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Reduction of the Lactose Content of Skim Milk by Continuous Countercurrent Cascade Ultrafiltration

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Abstract

Lactose was continuously removed from skim milk using two ultrafilters in series, with intermediate recycle. Reduced recycle flow rates at constant lactose stream (permeate product) flow rate resulted in slightly better removal of lactose from the milk feed, although this mode of operation increased the protein loss from the final milk retentate product. Increased permeate flow rate at constant recycle rate removed more lactose from the feed but also resulted in more loss of protein. However, at its maximum, the loss of protein represented only about 6% of the nutritional value of the milk. Under the experimental conditions studied, 58% of the lactose in the original skim milk could be continuously removed for the best combination of recycle and permeate flow rates. The experimental values of flow rates and compositions were reasonably well predicted by a mathematical model of the process.

INTRODUCTION

The treatment of protein calorie malnutrition with milk powder in developing countries has been a major source of controversy because of

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the high incidence of lactose malabsorption in these areas (*1*, *2*). The need for low lactose milk has been indicated by many nutritionists, since it is more easily absorbed by those with a lactose intolerance. In addition to peoples of developing countries, some infants and elderly in western countries exhibit intolerance to lactose and may also experience nutritional and calcium deficiencies (*3*).

Low lactose milk has been produced commercially by enzymatic hydrolysis of the lactose to glucose and galactose, and this product can be easily digested in liquid form by malabsorbers. However, in powder form it loses as much as 50% of its nutritional value because of so-called Maillard reactions which occur during production and storage of the material (*4*, *5*). Thus, hydrolyzed-milk powder has doubtful use as a protein source. Complete removal of the lactose by fermentation has been reported by Edelsten (*6*), but the resulting milk had an off-flavor.

Sorensen (*7*) reported on the production of a low lactose skim milk powder in which over 80% of the lactose is removed. The skim milk, originally containing about 10% solids, is batch ultrafiltered until the solids content reaches about 25%. There is a simultaneous reduction to about one-fifth of the starting volume. Maltodextrin is then added to compensate for loss of sweetness, and sodium and potassium salts replenish lost electrolytes. The mixture, now containing about 40% solids, is evaporated to 55% solids and then spray dried. The lactose content of the powder product was 86% lower than that of regular skim milk. Sorensen attributed a reduction of 71% to the ultrafiltration, with the remaining 15% occurring from the dilution effect of adding maltodextrin.

The present paper reports on attempts to continuously produce low-lactose milk by countercurrent cascade ultrafiltration with interstage recycle. The milk salts are continuously replenished by a make-up solution, and the effects of recycle and lactose stream removal rates are examined. Theoretical predictions of flow rates and compositions were made using the model of Ward et al. (*8*), and these are compared to experimental values.

THEORY

Ward et al. (*8*) developed a theoretical model to predict the concentration of different sized particles in each stream of a system of N ultrafilters in series. First, a sieving coefficient, S, which is a measure of the inverse of membrane retention is defined for each species for each membrane as

$$S = C_p \cdot 2/[C_f + C_r] \quad (1)$$

where C_p = concentration of species in permeate
C_f = concentration of species in feed
C_r = concentration of species in retentate

This simple relationship ignores concentration polarization, which is the undesired build-up of dissolved species at the membrane surface. To simplify the mathematics of the model, an average sieving coefficient for each species for all ultrafilters is calculated and considered as the overall sieving coefficient of the species in the system. (While we used "identical" membrane modules, no two modules are truly the same, and each one exhibits slightly different retention characteristics for each species.)

Figure 1 shows, schematically, the system of N ultrafilters in series. Volume and mass balances around each filter and node (where the recycle and feed streams mix) are used to arrive at a system of equations which may be solved by matrix techniques for the solute concentrations in the various streams (*8*).

