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Minimizing Water Use in Diafiltration of Whey Protein Concentrates

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ABSTRACT

A multistage countercurrent diafiltration process for whey protein concentrates (WPC) production is presented. Experimental tests of whey diafiltration have validated a mathematical model for the concentration of components in the permeate for the case of washing with a dilute solution containing whey components. This model has been used together with material balances in order to simulate the multistage process. Water saving with respect to a single-stage conventional diafiltration was demonstrated. In the case of a WPC production of 95.5% from the whey of a local dairy farm which contained 7.7 g/L proteins, 43 g/L lactose, 1.1 g/L nonproteic nitrogen, and 5.4 g/L ashes, the necessary volumes of water, V_D (volume of water per volume of treated solution), needed decreased from 5.4 for a single stage to 2.9 for a two-stage process down to 1.5 for a six-stage process. The adopted procedure can be easily applied to any diafiltration process.

INTRODUCTION

Whey proteins are of higher nutritive value than many other animal proteins. This makes whey an attractive raw material in the production of whey protein concentrates (WPC) (1). These ingredients have been used extensively

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in various segments of the food industry including dairy, bakery, meat industry, confectionery, beverage production, and the manufacture of baby and dietary foods (2). Because of progress in membrane technology, it is now possible to obtain WPC containing the desired quantities of soluble native proteins, lactose, and mineral matter (1). To obtain WPC with 65% or more proteins on a dry basis in the solid content, water is normally added during ultrafiltration to create a *diafiltration* operation whose main purpose is to remove large quantities of lactose from whey. The WPC thus obtained contains lower concentrations of lactose and mineral matter, which broadens their use in food product applications (1–3).

The diafiltration operation is characterized by high water consumption (4). It is well documented in the literature (5–8) that a remarkable water saving is achieved in a pre-ultrafiltrative concentration step in which the product concentration in the retentate is raised to the highest level consistent with an acceptable ultrafiltration flux. Nevertheless, water saving can be further improved by carrying out diafiltration in more than one stage. The aim of this work is to study a multistage countercurrent diafiltration process in the production of whey protein concentrates in which water consumption is lower than in conventional continuous diafiltration.

MATERIALS AND METHODS

Diafiltration Tests

The diafiltration tests were carried out in a laboratory experimental apparatus whose flow sheet is shown as Fig. 1. The solution to be treated [250 mL:

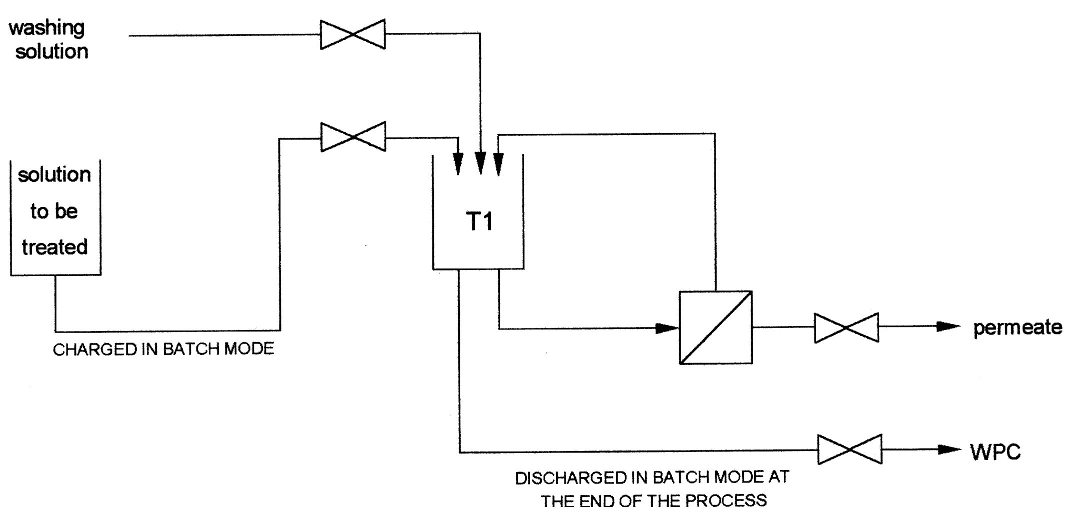

FIG. 1 Experimental apparatus employed for diafiltration tests.

TABLE 1
Whey Composition

pH	4.69
Proteins	7.7 g/L
Lactose	43 g/L
NPN	1.1 g/L
Total solids	5.72%
Ashes	0.54%

either 20 g/L lactose or whey (Table 1) according to experimental conditions] was placed in a thermostated beaker at 25°C in batch conditions. Then the continuous diafiltration process was started using either distilled water or a lactose solution of 3 g/L as the washing system. During diafiltration the washing solution was continuously added to the retentate in the feed beaker at a rate equal to the permeation rate. A laboratory scale plate and frame UF unit was used (Millipore, MINITAN-S.) equipped with a polysulfone membrane, 10,000 Da MWCO, total area 36 cm^2. The unit operated at a transmembrane pressure of 200 kPa, and the tangenital velocity was 0.3 m/s. Aliquot amounts (1.5 mL) of permeate were periodically sampled for lactose determination.

