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Effect of Fouling on Flux and on Energy Requirements in Reverse Osmosis of Skim Milk

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ABSTRACT

The effects of fouling on the permeate flux and on the power and energy required in the process of concentrating skim milk by a plate and frame type reverse osmosis unit with 990 type cellulose-acetate membranes were investigated. The permeate flow characteristics indicate that the fouling layer is rapidly formed and that its resistance is nearly constant over reasonable processing intervals. These fouling properties permit the correlation of permeate flow, overall mass transfer coefficient, and osmotic pressure difference in terms of the energy required per permeate volume versus time. This correlation shows a decrease in energy required per permeate volume with increasing mass transfer coefficient and time. These flow characteristics permit the estimation with confidence of the permeate flow energy requirements and the capacity of a full scale plant. Mass transfer coefficients were estimated for the membrane, the polarized layer, and the fouling layer. The overall mass transfer coefficient was then calculated. The polarized and fouling layers are films of components from the skim milk. Fouling was the controlling resistance.

INTRODUCTION

It has been reported that the concentration of dairy liquids by reverse osmosis leads to fouling of the membranes, which causes a decrease of flow with time (2, 7, 8). Because such fouling is an inherent part of the process, correlation of data for flow vs. time is needed to optimize equipment design.

This work was one phase of a project of the United States Department of Agriculture, partially funded by the Department of Energy, investigating low energy processes for concentration of fluid foods. Data were correlated in the solids concentration range of 8.8 to 25% by weight, because this is the range generally used for feed preparation (concentrated skim milk supplied to the evaporation step) and evaporation. Estimates of the cost and energy requirements for a commercial scale design are reported elsewhere (10).

MATERIALS AND METHODS

Experimental Procedures

A plate and frame module with a flat sheet membrane more convenient for the pilot plant operations of disassembly, cleaning, inspection, and replacement of membranes than spiral wound, tubular, and microfilament types. This type module is also being used in plant scale units and, therefore, poses no particular problem in scale up. The DDS (De Danske Sukkerfabrikker) Lab-Module size .72 m^2 (Figure 1) using 20 sandwiches of 990 type cellulose acetate membrane was used. The equipment is manufactured by De Danske Sukkerfabrikker,[3] DDS RO-Division of Denmark. Pasilac, Incorporated of Minneapolis, MN, is the United States supplier and agent. The feed circulating pump is a Rannie triplex high pressure piston homogenizer pump with a maximum capacity of 10 L/min at 8 MPa (1160 psig) operating pressure. A Reeves variable speed drive allows variations of the throughput of 2 to 10 L/min. A schematic diagram of the experimental flow arrangement is shown in Figure 2. A house cooling system, with cooling water at 5 to 8°C, controlled the skim milk at temperatures averaging from 15 to 21°C for the various runs.