The degree of separation which can be obtained with any particular separation process is indicated by the separation factor. One definition of separation factor is

$$\alpha^s_{XY} = (X_1/Y_1)/(X_2/Y_2) \quad (2)$$

where X_1 = mole fraction of Component X in Product 1
Y_1 = mole fraction of Component Y in Product 1
X_2 = mole fraction of Component X in Product 2
Y_2 = mole fraction of Component Y in Product 2

The numerical value of separation factor defined in this manner remains unchanged if mole fractions are replaced by mass fractions or molar or mass flow rates of individual components. If the separation factor is unity, no separation of Component X from Component Y has occurred. If the separation factor is greater than 1, Component X tends to concentrate in Product 1 while Component Y concentrates in Product 2. The opposite is true for a separation factor less than unity. In the present process the separation of lactose from the minor proteins, α-lactalbumin and β-lactoglobulin, is characterized by a separation factor since the membranes used totally retained the major protein, casein, while the minor proteins were only partially retained. All of the lactose permeated through the membranes.

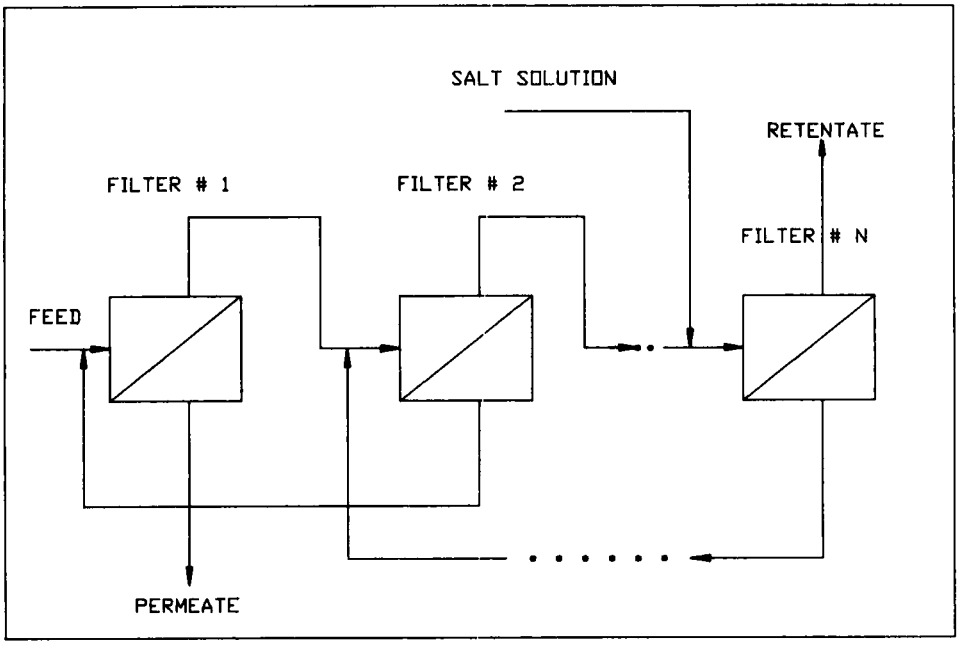

FIG. 1. System of N ultrafilters with intermediate recycle.

MATERIALS AND METHODS

Experimental System

The experimental system, shown schematically in Fig. 2, consisted of two Hemoflow F60 (Fresenius), polysulfone, hollow-fiber ultrafilters in series. Pumping was by Cole-Palmer Masterflex peristaltic pumps through Tygon tubing. Each filter contained 1.25 m^2 membrane surface area and the fiber internal diameter was 0.02 cm. The fresh skim milk was mixed with the recycled permeate stream from the second filter and then fed to the first filter. The permeate from the first filter was removed as the concentrated lactose solution which also contained dissolved milk salts. The retentate from the first filter was mixed with the make-up salt solution and fed to the second filter. The permeate from this filter was recycled to mix with the feed milk, while the retentate was the reduced-lactose milk product. Subsequent filters could be added to further enhance the lactose removal process.