Analytical Determinations

Lactose concentration was determined by the lactose/galactose UV method, Boehringer Mannheim. Protein concentration was determined by Kjeldahl's method for proteic nitrogen determination (heating digester VELP SCIENTIFICA mod. DK6; automatic steam distilling unit VELP SCIENTIFICA mod. UDK 130; automatic titrator CRISON mod. micro TT 2050).

GROUNDS OF THE PROPOSED CALCULATIONS

The system employed in the experimental tests was modeled as a CSTR. In fact, some experiments performed with a tracer (whose retention coefficient through the membrane was 0) evidenced perfect agreement between the experimental response to a step variation and the one calculated by a CSTR model.

In the case of washing with water, material balances yield the following expression for the component concentration in the retentate (4):

$$C = C_0 e^{-(1-\sigma)V_D} \tag{1}$$

and in the permeate:

$$C_P = C_0\,(1 - \sigma)e^{-(1-\sigma)V_D} \tag{2}$$

where C_0 = initial component concentration in the solution to be treated
σ = retention coefficient of the component
V_D = volumes of water per volume of solution to be treated

In the case of washing with a solution containing a component at concentration C_{IN}, a mathematical model has to be developed for the component concentration in the retentate. In the following, the equations for the model derivation are presented.

Let V_P = volume of liquid permeated
V_0 = volume of solution to be treated
N = amount of component in the system
P = probability a particle of the component will pass through the membrane
C = concentration of the component in the system (equal to N/V_0)

For small changes in the volume of liquid permeated (ΔV_P), the change in the amount of component in the system (tank T1, see Fig. 1), indicated as ΔN, is

$$\Delta N = -C\Delta V_P P + C_{IN}\Delta V_P \tag{3}$$

where $-C\Delta V_P P$ is the variation associated with the permeation process and $C_{IN}\Delta V_P$ is the amount of the component introduced in the system with the washing solution. It is clear that the total volume in the system remains constant (V_0), since the permeated volume is equal to the inlet volume. The probability that one particle will pass through the membrane can be expressed as a function of the retention coefficient (σ) as follows (4):

$$P = 1 - \sigma = C_P/C \tag{4}$$

Dividing both sides of Eq. (3) by the initial feed volume, V_0, substituting Eq. (4), and converting to differential form yields

$$dC = -C\,dV_D(1 - \sigma) + C_{IN}\,dV_D \tag{5}$$

Integrating with initial conditions $C = C_0$ at $V_D = 0$ yields the following expression for the component concentration in the retentate as a function of permeated volumes:

$$C = \frac{1}{1-\sigma}\{C_{IN} - [C_{IN} - C_0(1-\sigma)]e^{-V_D(1-\sigma)}\} \tag{6}$$

The component concentration in the permeate, C_P, can be determined by mul-

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tiplying the concentration C (Eq. 6) by the probability that one particle will pass through the membrane, P (Eq. 4), as follows:

$$C_P = CP = C_{IN} - [C_{IN} - C_0(1 - \sigma)]e^{-V_D(1-\sigma)} \qquad (7)$$

Equations (6) and (7) give the component concentration in the retentate and in the permeate, respectively, during a diafiltration process with a component in the washing solution (9).

RESULTS AND DISCUSSION

This section reports the results obtained in both experimental trials and simulation tests. The experimental work was performed in order to validate Eqs. (2) and (7). In particular, trials with a lactose solution as the washing system were performed for the reproduction of every single stage of a multistage process (excluding the last one, where washing is with water). In this way an equation representative of each stage is available and applicable in the simulation tests where a multistage diafiltration process was examined with the number of stages ranging from 2 to 6. The experimental trials also allow estimation of the lactose retention coefficient in the presence of whey, and this value has been used during the simulations.

Experimental Results

Some single-stage continuous diafiltration tests have been performed in a laboratory-scale apparatus using either distilled water or lactose (3 g/L) as the washing solution in order to validate Eqs. (2) and (7). The solution to be treated was either lactose (20 g/L) or whey. Figures 2 and 3 show the profiles of lactose concentration in the permeate (referred to the initial concentration) versus the permeated volumes during the experimental tests. The continuous lines have been calculated by Eq. (2) for washing with water and by Eq. (7) for washing with lactose (3 g/L). The lactose retention coefficient was experimentally estimated (according to Eq. 4) at 0 for a pure lactose solution 20 (g/L) and at 0.07 ($\pm$ 0.01) for lactose in real whey. There is good agreement between experimental (points) and calculated data in both tests. In particular, the curves obtained with lactose (3 g/L) as the washing solution validate the developed model. For the lactose retention coefficient in the presence of whey, a value equal to 0 can be used. In fact, even if it is statistically different with respect to the experimentally estimated value (that is, 0.07 $\pm$ 0.01), it gives results that agree quite well with experimental data when used in Eq. (7). Nevertheless, a value of 0.07 has been assumed in the simulations that follow.