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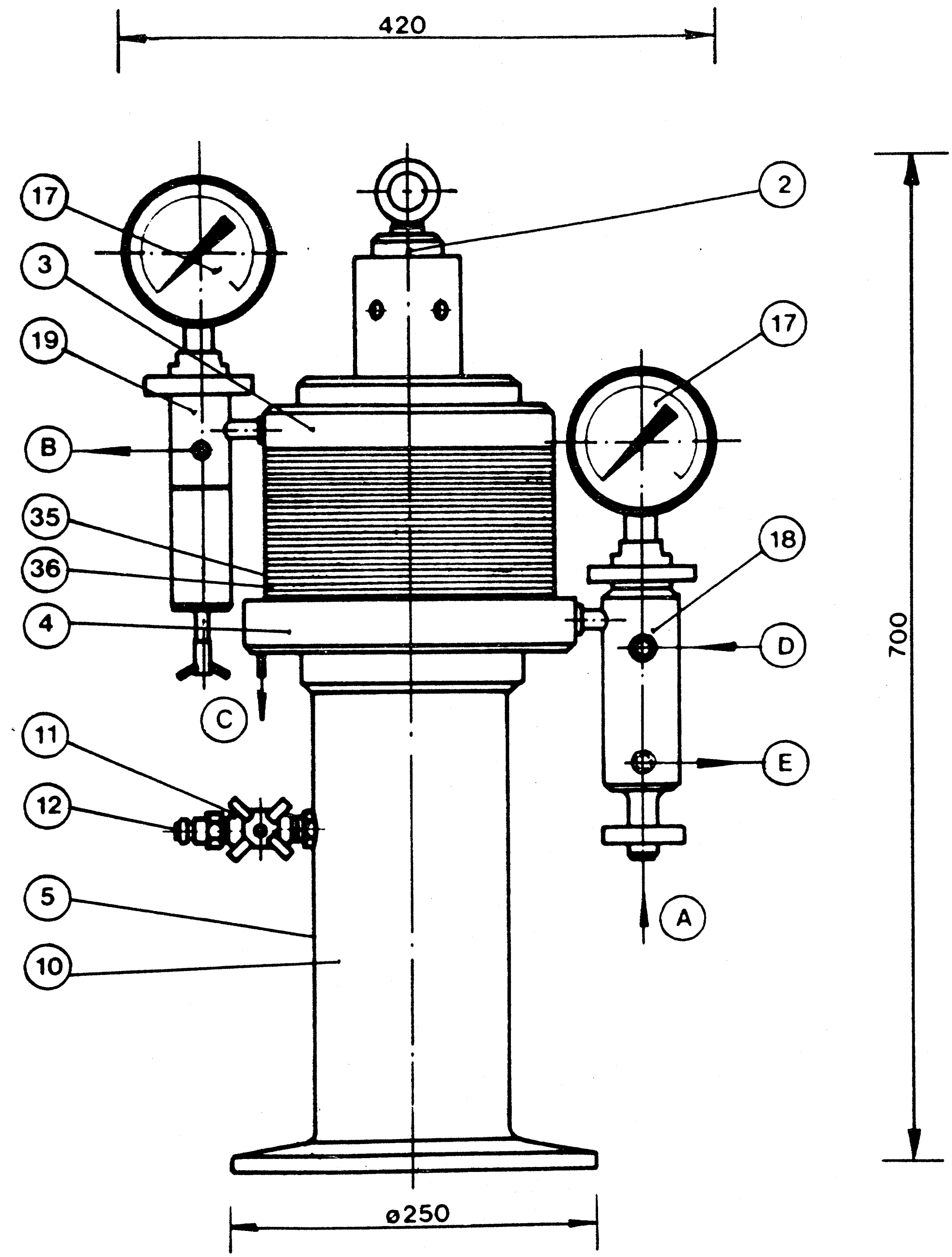

Figure 1. Reverse osmosis laboratory module. Legend: 2. center bolt, 3. top flange, 4. bottom flange, 5. base, 10. cylinder (built in), 11. valve, 12. coupling, 17. pressure gauge, 18. pressure gauge pocket with heat exchanger, 19. back pressure value. 35. membrane support disc, 36. intermediate disc, A. inlet, B. outlet, C. drain pipe, D. cooling/heating medium inlet, and E. cooling/heating medium outlet.

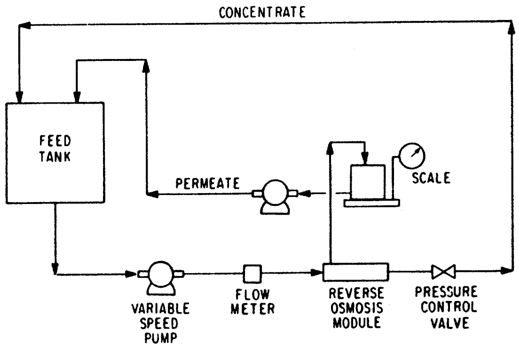

Figure 2. Flow arrangement for reverse osmosis equipment.

The temperature variation within each run was about ± 1°C. Conditions chosen for this study duplicate present commercial practice in concentrating skim milk from 8.8% total solids to 18% (factor of 2 ×) or in the extreme, 25% (2.8 × concentration). Also, it has been reported (10) that flux was unaffected by the volume processed when performing concentration.

Hiddink (2) has shown recirculation rate to be a critical variable; an optimum rate should be used to maximize overall flux over the entire operating period. The 8 L/min rate recommended by the manufacturer was used in all runs. It was observed that flux behaved as reported by Hiddink (2).