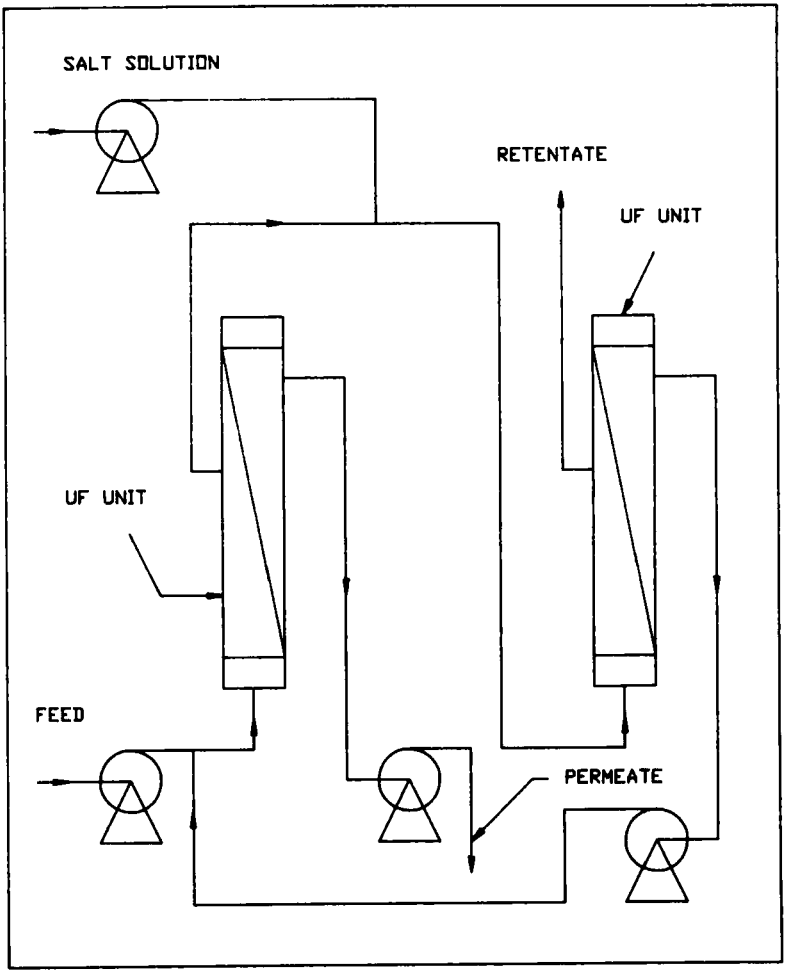

FIG. 2. Schematic of experimental apparatus.

Procedure

Initially the sieving coefficients for lactose, casein, and the minor proteins α-lactalbumin and β-lactoglobulin were determined for each filter. Though the two filters were "identical," there are no truly identical ultrafilters. For mathematical convenience an average sieving coefficient was calculated for each component. Skim milk was fed to the two ultrafilters in series and the retentate removal rate from the second filter was adjusted to this same value. Similarly, the salt solution feed rate was set equal to the permeate withdrawal rate from the first filter. Because of limitations in the pumping system, the maximum feed rate possible was 12.5 mL/min, and this was used in all runs. Thus the retentate or low-lactose milk rate was also 12.5 mL/min. The intermediate recycle rate was set at 3, 7, and 12.5 mL/min in successive runs. For these runs the permeate withdrawal rate from the first filter was set at 6 mL/min, as was the make-up salt solution feed rate. A second set of runs kept the recycle constant at 12.5 mL/min while the permeate was removed at 3.1, 6.1, and 12.5 mL/min. In each run, at least 1 h was allowed for steady state to be achieved and then approximately 200 mL each of the feed, permeate, and retentate were collected for analysis of lactose and protein concentrations.

Analysis

Since lactose is optically active, its concentration was determined by polarimetry using a Perkin-Elmer Model 241 MC polarimeter set with a sodium lamp (*10*). The proteins were determined by passage of samples through a Sephadex-100 gel column, fractionation of the eluent by fraction collector, and analysis of each fraction by a Perkin-Elmer Model 330 spectrophotometer.

Materials

To assure reasonable consistency of feed material, "Weight Watchers" brand skim milk was used in all runs. The make-up salt solution composition approximated the salt content of milk whey as found in the literature (*9*). Its composition is shown in Table 1. Jenness (*9*) also indicates that skim milk contains about 3.3% protein, about 80% of which is casein. The remainder consists of a mixture of globulins and albumins, of which β-lactoglobulin and α-lactalbumin are representative.

TABLE 1
Approximate Salt Content of Whey[a] (9)

Compound	% Concentration in water
Calcium chloride	0.1109
Sodium phosphate dibasic	0.1846
Potassium chloride	0.0671
Potassium sulfate	0.0181
Potassium bicarbonate	0.0328
Sodium bicarbonate	Enough to raise pH from 3.8 to 6.8 (pH of milk)

[a]Since the cited reference lists composition only by constituent ions, the above list of compounds and compositions were proposed as giving the closest total salt composition. Other combinations are also possible.