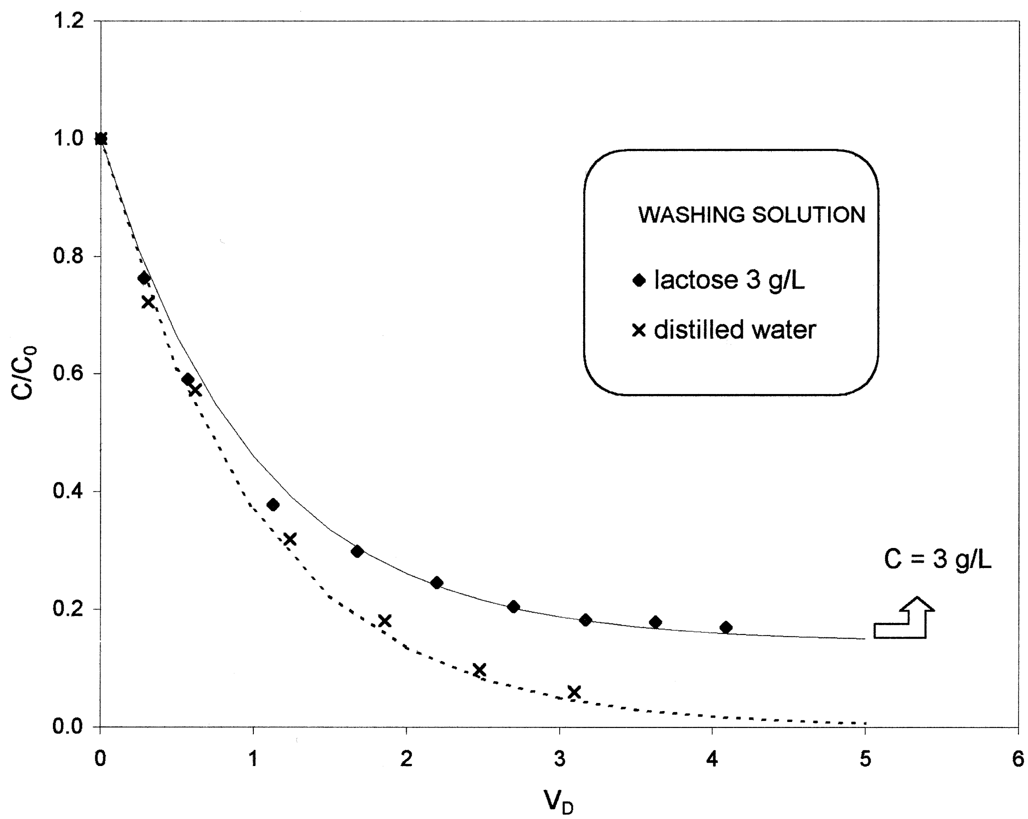

FIG. 2 Experimental (points) and predicted (lines) dimensionless lactose concentration vs diluted volumes profiles during diafiltration of a lactose solution of 20 g/L using distilled water and lactose (3 g/L) as washing solutions. Lines have been calculated by Eqs. (2) and (7), where the retention coefficients were fixed as experimentally determined (see text for details).

Multistage Countercurrent Diafiltration Simulation

Conventional continuous diafiltration involves adding a washing solution at the appropriate pH and temperature to the feed tank at the same rate (F) as the permeate flux, thus keeping the feed volume, V_0, constant during processing. This is a single-stage operation: when the component concentration reaches a target value, the operation is stopped. The experimental tests performed have been a single-stage continuous diafiltration. Obviously, in the case of a washing solution containing a component at a concentration C_{IN}, as shown by Figs. 2 and 3, the target value of the component concentration in the retentate cannot be lower than C_{IN}.

In order to produce WPC at a target value $> 80\%$ and at the same time save water, a countercurrent multistage process might be performed where pure

