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WHEY PROTEIN CONCENTRATE PRODUCTION IN A PILOT SCALE TWO-STAGE DIAFILTRATION PROCESS

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ABSTRACT

A pilot scale two-stage batch diafiltration process for whey protein concentrate (WPC) production is presented in this work. This process has two main advantages: a significant water saving with respect to a single-stage diafiltration process and a membrane surface saving with respect to a continuous multistage process. Every unit operation of the process has been experimented in a pilot scale (ultrafiltration, diafiltration, drying), in order to produce a WPC powder. Lactose content decreased from about 75% (of whey) to 4.5% (calculated as mass of lactose per total solute mass) and proteins increased from 15% to 83% (calculated as mass of proteins per total solute mass), with a water consumption of about 1.5 L/L

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of whey. Permeability tests enabled the calculation of the main mass-transfer resistances through the membrane: the intrinsic membrane resistance, R_M, was 13.8 ± 0.1, the one due to fouling, R_F, was 3.68 ± 0.04, and the one due to polarization, R_G, was estimated at 2.4 ± 0.4 (at 150 kPa TMP) 10^9 Pa s m^{-1}, at 25°C. These values are in agreement with data reported in the literature. A study performed on the membrane retention properties showed that not only peptides but also two native proteins (β-lactoglobulin and α-lactoalbumin) pass in the permeate during ultrafiltration. Consequently, a significant protein loss takes place during the process. Diafiltration tests performed in a two-stage countercurrent operation demonstrated the water saving with respect to a single-stage process (46%). Furthermore, experimental data have been used in order to evidence the capability of a previously developed mathematical model to predict the components' concentration during diafiltration.

INTRODUCTION

Whey proteins are of higher nutritive value than many other animal proteins (13). These ingredients have been used extensively in various segments of the food industry including dairy, bakery, meat industry, confectionery, beverage production, and the manufacture of baby and dietary foods (6). Due to progress in membrane technology, it is possible now to obtain WPC containing the desired quantities of soluble native proteins, lactose, and mineral matter (13). To obtain WPC with high protein concentration, water is normally added during ultrafiltration, realizing a *diafiltration* operation, whose main purpose is to remove residual quantities of lactose and minerals from whey. WPC thus obtained is widely employable in food product applications. Diafiltration is characterized by high water consumption (3). It is well documented in the literature (2, 8, 9, 11) that a remarkable water saving is achieved in a prediafiltrative concentration step in which the product concentration in UF 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. Furthermore, a batch countercurrent operation may be considered instead of a continuous one, in order to make further savings in membrane area. A detailed description of the batch countercurrent process is given in a previous work (1), where a mathematical model has been presented in order to predict the components' concentration during the process. In that paper, the water saving with respect to a single-stage process was also outlined. Figure 1 shows the flowsheet of a two-stage batch countercurrent diafiltration. The operation takes place in two steps: first the feed

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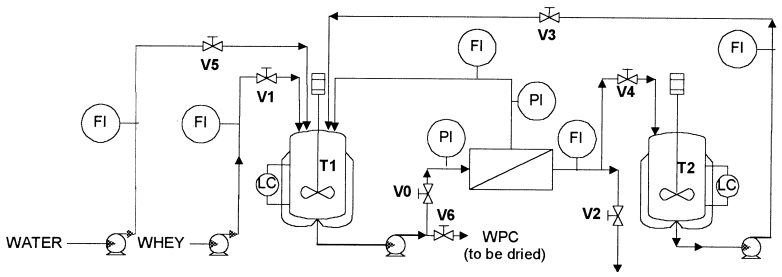

Figure 1. Flowsheet of the experimental apparatus for a two-stage batch diafiltration.

(whey) is washed with a solution containing all the whey components passed through the membrane, which was permeated in the previous diafiltration with water. Secondly, it is washed with water. The permeate of this second step is stored and used for the next washing. In particular, the process takes place according to the following steps:

1. Whey is charged in tank 1 (T1) in batch mode. Valve 1 (V1) is open and other valves are closed during the filling step (V_0 will be the final volume in T1).
2. Then valve 1 is closed and a diafiltration process takes place (valve V0 is open). The washing solution is added at constant V_0 (valves V2 and V3 are open, others are closed) and the retentate is completely recirculated. In this first washing, valve V2 is open in order to remove from tank 1 most of components with approximately $\sigma = 0$.
3. A final washing is carried out with pure water (valve V3 is closed while valve V5 is open) and the permeate is collected in the storage tank T2 (in this case valve V2 is closed and valve V4 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 V6 open and V0 closed) and sent to drying for WPC production.