RESULTS AND DISCUSSION

The results of the study are presented and discussed in three separate sections. First, the measurement of sieving coefficients for each component is presented. Then the effect of changing recycle flow rate or recycle ratio, R, defined as recycle rate divided by fresh feed rate, is examined. Finally, the effect of varying the permeate flow rate, analogous to varying the make-up solvent flow rate, is discussed.

Determination of Sieving Coefficients

Initially the sieving coefficients for each of the components lactose, casein, and minor proteins were evaluated. Because the proteins α-lactalbumin and β-lactoglobulin could not be adequately separated on the Sephadex-100 gel, they were treated as a single entity and one sieving coefficient was calculated for the combined material. This was considered reasonable since they are close in molecular weight relative to the major protein, casein, which comprises about 80% of the total protein in milk. Casein has an average molecular weight of about 67,000, and it exists in milk as very large particles complexed with calcium and phosphate. Lactoglobulin exists as a dimer of average molecular weight 35,000, and lactalbumin has an average molecular weight of about 16,000. The Hemoflow F60 membranes were expected to retain most of the casein, pass all of the lactose and salts, and fractionate the minor proteins. This proved to be the case since the average sieving coefficient

for casein was calculated to be 0.0, while that of lactose was unity. The minor proteins had sieving coefficients of 0.47 in the first filter and 0.33 in the second, giving an average coefficient of 0.4 for the two filter system.

Effect of Recycle Flow Rate/Ratio

With the feed set at a constant 12.5 mL/min and the permeate at 6 mL/min, the recycle rate was changed from 3 to 6 to 12.5 mL/min. This corresponded to recycle ratio values of 0.24, 0.48, and 1.0. Figures 3 and 4 show the effect of recycle on lactose and minor protein recoveries, respectively, in the two exiting streams. Figure 3 shows that the lactose recovery in the permeate stream, the stream we wish to concentrate in lactose, decreased slightly, from 41 to 37% as the recycle ratio increased from 0.24 to 1.0. Figure 4 shows that for all three recycle ratios the recovery of minor proteins in the retentate milk product was 75% or higher. This was a hoped-for result since we desired not to deplete the

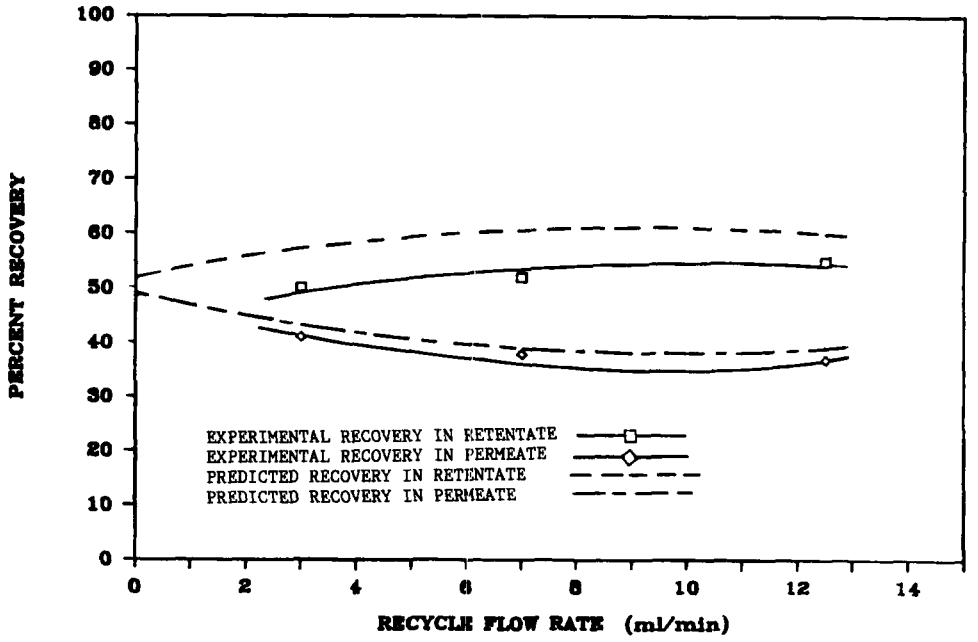

FIG. 3. Dependence of lactose recovery on recycle flow rate.