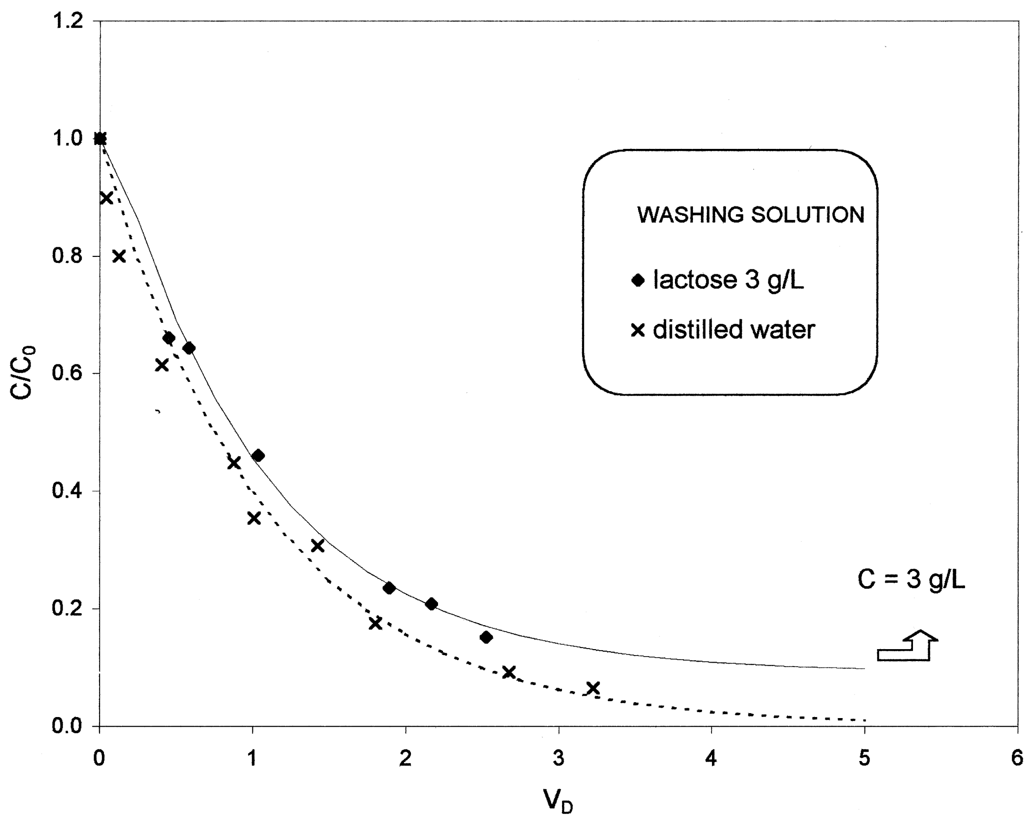

FIG. 3 Experimental (points) and predicted (lines) dimensionless lactose concentration vs diluted volumes profiles during whey diafiltration using distilled water and lactose (3 g/L) as washing solutions. Lines have been calculated by Eqs. (2) and (7), where the retention coefficients were fixed as experimentally determined (see text for details).

water is used as the washing solution only in the last stage. In this case the permeated volumes are equal in each stage. Figure 4 shows the flow sheet for a two-stage diafiltration. The operation takes place in two steps: first, the feed (whey) is washed with a solution containing all the whey components passed through the membrane, which was permeated in the previous diafiltration with water, and second, it is washed with water. The permeate of this second step is stored and used for the next washing.

The process takes place according to the following steps:

1. Whey is charged to Tank 1 (T1) in a batch mode. Valve 1 is open and the other valves are closed during the filling step (V_0 will be the final volume in T1).
2. Valve 1 is closed and a diafiltration process takes place. The washing so-

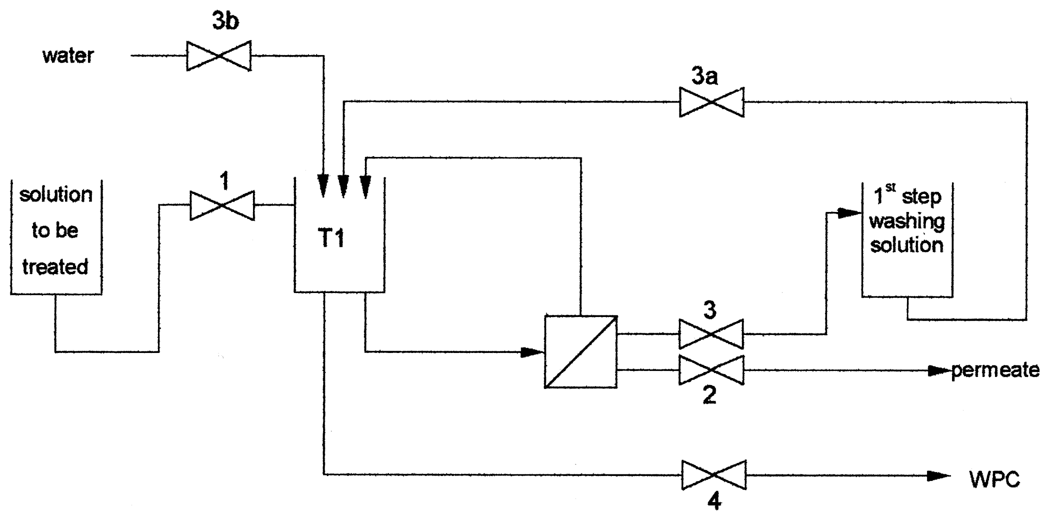

FIG. 4 Two stages countercurrent diafiltration flow sheet (see text for details).

lution is added at constant V_0 (Valves 3a and 2 are open; Valves 3, 3b, and 4 are closed) and the retentate is completely recirculated. Valve 3a is open in this first washing in order to remove most of components from Tank 1 with approximately $\sigma = 0$.