The Reynolds number cannot be easily determined in this type of equipment because of the difficulty in calculating the wetted wall diameter of the radial channel. The flow path of the liquids in the membrane chamber is shown in Figure 3. The 990 cellulose acetate membranes are being used in the industry. They give good flux and allow only small amounts of mineral salts of calcium, sodium, and potassium to pass into the permeate, not unlike entrainment in the condensate from thermal evaporation.

The pure water permeability constant was determined before each run and again after cleaning the membranes to determine the condition of the membranes at that time. It is well-documented that membrane permeability decreases with time or "age" of membrane due to compacting from the high pressures used in the operation. Further, a sodium chloride solution of 2000 ppm was run through the equipment before and after each trial to "define"

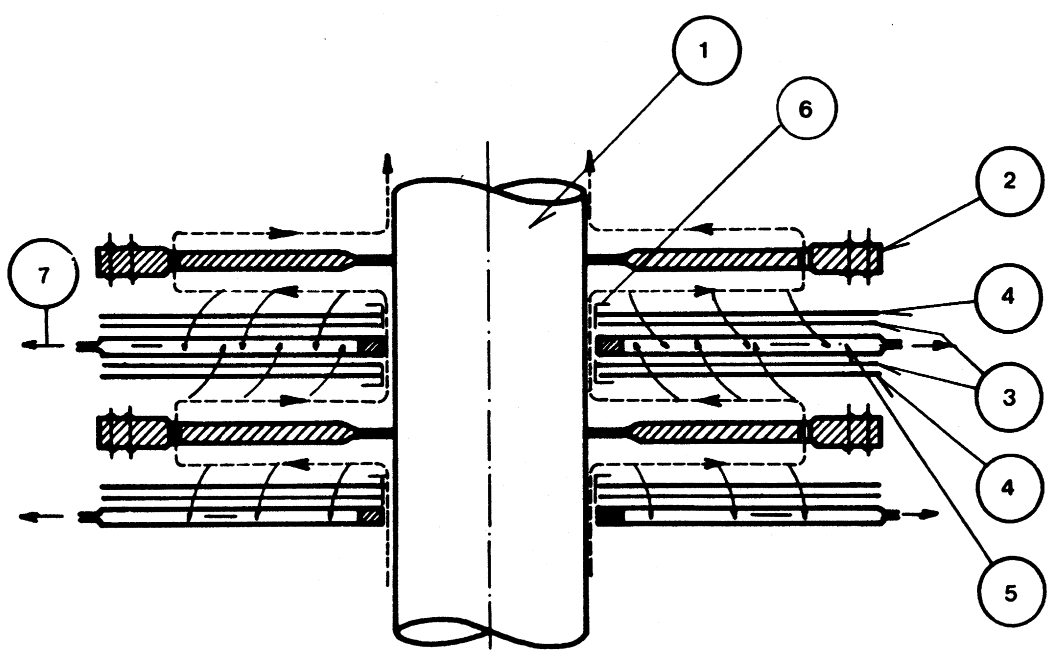

Figure 3. Flow path through module. 1. center bolt, 2. spacer, 3. drain paper, 4. membranes, 5. membrane support plate, 6. neck ring, and 7. permeate.

the "specific" membrane used and its condition (9). In this manner, unusual salt retentions would signal membrane deterioration as well as defining the membrane in Sourirajan's terms (4).

We determined flux values by weighing the amounts of permeate collected during 10, 15, or 30-min intervals for water, salt solution, and milk permeates, respectively. A series of runs were planned to evaluate the flux while operating at constant concentrations up to 35% solids to investigate the variations of flux with time at constant concentration. Concentrations of the skim milk and the electrical conductivities of the permeates were measured during the runs, and no significant variations were noted. Also, no changes in the visual appearances or odors of the fluids were noted. Therefore, we have no reason to believe that pH change or change in microorganism count is a factor in our experiments.