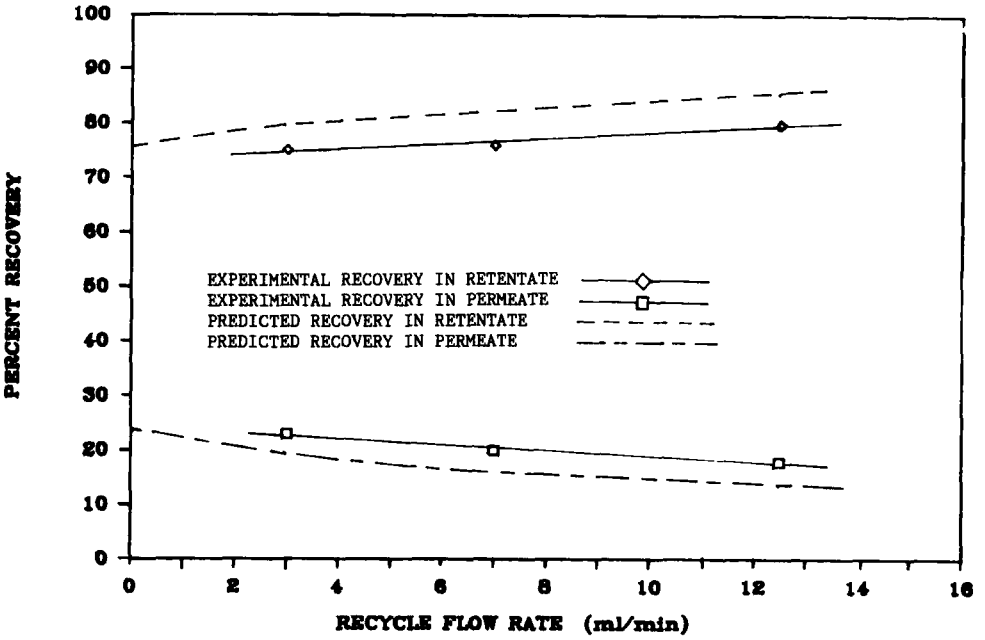

FIG. 4. Dependence of recovery of minor proteins on recycle flow rate.

milk of much of its protein nutritional value in the process of removing the lactose. In all cases the predictions of the system model closely followed the trend of the data. The slight differences between the predicted and actual curves can be attributed to the use of an average sieving coefficient for the two ultrafilters and to ignoring concentration polarization at the membrane surface.

Figure 5 shows the variation in separation factor between lactose and the minor proteins as a function of recycle rate. The experimental separation factor increased slightly from about 2.75 to 3.2 as the recycle flow rate was increased. The model predicted somewhat higher values, again due to the failure of the model to account for concentration polarization. Though the change in separation factor was not substantial as the recycle flow rate was increased, nonetheless it appeared that there was a slight gain to be made by running the system at a higher recycle ratio. Ward (*8*) predicts that a recycle ratio of unity gives the optimum trade-off between extraction of solute and separation factor.