3. A final washing is carried out with pure water (Valve 3a is closed while Valve 3b is open), and the permeate is collected (in this case Valve 2 is closed and Valve 3 is open) for the first washing (Step 2) of the next batch of whey.
4. As soon as the WPC reaches the target value, the solution in Tank T1 is discharged (Valve 4 is open) and sent to drying for WPC production.

In what follows an expression for a particular component concentration in the retentate at the end of the two-stage process is determined as a function of permeated volumes in each stage, V_D. In the case of WPC production, this component can be lactose, or proteins, or ashes, etc. Let the solution to be treated contain the component at a concentration C_0. After the first stage, where washing is performed with a solution at a component concentration $C_{IN,1}$, the concentration of the component in the retentate is given by Eq. (6). This concentration is the component concentration at the beginning of the second stage, where washing is performed with water. Consequently, according to Eq. (6), the following expression gives the initial component concentration for the second stage, $C_{0,2}$:

$$C_{0,2} = \frac{1}{1-\sigma}\{C_{IN,1} - [C_{IN,1} - C_0(1-\sigma)]e^{-V_D(1-\sigma)}\} \qquad (8)$$

The concentration $C_{IN,1}$ is the component concentration in the permeate stored during the washing step with water obtained in the previous diafiltration process. Consequently, it can be calculated by the following expression:

$$C_{\mathrm{IN},1} = \frac{1}{V_\mathrm{D}} \int_0^{V_\mathrm{D}} C_{\mathrm{P},2}\, dV_\mathrm{D} \tag{9}$$

where $C_{\mathrm{P},2}$ is given by Eq. (2), with the initial concentration $C_{0,2}$. Combining Eqs. (2) and (9) and integrating yields

$$C_{\mathrm{IN},1} = \frac{C_{0,2}}{V_\mathrm{D}}[1 - e^{-V_\mathrm{D}(1-\sigma)}] \tag{10}$$

The component concentration in the retentate at the end of the water washing step can be calculated by Eq. (1), with an initial concentration $C_{0,2}$:

$$C = C_{0,2} e^{-V_\mathrm{D}(1-\sigma)} \tag{11}$$

This concentration represents the component concentration in the retentate at the end of the two-step countercurrent process. By combining Eqs. (8), (10), and (11), the following expression is obtained where the component concentration at the end of the process is a function of only the permeated volumes, V_D, and the component concentration in the solution to be treated, C_0:

$$C = C_0 V_\mathrm{D}(1-\sigma)\frac{e^{-2V_\mathrm{D}(1-\sigma)}}{V_\mathrm{D} - V_\mathrm{D}\sigma - 1 + 2e^{-V_\mathrm{D}(1-\sigma)} - e^{-2V_\mathrm{D}(1-\sigma)}} \tag{12}$$

[We wish to point out that the equivalent expression for C in Ref. 9 contains a misprint. In fact, the exponential term is $-2V_\mathrm{D}(1-\sigma)$ and not $(-V_\mathrm{D}(1-\sigma))^2$ as in that reference.]

An analogous procedure can be followed for an n-stages countercurrent diafiltration process (Fig. 5). In each stage the initial feed concentration for the

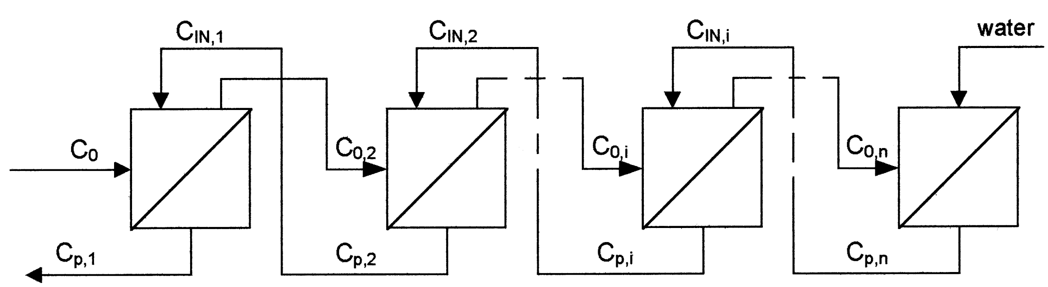

FIG. 5 Multistage countercurrent diafiltration block diagram.

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subsequent stage is calculated together with the washing solution concentration for the previous stage. As concerns the first stage, Eq. (8) gives the initial component concentration for the second stage.