In the case of an *n*-stage batch diafiltration, *n*-1 storage tanks (like tank T2) and just one membrane module would be necessary for the process. The saving in membrane area with respect to a continuous process is consequently obvious where *n* modules are requested (Fig. 2 shows the block diagram of a two-stage continuous countercurrent diafiltration, as example) (3). Nevertheless, this saving led to an increase in the plant complexity, and this aspect also has to be taken into account during the process design step.

The aim of the present work was to produce a powder of whey protein concentrate (>80%, on a dry basis) in a pilot scale. The membrane processes have

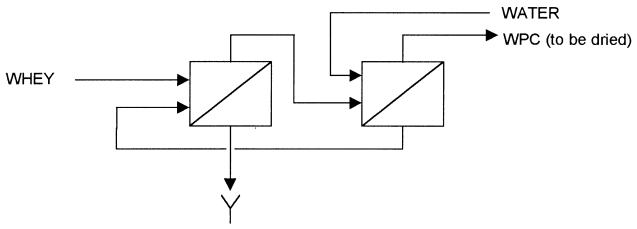

Figure 2. Block diagram of a two-stage continuous countercurrent diafiltration (3).

been characterized, as concern mass-transfer resistances through the membrane and protein retention properties. Furthermore, the diafiltration experimental data (performed in a two-stage batch countercurrent operation) have been used to successfully test the model developed in the previous paper (1).

THEORY

Mass-Transfer Resistances

The resistance model (3) was applied for the fitting of permeability data. In the case of a clean membrane and water as the feed:

$$J_p = \frac{TMP}{R_M} \tag{1}$$

where

J_p [$L/h/m^2$]	is the permeate flux;
TMP [kPa]	is the transmembrane pressure; and
R_M [kPa h m^2/L]	is the membrane intrinsic resistance.

In the case of whey as the feed:

$$J_p = \frac{TMP}{R_M + R_F + R_G} \tag{2}$$

where

R_F [kPa h m^2/L]	is the resistance due to membrane fouling, not dependent on pressure; and
R_G [kPa h m^2/L]	is the resistance due to concentration polarization and boundary layer and it is a function of the applied pressure.

The permeate viscosity was lumped in with the membrane resistance (Eqs. 1 and 2), considering that temperature was constant. Equation (2) can be modified as follows (3):

$$J_p = \frac{TMP}{R'_M + \phi \cdot TMP} \tag{3}$$

where R'_M is equal to $R_M + R_F$, and it can be determined using water as the feed on a fouled membrane, and ϕ TMP is equal to R_G.

Flux Decline

The following empirical equation has been fitted to permeate flux vs. time declines (3):

$$J_p = J_\infty + D\, e^{-t/\tau} \tag{4}$$

where

J_∞ [L/h/m^2]	is the steady-state flux;
D [L/h/m^2]	corresponds to the drop in flux in the time interval from the start of the experiment to the development of steady state, and it is indicated in the following as *stationary flux decline;* and
τ [min]	is the decline time constant, that is the time where the 63% of the stationary flux decline is achieved.

Retention Coefficient

The membrane retention coefficient was calculated according to the following relation:

$$\sigma = 1 - \frac{C_P}{C_R} \tag{5}$$

in which C_P [g/L] and C_R [g/L] are the measured component concentrations in the permeate and in the retentate, respectively.

Single-Stage Diafiltration

The system employed in the experimental tests has been modeled as a continuous stirred tank reactor (CSTR). In fact, some experiments performed with

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a tracer—whose retention coefficient through the membrane was 0—showed a perfect agreement between the experimental response to a step variation and the calculated one by a CSTR model.