Fresh skim milk obtained from a local dairy outlet was used to prepare feeds at different concentrations. The milk was stored 4 days, at most, at a temperature of 3.3 to 5°C before being used in the experiments. Thirty-eight to 76 L of feed were prepared. Milk was concentrated in the reverse osmosis unit in a batch operation. Fat content of the skim milk was negligible, and no fat was noticed on the membranes upon visual inspection. This procedure differs from the way milk would be processed in a sanitary commercial plant, because it had a longer residence time in the equipment and it was exposed to air in the feed tank. However, we thought this was the best way to obtain the desired feed compositions without heat, as required in the concentration of the milk by evaporation. Therefore, we think no appreciable change in milk properties will occur and that the data and results are applicable to a full-scale plant.

RESULTS AND DISCUSSION

Correlation of Flux Versus Time

The correlation of flux in liters/(m^2/h) vs. time was derived analytically from the correlation of total volume of permeate collected vs. time. Time was .5 to 6.5 h. The data for total volume per square meter (L/m^2) vs. time are shown in Table 1. The figures in the body of the table are obtained by dividing the total volume of permeate collected up to that time by the membrane area of the reverse osmosis unit (.72 m^2). These data are closely correlated by a power curve:

$$y = ax^b \qquad [1]$$

y = cumulative volume of permeate collected per unit area of membrane, L/m^2, x = time, h.

Correlation coefficients (r^2) are .99 or greater for these curves. The first derivative of Equation [1] with respect to time will give the following relationship of permeate flux with time:

$$\text{flux} = dy/dx = abx^{b-1} \qquad [2]$$

Flux calculated from Equation [2] at .5-h intervals is listed in Table 2 for various feed compositions. For instance, Run No. A, which has a feed concentration of 8.59%, had a flux of 7.628 after 1.0 h. Plots of these curves are shown in Figure 4. Correlations of flux vs. time are of the same form as reported (2, 8) for fouling in reverse osmosis of whey.

Calculation of Fouling Mass Transfer Coefficient

The correlations of flux vs. time presented in Table 2 permit calculation of the fouling mass transfer coefficient in a manner similar to that shown by Hiddink (2). The general equation for flux is:

$$F = K_{mpf}(\Delta P - \Delta P_{ocf}) = ab\,t^{(b-1)} \qquad [3]$$

where:

F = flux, L/(m^2h),
ΔP = difference in pressure across membrane, atm,
ΔP_{ocf} = difference in osmotic pressure across membrane,
t = time in h,
a,b,b−1 = coefficients in Equation [2]
K_{mpf} = overall mass transfer coefficient, L/(m^2/h/atm),

Overall resistance to permeation comprises a polarized membrane resistance, $1/K_{mp}$, and a fouling resistance, $1/K_f$ such that:

TABLE 1. Total permeate volume, $V(L/m^2)$ collected over time, t. Data correlated by $V = at^b$.

	Feed concentration			
Time	8.59 Run A	11.48 Run B	15.69 Run C	34.12 Run D
(h)				
.5	4.85	4.28	3.33	2.54
1.0	9.13	8.03	6.72	4.81
1.5	12.31	11.53	9.76	7.01
2.0	16.09	14.87	12.78	9.19
2.5	19.70	18.87	15.69	11.36
3.0	23.10	23.36	18.57	13.54
3.5	26.37	26.36	21.42	15.71
4.0	29.51	29.27	24.24	17.87
4.5	32.55	32.12	27.16	20.00
5.0	35.57	34.89	29.91	22.12
5.5	38.37	37.59	32.74	24.24
6.0	41.18	40.25	35.57	26.33
a	8.92	8.16	6.58	4.82
b	.8547	.8962	.9436	.9467
Standard error of estimate[1]	.32	.64	.24	.11
Degrees of freedom	10	10	10	10
R-square[1]	.999	.999	.999	.999

[1]See (1).

$$R_o = 1/K_{mpf} = 1/K_{mp} + 1/K_f \qquad [4]$$

The polarized membrane resistance may be considered as consisting of a membrane hydraulic resistance, $1/K_m$, and a polarized layer resistance, $1/K_p$, such that:

$$1/K_{mp} = 1/K_p + 1/K_m \qquad [5]$$

and K_{mp} may then be calculated from:

$$K_{mp} = \frac{ab\ t^{b-1}}{\Delta P - \Delta P_{ocf}} \qquad [6]$$

TABLE 2. Flux $(L/(m^2/h))$ vs. time.