2nd stage: washing solution with a component concentration $C_{\mathrm{IN},2}$:

$$C_{0,3} = \frac{1}{1-\sigma}\{C_{\mathrm{IN},2} - [C_{\mathrm{IN},2} - C_{0,2}(1-\sigma)]e^{-V_{\mathrm{D}}(1-\sigma)}\} \tag{13}$$

$$\begin{aligned} C_{\mathrm{IN},1} &= \frac{1}{V_{\mathrm{D}}}\int_0^{V_{\mathrm{D}}} C_{\mathrm{P},2}\,dV_{\mathrm{D}} \\ &= \frac{C_{\mathrm{IN},2} - C_{\mathrm{IN},2} - C_{0,2}(1-\sigma)}{V_{\mathrm{D}}(1-\sigma)}[1-e^{-V_{\mathrm{D}}(1-\sigma)}] \end{aligned} \tag{14}$$

ith stage: washing solution with a component concentration $C_{\mathrm{IN},i}$:

$$C_{0,i+1} = \frac{1}{1-\sigma}\{C_{\mathrm{IN},i} - [C_{\mathrm{IN},i} - C_{0,i}(1-\sigma)]e^{-V_{\mathrm{D}}(1-\sigma)}\} \tag{15}$$

$$\begin{aligned} C_{\mathrm{IN},i-1} &= \frac{1}{V_{\mathrm{D}}}\int_0^{V_{\mathrm{D}}} C_{\mathrm{P},i}\,dV_{\mathrm{D}} \\ &= C_{\mathrm{IN},i} - \frac{C_{\mathrm{IN},i} - C_{0,i}(1-\sigma)}{V_{\mathrm{D}}(1-\sigma)}[1-e^{-V_{\mathrm{D}}(1-\sigma)}] \end{aligned} \tag{16}$$

nth stage: washing with water:

$$C = C_{0,n}e^{-(1-\sigma)V_{\mathrm{D}}} \tag{17}$$

$$C_{\mathrm{IN},n-1} = \frac{C_{0,n}}{V_{\mathrm{D}}}[1-e^{-V_{\mathrm{D}}(1-\sigma)}] \tag{18}$$

where $C_{\mathrm{P},i}$ = component concentration in the permeate of stage i
$C_{0,i}$ = component concentration at the beginning of stage i
$C_{\mathrm{IN},i}$ = component concentration in the washing solution of stage i
C_0 = component concentration in the solution to be treated
C = component concentration in the retentate at the end of the n-stages countercurrent diafiltration process

In the case of a number of stages greater than 3, Eqs. (16) to (22) do not give an analytical solution like Eq. (15), but they have to be solved using an iterative method, as follows. A value is assumed for $C_{\mathrm{IN},1}$, and $C_{0,2}$ is calculated by using Eq. (8). All variables are then calculated progressively: $C_{\mathrm{IN},i}$ from the

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implicit Eq. (16) and $C_{0,i+1}$ from Eq. (15). Convergence is obtained if Eq. (18) is satisfied; otherwise a new value has to be chosen for $C_{IN,1}$.

Equations (12) to (18) have been applied for the simulation of a multistage countercurrent diafiltration process for the production of WPC. Whey composition is reported in Table 1. The retention coefficients were assumed as follows: 0.99 for proteins, 0 for nonproteinic nitrogen, 0.2 for ashes (4), and 0.07 for lactose, as experimentally determined.

Figure 6 reports the calculated volumes of water as a function of the target WPC at the end of the process. The volumes of water as a function of the number of stages for a target WPC of 95.5% are also indicated in the figure. The simulations have been performed for a process with up to six stages. It is evident, as expected, that as the number of stages increases, the volumes of water necessary decrease. Nevertheless, the incremental water saving diminishes as the number of stages increases. This aspect is shown by Fig. 7 where the water saving with respect to single-stage diafiltration is reported as a function