In the case of washing with water, material balances on a component yield the following expression for the component concentration in the retentate (3):

$$C = C_0\, e^{-(1-\sigma)V_D} \tag{6}$$

and in the permeate

$$C_P = C_0(1 - \sigma)\, e^{-(1-\sigma)V_D} \tag{7}$$

where C_0 initial component concentration in the solution to be treated;

σ retention coefficient of the component; and

V_D volumes of water per volume of solution to be treated.

In the case of washing with a solution containing a component at a concentration C_{IN}, a mathematical model has been developed for the component concentration in the retentate (1). The following equations have been achieved for the component concentration in the retentate, C, and in the permeate, C_P, as a function of permeated volumes:

$$C = \frac{1}{1 - \sigma}\,\{C_{IN} - [C_{IN} - C_0(1 - \sigma)]\cdot e^{-V_D(1-\sigma)} \tag{8}$$

$$C_P = C_{IN} - [C_{IN} - C_0(1 - \sigma)]\cdot e^{-V_D(1-\sigma)} \tag{9}$$

Two-Stage Batch Countercurrent Diafiltration

Conventional continuous diafiltration involves adding a washing solution at the appropriate pH and temperature to the feed tank at the same rate as the permeate flux, thus keeping 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. Obviously, in the case of a washing solution containing a component at a concentration C_{IN}, the target value of the component concentration in the retentate cannot be lower than C_{IN}. In order to produce WPC with a target value >80% protein and at the same time save water, a countercurrent multistage process might be performed, where pure water is used as the washing solution just in the last stage. In this case, the permeated volumes are equal in each stage. A further saving might be realized if a batch operation is considered instead of a continuous one. In fact, just one membrane module and one storage tank are necessary. On the other hand two membrane modules would be necessary in the case of a two-stage continuous process.

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In a previous work (1), the following expression for a particular component concentration in the retentate at the end of the two-stage process was determined as a function of permeated volumes in each stage, V_D. In the case of WPC production, this component can be either lactose, proteins, or ashes, and so on:

$$C = C_0 \cdot V_D \cdot (1 - \sigma) \frac{e^{-2V_D(1-\sigma)}}{V_D - V_D \cdot \sigma - 1 + 2 \cdot e^{-V_D(1-\sigma)} - e^{-2V_D(1-\sigma)}} \qquad (10)$$

where C_0 is the component initial concentration (i.e., the concentration of whey).

MATERIALS AND METHODS

Solutions

Whey was provided by a dairy farm near L'Aquila, Italy. Before each trial, it was acidified to pH 4 with H_2SO_4, in order to prevent contamination. Table 1 shows whey composition before acidification.

Membrane

Ultrafiltration and diafiltration tests were carried out in a pilot scale experimental apparatus (SGI, France). The membrane was a spiral wound Amicon type S10Y10. It is made of regenerated cellulose, with 10,000 Da MWCO, area 0.93 m^2, nominal channel height 0.7 mm, pH tolerance 2–13. After each trial, the membrane was cleaned according to a standard washing procedure suggested by the constructor: a first acid washing (15 min, 45°C, pH 2) is followed by an acid washing (20 min, 50°C, pH 12), and a final membrane reconditioning by deionized water.

Table 1. Chemical Composition (g/L) of Whey

pH	6.3
Lactose	43.5
Proteins	7.7
Total dry matter	64.8
Ashes	5.8

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Ultrafiltration Tests

For the permeability curve determination, ultrafiltration was started with recirculation of both retentate and permeate, in order to have constant operating conditions during the test. After a period of recirculation, the permeate was collected for flux determination. Then, the inlet pressure was increased and after a further interval a subsequent permeate flux determination was performed. For the flux decline determination, the retentate only was recirculated during the ultrafiltration process, and the permeate was periodically collected for flux determination. Main operating conditions were: tangential velocity 0.7 m/s and temperature 25°C. Periodically, aliquots (1.5 mL) of permeate were sampled for determination of whey components.

For the determination of retention coefficients, both retentate and permeate were recirculated, and they were periodically (after 30, 60, 90, and 120 min) sampled for determination of whey components

Diafiltration Tests

The diafiltration tests were carried out in a two-stage batch countercurrent operation. The experimental apparatus flowsheet is shown in Fig. 1 (see "Theory" section (for details on the operation). The feed volume was 26 L, and the main operating conditions were: transmembrane pressure 120 kPa; tangential velocity 0.7 m/s; temperature 25°C. Periodically, aliquot amounts (1.5 mL) of permeate were sampled for whey components' determination.