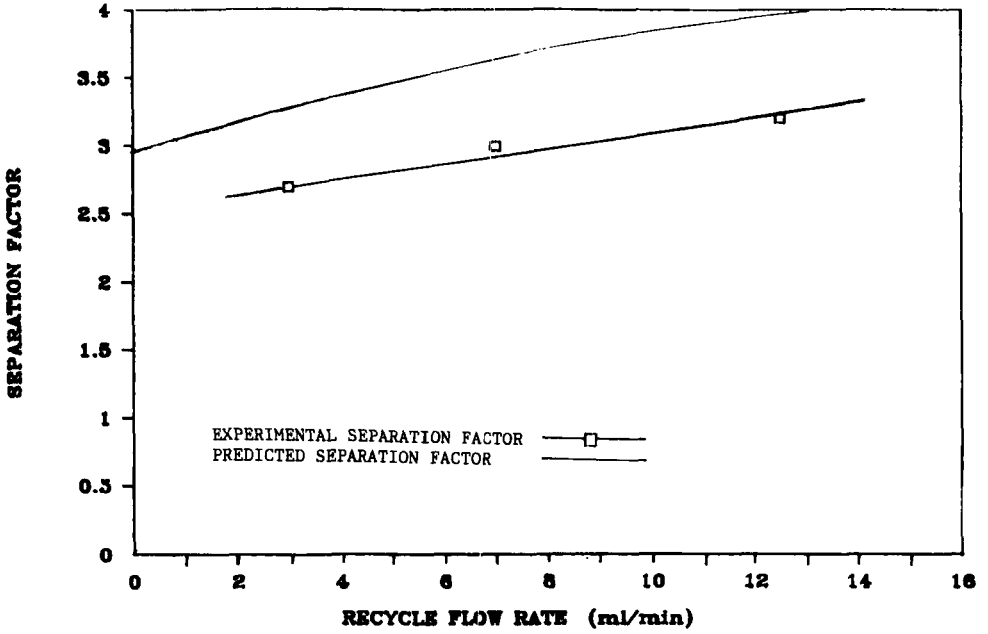

FIG. 5. Dependence of separation factor on recycle flow rate.

Effect of Permeate Flow Rate

Following the results of the recycle rate study, the feed and recycle rates were set at 12.5 mL/min (recycle ratio of 1.0), and the permeate flow rate was varied from 3.1 to 6.1 to 12.5 mL/min. In so doing, the make-up salt solution flow rate was also set to the same values as the permeate rate to facilitate the application of the mathematical model. Figures 6 and 7 show the effect of permeate flow rate on lactose and minor protein recovery, respectively, in each of the product streams. Figure 6 shows that the lactose recovery in the permeate increased dramatically from 21 to 58% as the permeate rate rose from 3.1 to 12.5 mL/min. However, Figure 7 indicates that for the same range of permeate flow rates the amount of minor proteins lost in the lactose-rich permeate stream also increased from 10 to 38%. This is undesirable, but its effect is outweighed by the increase in lactose removal from the milk, which is the main purpose of the process.

Even though the recovery of minor proteins in the permeate increased as the permeate flow rate increased, Fig. 8 shows that the separation

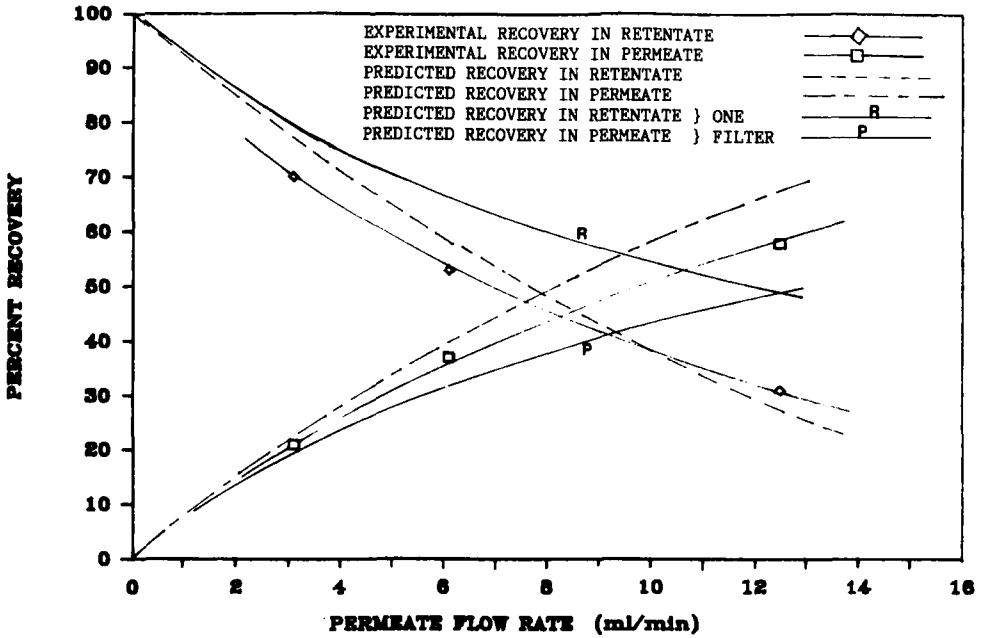

FIG. 6. Dependence of lactose recovery on permeate flow rate.

factor of lactose from proteins also increased from about 2.7 to 3 as permeate rate increased from 3.1 to 12.5 mL/min. That is to say that, though more protein was lost at the higher permeate rate, it was relatively less than was lost at lower flow rates. In addition, the loss of 38% of the minor proteins in the permeate represents a loss of only about 6% of the total protein nutritional value of the skim milk.

