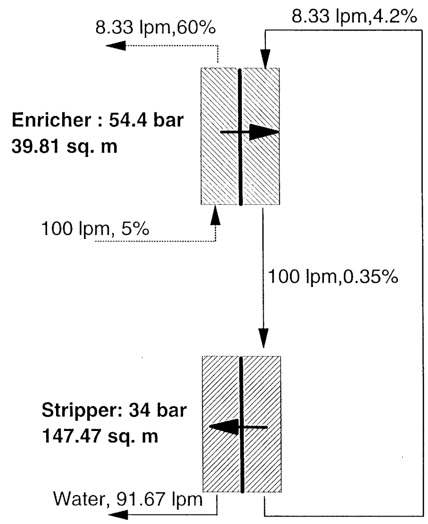

Figure 6. Area requirements for lower operating pressure of the stripper calculated using model parameters indicated in the text.

The total area was calculated as 187.3 m^2. Operating both the enricher and stripper at 54.4 bar resulted in a total area of 116.3 m^2. The increase in membrane area (by approximately 25%) is solely due to higher area requirements in the stripper. Keeping in mind the ever decreasing costs of reverse osmosis membranes and ever increasing power costs, one should operate the stripper at lower driving pressures for more favorable economic conditions.

Figure 7 shows a configuration for a natural source of brine (3.5% salt) utilized to concentrate the sucrose solution from 5 to 60% while the maximum operating pressure is kept at approximately 54.4 bar (800 psi). The final salt stream, to be discarded back into the sea, is only slightly diluted compared to what is drawn from the sea. This configuration of two enrichers and one stripper first concentrates the sucrose stream from 5.0 to 48.1% in the first enricher at 34 bar. The second enricher then further concentrates the sucrose solution from 48.1 to 60.0% at 54.4 bar. The above configuration requires approximately 235 m^2 of membrane area. This increased membrane cost would be offset by lower operating and installation costs of the pumps. Because the first enricher and the stripper both operate at lower pressures, they both require high flow-rate, low-pressure pumps that are much cheaper than high flow-rate, high-pressure pumps. The second enricher would require low flow-rate, high-pressure pumps that are also cheaper than high flow-rate, high-pressure pumps. This configuration is potentially interesting for cases in which a natural source of brine is available.

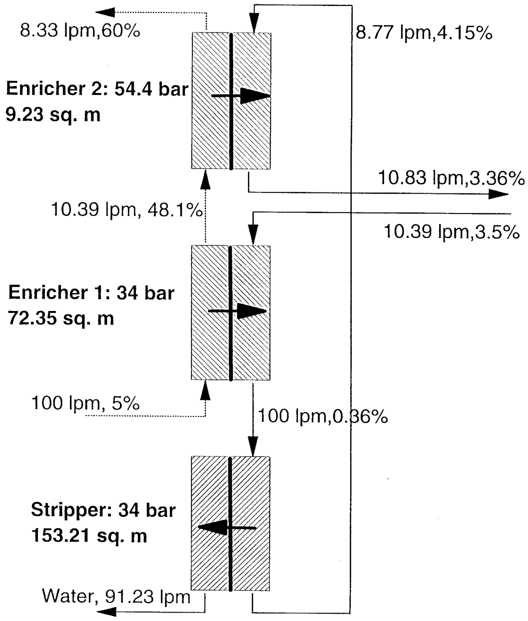

Figure 7. Area requirements for a modified enricher-stripper configuration for the case of a natural brine source (seawater).

CONCLUSIONS

The recently proposed, theoretically designed, enricher-stripper configuration for the concentration of sucrose solutions from 5 to 60° Brix combining conventional reverse osmosis and osmotic dehydration was further investigated with a hydrophobic high-rejection reverse-osmosis membrane in the enricher and a high-rejection, thin-film, composite, reverse-osmosis membrane in the stripper. A graphical method for estimating operating pressure limits in the enricher and the stripper is presented. The type of membrane used in the enricher, the operating pressure of the enricher and the stripper, the ratio of the salt-solution elution, and the flow rate of the sucrose solution into the enricher affect the optimum salt concentration that should be fed into the enricher to minimize the total membrane area. For a specific set of operating pressures, the optimum salt concentration reduces as the ratio of flow rates increases. A configuration is proposed that would enable the concentration of a sucrose solution from 5 to 60% using a natural source of brine (seawater, 3.5% salt) under a limited maximum operating pressure of 54.4 bar (800 psi). The proposed configuration may reduce operating and installation costs due to the management of flow rate and pressure requirements. The seawater can be eventually discharged back to the sea.

NOMENCLATURE

a	area (L^2)
A	water permeability coefficient ($M^{-1}L^{2}T$)
C	concentration (ML^{-3})
F	dilution factor ($-$)
J_w	water flux (LT)
ΔP	pressure ($ML^{-1}T^{-2}$)
Q	flow rate ($L^{2}T^{-1}$)
q	constant defined in Eqs. (10) & (12)
R	membrane rejection coefficient ($-$)
x	constant defined in text

Greek Letters

α_1, α_2	osmotic pressure coefficients for sucrose
β_1,β_2	osmotic pressure coefficients for salt
π	osmotic pressure

Subscripts

f	sucrose (feed)
i	inlet
o	outlet
p	salt (permeate)
sal	salt
suc	sucrose

Superscripts

E	Enricher
S	Stripper

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