	Feed concentration			
Time	8.59% Run A	11.48% Run B	15.69% Run C	34.12% Run D
(h)				
.5	8.44	7.82	6.47	4.73
1.0	7.63	7.27	6.22	4.56
1.5	7.20	6.97	6.08	4.46
2.0	6.90	6.77	5.98	4.39
2.5	6.68	6.61	5.90	4.34
3.0	6.50	6.49	5.84	4.30
3.5	6.36	6.39	5.79	4.27
4.0	6.24	6.30	5.75	4.23
4.5	6.13	6.22	5.71	4.21
5.0	6.04	6.16	5.68	4.18
5.5	5.95	6.09	5.65	4.16
6.0	5.88	6.04	5.62	4.14

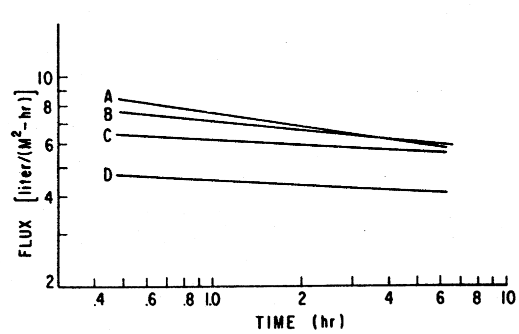

Figure 4. Correlation of flux $(L/(m^2/h)$ with time (h). Feed concentrations (weight %) A. 8.59, B. 11.48, C. 15.69, and D. 34.12.

under the conditions, at t = 1/60 h, when resistance consists of the membrane and polarized layer, since fouling is insignificaat this early in the process. Because $\Delta P_{ocf} = 0$, K_m may be evaluated from the pure water permeability data taken in the laboratory as:

$$K_m = F_{pw}/\Delta P \quad [7]$$

where F_{pw} = pure water flux. Therefore, the polarized layer resistance may be calculated as:

$$1/K_p = R_p = 1/K_{mp} - 1/K_m \quad [8]$$

At any time, flux is given by:

$$F_t = K_{mpft}\,(\Delta P_t - \Delta P_{ocf}) \quad [9]$$

So that K_{mpft} may be calculated.

$$K_{mpft} = \frac{F_t}{(\Delta P_t - \Delta P_{ocf})} = \frac{ab\,t^{b-1}}{\Delta P_t - \Delta P_{ocf}} \quad [10]$$

We chose t = 6 h. Therefore, K_f may be calculated from:

$$1/K_f = 1/K_{mpf} - 1/K_{mp} \quad [11]$$

Table 3 presents a summary of these calculations for the various feed compositions. It is obvious that fouling resistance is the controlling resistance. For instance, for Run B $(1/K_f) = 8.12$ and total resistance is $(1/K_{mpf}) = 8.35$. Therefore, fouling resistance is 97% of total resistance after 6 h of operation. The F_s, shown in Table 3, is the permeability of a sodium chloride solution (2000 ppm). The order of magnitude of the resistance for fouling appears consistent with those reported for skim milk by Hiddink (2).

Correlation of Power and Energy Required for Permeate Flow

Flow vs. time correlations were used to derive correlations of power required for permeate flow vs. time and also to derive correlations of energy required per volume of permeate vs. time.

From the previous section, permeate flux, $F[L/(h/m^2)]$ is given by:

TABLE 3. Mass transfer coefficients (K).

Run	Feed concentration	F L/(m²/h) t=1 min	F L/(m²/h) t=6 h	F_{pw}	F_s	P atm	P_o atm	K_{mp}	K_m	K_{mpf}	K_p	K_f
	(%)			— (L/(m²/h))—					—— (L/(m²/h/atm)) ——			
A	8.59	13.83	5.88	63.3	57.58	58.18	5.40	.2619	1.088	.1113	.3449	.1147
B	11.48	11.18	6.07	55.8	48.61	58.18	7.50	.2207	.9591	.1198	.2867	.1231
C	15.69	7.82	5.61	57.6	48.61	58.18	11.50	.1676	.9900	.1202	.2017	.1227
D	34.12	5.67	4.15	55.8	48.6	58.18	33.00	.2254	.9591	.1647	.2947	.1711

$$F = a\, b\, t^{b-1} \quad [12]$$

The power required for pumping the permeate only through the membrane and only at this point in the system may be derived by an energy balance for an incompressible fluid across the membrane from equations given in (5) and by:

$$B = (\Delta P)F/C_z \quad [13]$$

where C_z = 3.5526 × 10^4 (L/atm)/(kWh).