The mathematical model predicts the concentrations of proteins and lactose in the product streams quite well, though it does not do as well at predicting the separation factor. As mentioned earlier, this is probably due to a combination of using a single sieving coefficient to characterize the system, ignoring concentration polarization and perhaps also to assuming a linear concentration profile along the length of the hollow fibers. Figures 6 through 8 also indicate that the system of two filters in series gives higher recoveries of lactose and proteins in the permeate, as well as a higher separation factor of these components from each other, than does a single ultrafilter. In the case of lactose this represents an improvement, while for the proteins it is a disimprovement, insofar as more protein is lost in the dual system than in the single ultrafilter. For

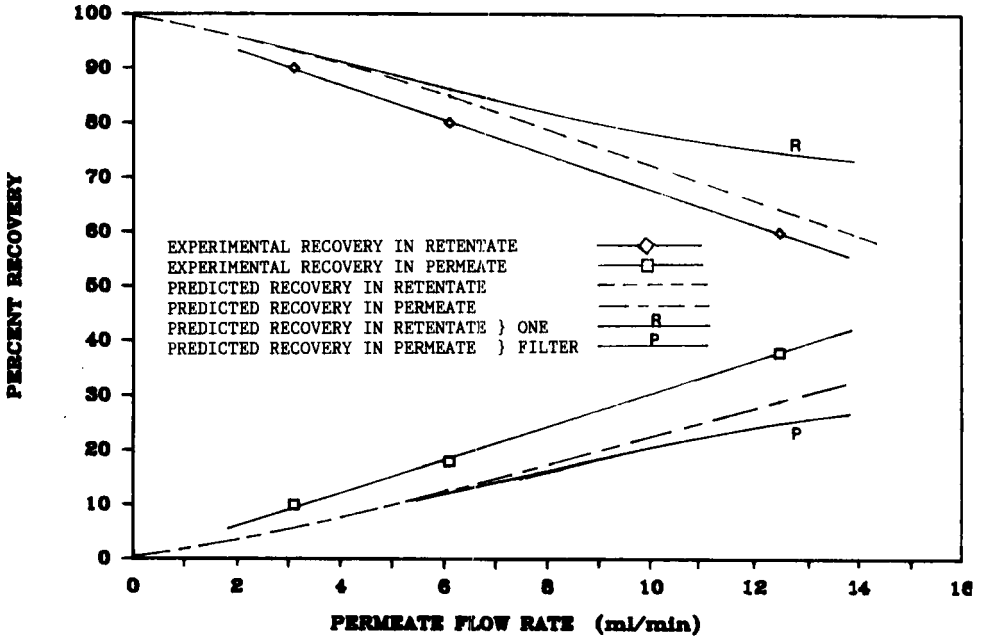

FIG. 7. Dependence of recovery of minor proteins on permeate flow rate.

the reasons cited above, however, this loss of protein is far outweighed by the increased removal of lactose from the milk. Based on the current results, and on the model predictions of Ward et al. (*8*), it is also predicted that the addition of a third filter in series will further increase the lactose removal from the milk, though at the expense of some more loss of minor proteins.

CONCLUSIONS

A system of two ultrafilters in series was used to remove much of the lactose from skim milk. The study showed that the permeate (lactose-rich stream) flow rate had a major effect on the total lactose removal from the feed milk. Lactose recoveries in the permeate of as high as 58% could be achieved when the permeate flow rate was identical to the feed rate, at a recycle ratio (recycle flow rate to feed rate) of unity. This may be compared to the 71% recovery rate quoted by Sorensen (*7*) as being due to ultrafiltration alone in a single batch ultrafilter with recycle of retentate.