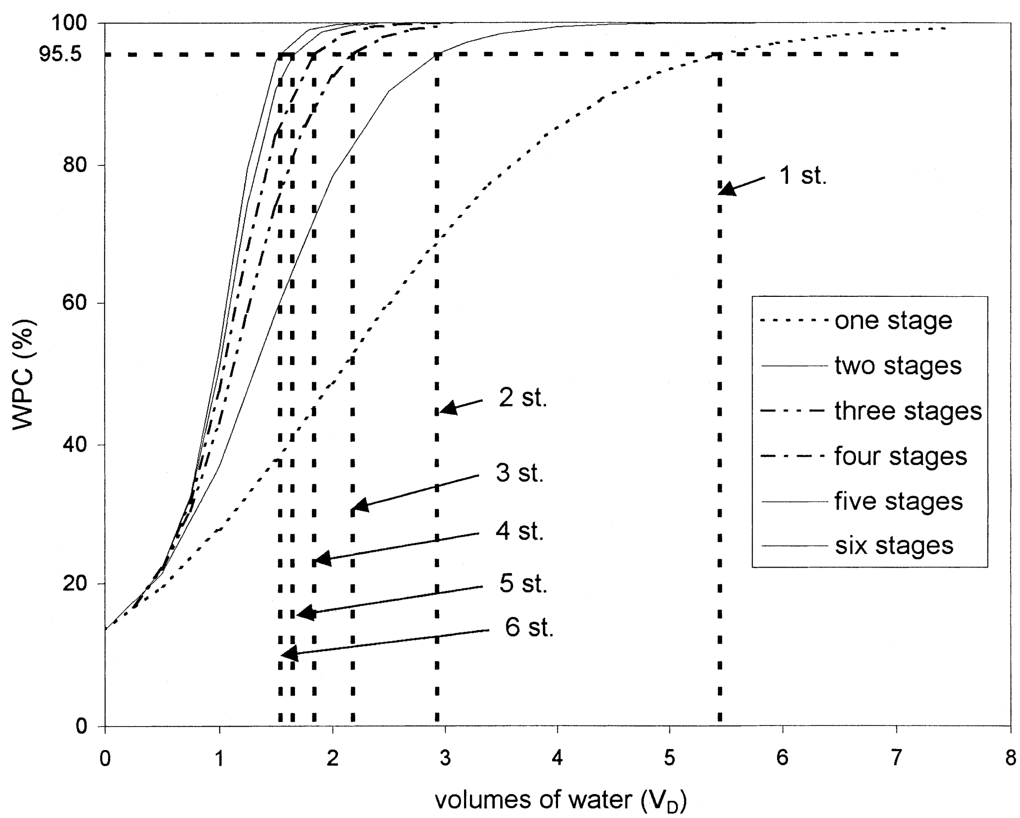

FIG. 6 Volumes of water as a function of target WPC (%) from one stage to six stages. The necessary water for a target WPC of 95.5% is evidenced.

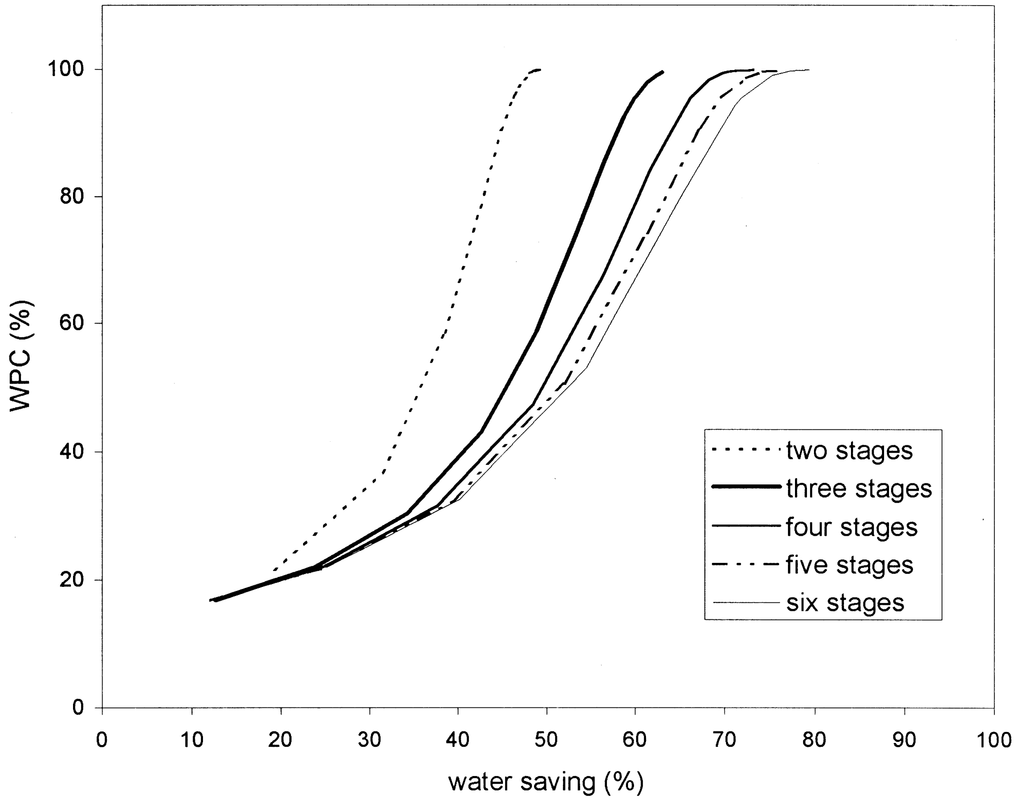

FIG. 7 Water saving with respect to a single stage as a function of target WPC.

of the target WPC. In particular, it is evident that as the number of stages increases from four to six, the water saving does not increase significantly.

Figure 8 shows the WPC profile in the retentate during single-stage and two-stage processes for a target WPC of 95.5%. As shown by Fig. 6, the permeated volumes are 5.5 in the single-stage process and 2.9 in each stage of the two-stage process. Note that Fig. 8 shows that in the first of the two stages, where whey is washed with a solution containing all the components (proteins, lactose, NPN, ashes), a preliminary washing from WPC 14% to about 60% is achieved. In the second step, washing with water realizes a further concentration from 60% to 95.5%.