From (3), the pressure drop across the membrane, Δp (atm), required for a given flux, F, and osmotic pressure difference, ΔP_o, is:

$$\Delta P = F/K_{mpf} + \Delta P_o \quad [14]$$

where K_{mpf} = overall mass transfer coefficient in L/(h/m^2/atm) as in the previous section. Equation [14] assumes that the mass transfer coefficient, K_{mpf}, is constant.

Strictly speaking, this assumption for Equation [14] is not true. However, the flux vs. time curves show that flux decreases very slowly, indicating that the mass transfer coefficient decreases rapidly to almost a constant value. Similar observations are reported (6). Therefore, the error introduced by this assumption is not great, because the greatest error would occur in calculating the energy for the period when the coefficient decrease is greatest, say the first half hour, whereas the total period for the calculation is 6 h. Plant operating periods are about 20 h before the cleaning cycle. At this point, it would be appropriate to emphasize that ΔP, in Equations [13] and [14], is the pressure drop across the membrane for permeate flow, and that the energy requirement calculated from this ΔP is only for permeate flow immediately across the membrane. Therefore, for a unit area of the stage or module, the power may be calculated from the equation resulting from the combination of Equations [13] and [14]:

$$B = (F/K_{mpf} + \Delta P_o)F/C_z \quad [15]$$

where B = power, kW/m^2. The energy required may be calculated by integrating the power over time and dividing the latter energy by the integral of flow over time. Use of the integrals is required because flow and power vary with time. The energy required is given by:

$$E = \int_o^t B\, dt \quad [16]$$

where E = kWh/m^2. The volume of permeate is given by:

$$V = \int_o^t F\, dt \quad [17]$$

where V = L/m^2. The energy, E_V, required per liter of permeate is then calculated by dividing Equation [16] by Equation [17]:

$$E_V = \frac{\int_o^t B\, dt}{\int_o^t F\, dt} \quad [18]$$

where E_V = kWh/L. Permeate volume is easily obtained from Equations [12] and [17] as:

$$V = at^b \quad [19]$$

Power, B, is derived as a function of time from Equations [12] and [15]:

$$B = \frac{a^2\, b^2\, t^{(2b-2)}}{K_{mpf}C_z} + \frac{ab(\Delta P_o)t^{(b-1)}}{C_z} \quad [20]$$

Substituting B from Equation [20] into Equation [16] and integrating over the interval from o to t results in:

$$E = \frac{a^2b^2}{K_{mpf}C_z(2b-1)}\, t^{(2b-1)} + \frac{a\,(\Delta P_o)\, t^b}{C_z} \quad [21]$$

Combining Equations [21] and [19] to calculate energy per liter of permeate yields:

$$E_V = \frac{ab^2\, t^{b-1}}{K_{mpf}C_z\,(2b-1)} + \frac{\Delta P_o}{C_z} \quad [22]$$

Figure 5 shows the plot of Equation [22]. This equation correlates the flow of permeate, the mass transfer coefficient, the osmotic pressure difference, and time with the total energy required per volume of permeate. Referring to Figure 5, the curves are labeled with their corresponding concentration and overall mass

TABLE 4. Calculated energy (E, E_V), power (B), permeate volume (V), and flux (F).