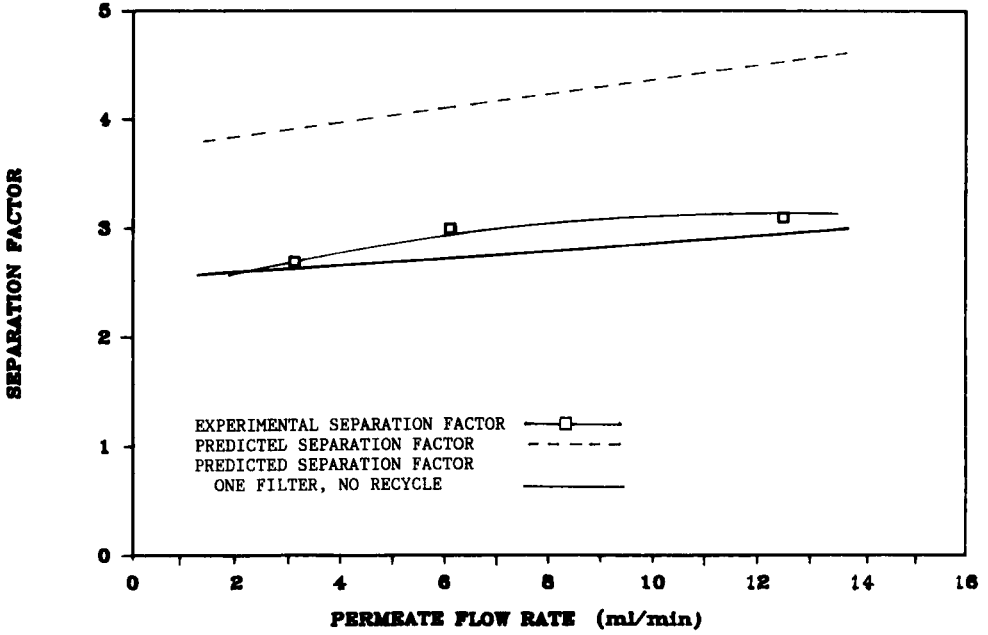

FIG. 8. Dependence of separation factor on permeate flow rate.

We feel that the addition of a third filter to our system will further increase the lactose removal and bring our system closer to the recovery experienced by Sorensen. The milk powder made from Sorensen's low-lactose milk was found to be acceptable to lactose maladsorbers and hence our product should also be satisfactory.

One minor disadvantage to our process is that in removing the lactose, the process also removed about 6% of the protein nutritional value of the milk in the form of the minor proteins, α-lactalbumin and β-lactoglobulin. However, we feel the removal of the lactose which was achieved far outweighs this slight loss of protein, which is certainly much lower than the 50% nutritional loss quoted by Burvall (*4*, *5*) as being lost by denaturation of hydrolyzed, low-lactose milk. One may also assume that some protein was lost in the permeate in Sorensen's experiments (*7*), although these data are not presented in his paper. In addition, the lactose removal in the present process is much superior to that which can be achieved with a single pass through one ultrafilter, while the incremental cost of the extra filter and pumps is minimal.

REFERENCES

1. F. J. Simoons, J. D. Johnson, and N. Kretchmer, *Pediatrics, 59,* 98–107 (1977).
2. F. J. Simoons, *Digest. Dis., 23,* 963–980 (1978).
3. L. Nagy, Gy. Mozsik, M. Garamszegi, E. Sasreti, Cs. Ruzsa, and T. Javor, *Acta Med. Hung., 40*(4), 239–245 (1983).
4. A. Burvall, N.-G. Asp, A. Dahlqvist, and R. Oste, *J. Dairy Res., 44,* 549–553 (1977).
5. A. Burvall, N.-G. Asp, A. Bosson, C. San Jose, and A. Dahlqvist, *Ibid., 45,* 381–389 (1978).
6. D. Edelsten, F. Ebbesen, and J. Hertel, *Milchwissenschaft, 34,* 733–734 (1979).
7. K. Sorensen, M. Meersohn, J. Sonne, L. Larsen, D. Edelsten, and E. Gudmand-Hoyer, *Scand. J. Gastroenterol., 18*(8), 1063–1068 (1983).
8. R. Ward, E. Klein, P. Feldhof, and T. Turnham, Paper presented at Conference on Chemical Processes, Cincinnati, Ohio, September 1984.
9. R. Jenness, *Principles of Dairy Chemistry,* Wiley, New York, 1976.
10. *Official Methods of Analysis,* Association of Official Agricultural Chemists, Washington, D.C., 1975.

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