CONCLUSIONS

A multistage countercurrent diafiltration process for WPC production is presented in this work. Experimental tests have validated a model developed

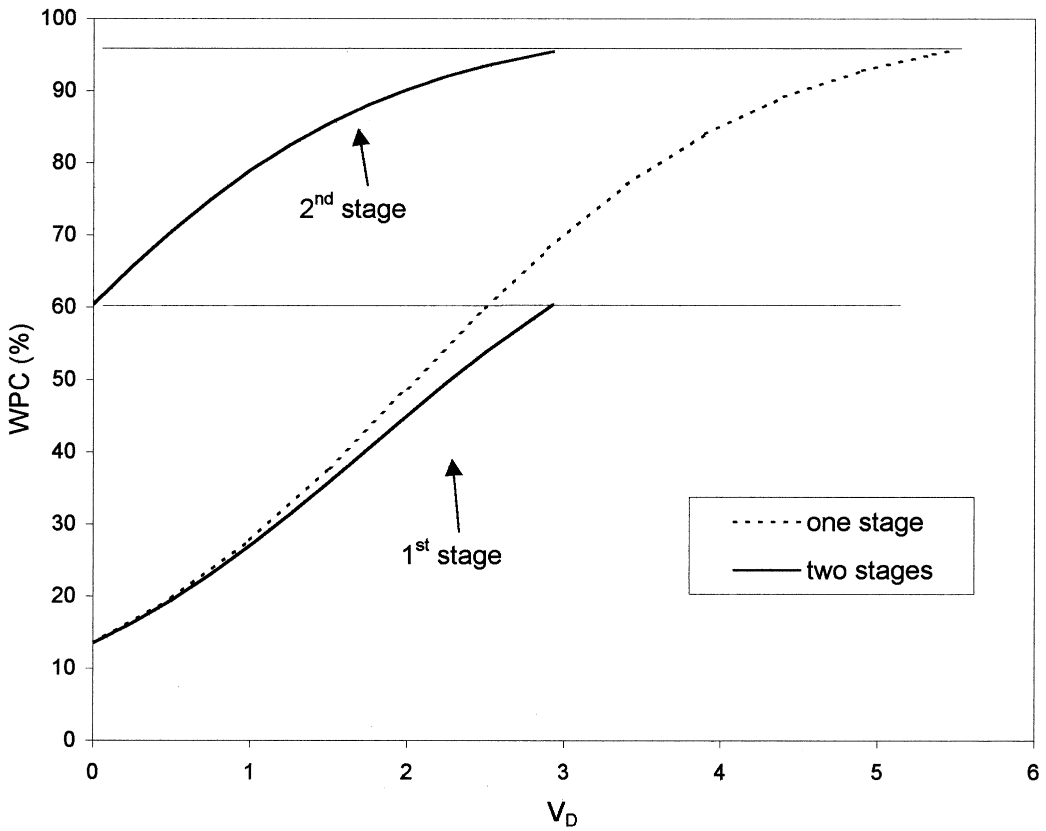

FIG. 8 WPC profiles during a single stage and a two stages diafiltration process for a target WPC of 95.5%.

for a particular component concentration in the retentate during a diafiltration where washing is performed with a solution containing that component. Material balances and the developed model—together with a model found in the literature for water as the washing system (4)—have allowed simulation of a multistage process which demonstrates water saving with respect to a single-stage conventional diafiltration. This aspect is very interesting considering that water costs are quite important in the overall operating costs of a diafiltration operation. Furthermore, and considering that investment costs depend primarily on the filtrating surface (4), the countercurrent process might be realized in industry with a one-membrane system and several holding tanks rather than with multiple membrane systems that operate continuously. This is true even with the relatively low added value of WPC. In the case of high added value products (such as pharmaceuticals), a continuous process might be justified. In the case of WPC 95.5% production from a whey containing 7.7

g/L proteins, 43 g/L lactose, 1.1 g/L nonproteic nitrogen, and 5.4 g/L ashes, simulations have shown that the volumes of water decrease from 5.4 for a single-stage process to 2.9 for a two-stage process down to 1.5 for a six-stage process. Furthermore, the water saving does not increase significantly with the number of stages when the number of stages is relatively high. Consequently, an optimization technique might be used for the determination of the optimum number of stages. Further work is in progress in this direction, both in simulation and in a real case application on the pilot scale. The adopted procedure can be easily applied to any diafiltration process.

ACKNOWLEDGMENTS

The authors are grateful to Dr. Daniela Cifoni and Mrs. Lia Mosca for their helpful collaboration in the experimental work.

SYMBOLS

C	component concentration in the retentate at the end of the n-stages countercurrent diafiltration process (mg/L)
C_0	initial component concentration in the solution to be treated (mg/L)
$C_{0,i}$	component concentration at the beginning of stage i (mg/L)
C_{IN}	component concentration in the washing solution (mg/L)
$C_{\mathrm{IN},i}$	component concentration in the washing solution of stage i (mg/L)
C_{p}	component concentration in the permeate (mg/L)
$C_{\mathrm{p},i}$	component concentration in the permeate of stage i (mg/L)
N	amount of component in the system (mg)
P	probability that a particle of the component will pass through the membrane
V_{D}	volumes permeated per volume of solution to be treated
V_{P}	volume of liquid permeated (L)
σ	component retention coefficient

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