Run	Feed concentration	a Constant	b Constant	K_{mpf} Constant	ΔP_O atm	t	E kWh/m²	E_V kWh/L	B kW/m²	V	F
	(%)					(h)		———— ($\times 10^{-3}$) ————		(L/m²)	(L/m²/h)
A	8.59	8.92	.8547	.1113	5.40	.1	4.24	3.40	30.3	1.25	10.7
						1.0	22.1	2.48	15.9	8.92	7.63
						2.0	36.3	2.25	13.1	16.1	6.90
						6.0	80.1	1.94	9.63	41.3	5.88
						10.0	115.8	1.81	8.36	63.9	5.46
						20.0	191.1	1.66	6.91	115.5	4.94
						100.0	613.0	1.34	4.45	457.0	3.91
B	11.48	8.16	.8962	.1198	7.50	.1	2.78	2.68	2.22	1.04	9.28
						1.0	3.07	2.15	14.1	8.16	7.31
						2.0	17.6	2.02	12.3	15.2	6.80
						6.0	74.2	1.82	9.94	40.7	6.07
						10.0	111.9	1.74	9.01	64.3	5.76
						20.0	195.5	1.64	7.89	119.6	5.36
						100.0	716.4	1.42	5.79	505.9	4.53
C	15.69	6.58	.9436	.1202	11.50	.1	1.56	2.08	14.0	.749	7.07
						1.0	12.3	1.87	11.0	6.58	6.21
						2.0	22.9	1.81	10.3	12.7	5.97
						6.0	61.4	1.72	9.19	35.7	5.61
						10.0	97.2	1.68	8.73	57.8	5.45
						20.0	181.1	1.63	8.14	111.1	5.24
						100.0	769.6	1.52	6.92	507.5	4.79
D	34.12	4.82	.9467	.1647	33.0	.1	1.02	1.86	9.34	.545	5.16
						1.0	8.46	1.76	7.80	4.82	4.56
						2.0	16.0	1.73	7.39	9.29	4.40
						6.0	44.2	1.68	6.79	26.3	4.15
						10.0	70.8	1.66	6.53	42.6	4.03
						20.0	134.2	1.63	6.20	82.2	3.89
						100.0	594.1	1.58	5.49	377.1	3.57

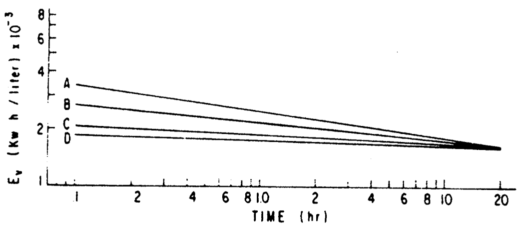

Figure 5. Correlation of energy per volume of permeate (kWh/L) with time (h). Feed concentrations (weight %) and mass transfer coefficient [L/(m^2/h/atm)]: A. 8.59, .1121; B. 11.48, .1255; C. 15.69, .1324; and D. 34.12, .1720.

transfer coefficient. In general, the curves demonstrate the trend we would expect from Equation [22]: that more energy is required as the overall mass transfer coefficient decreases. Examination of Figure 5 suggests that E_V, energy required per volume of permeate, presents a good correlation of flow conditions. The normal or intuitive expectation about energy requirement would be that as resistance increases, increased energy is required. However, one must take into consideration the conditions of the system under study. The pressure drop across the membrane is being held constant. Therefore, the flow through the membrane slowly decreases as the fouling resistance is slowly increasing. The energy requirement depends on flow and pressure drop. Thus, because pressure drop is constant and flow is decreasing, the energy requirement decreases with time. Therefore, the energy required per total permeate volume collected will decrease with time as shown by the calculations presented in the paper. These calculations use the experimentally determined permeate volume versus time data. Table 4 presents the data calculated by Equations [12] and [19 to 22]. Again, it would be appropriate to state that E_V represents only the energy requirement for the permeate flow across the membrane, and this energy is required to overcome the effects of membrane resistance, polarized layer resistance, and fouling layer resistance. Therefore, this energy requirement may be thought of as the minimum energy required for permeate flow due to reversible (membrane resistance) and irreversible (fouling) effects.

CONCLUSIONS

The effect of fouling on the permeate flow in the reverse osmosis processing of skim milk is similar to that reported previously (8). However, it is possible to correlate mathematically the permeate flow, the overall mass transfer coefficient, and the osmotic pressure difference in terms of the energy required per permeate volume, E_V, vs. time.

There appears to be an overall trend of decreasing energy requirement with increasing mass transfer coefficient. The flow characteristics indicate that the fouling layer is rapidly formed and that its resistance is practically constant over reasonable processing intervals. These characteristics permit the estimation of the energy requirements and design capacity of a large-scale plant with confidence. Such a design is reported (10).

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