Drying Tests

A Niro spray dryer was used for the production of WPC powder. The residual moisture was 4%, with an inlet air temperature of 190°C and an outlet air temperature of 90°C.

Analytical Determinations

Lactose concentration was determined through lactose/galactose UV method, Boehringer Mannheim. Protein concentration was determined by the Kjeldahl's method for protein nitrogen (heating digester Velp Scientifica mod. DK6; automatic steam distilling unit Velp Scientifica mod. UDK 130; automatic titrator Crison mod. microTT 2050); the analyzed values of elemental N were multiplied by 6.38 in order to obtain protein concentration.

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Single proteins in the collected samples have been determined by liquid chromatography (Waters pump, model 510; Waters universal liquid chromatograph injector, model U6K; LC Spectrophotometer, Waters Lambda Max model 481, set at 280 nm; Biorad Chromatographic Column, model Biosil Sec-250 (300 × 7.8 mm) and Biorad Biosil 250 Guard Column (80 × 7.8 mm)).

RESULTS

Ultrafiltration

A preliminary investigation was performed in order to characterize membrane performance in terms of mass-transfer resistances, flux decline, and retention properties.

Permeate fluxes, *Jp*, were experimentally determined (data not shown) vs. transmembrane pressure, TMP, during permeability tests where the feed was: 1) water (on clean membrane); 2) whey and 3) water (on the fouled membrane), respectively. A straight-line relationship was achieved in the case of water, both with the clean and the fouled membrane. On the other hand, experimental data with whey presented a deviation from linearity, as a consequence of the mass-transfer control (3). Equation (1) was fitted to experimental data with water on clean membrane, in order to estimate the intrinsic membrane resistance, R_M. Subsequently, the resistance R'_M—which takes into account the intrinsic resistance, R_M, and the one due to fouling, R_F—was estimated from the slope of the regression line based on experimental data obtained using water on a fouled membrane. Equation (3) was then fitted to experimental data with whey through a nonlinear regression procedure (least squares method, modified by Mezaki) (7), with ϕ as adjustable parameter. The following resistances were determined: R_M 13.8 ± 0.1, R_F 3.68 ± 0.04, and R_G 2.4 ± 0.4 (at 150 kPa TMP) 10^9 Pa s m^{-1}. These values are in agreement with data reported in the literature (3, 4, 5, 10, 12), taking into account the different operating conditions.

Equation (4) was then fitted to experimental data of permeate flux vs. time (not shown here) through a nonlinear regression technique (7), and the following values were estimated for the three adjustable parameters: $J_\infty = 9 \pm 2$ L/h/m^2; $D = 15 \pm 2$ L/h/m^2; and $\tau = 40 \pm 10$ min. These values may be helpful in order to predict flux decline in the investigated operating conditions.

A further investigation was performed in order to characterize membrane retention properties. The retention coefficients of individual constituents were calculated according to Eq. (5) from experimental concentrations in the permeate and retentate, respectively. Table 2 shows the obtained values. It is evident that protein retention coefficient is equal to about 0.8, with a consequent loss of proteins. This aspect has to be taken into account during the process design step, as it influences

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Table 2. Experimental Values for Whey Components' Retention Coefficients

Component	σ
Proteins	0.78 ± 0.01
Lactose	0.05 ± 0.01
Ashes	0.010 ± 0.006

significantly the protein yields. With regard to lactose and ash, a value of 0 was used in subsequent calculations, instead of the experimentally determined value. In fact, there was no practical difference in the obtained results, even if the experimental retention coefficients were statistically different from zero. Permeate and retentate samples taken after 30, 60, 90, and 120 min of processing were analyzed using HPLC, in order to evaluate the retention coefficients of individual proteins and the effect of time on these quantities. Table 3 shows the estimated values, whereas Fig. 3 shows a chromatogram of the permeate after 30 min of processing (other chromatograms were similar and they are not reported here). Results in Table 3 suggest that there is no effect of time of processing on retention coefficient. In fact, σ values determined after various times are not significantly different. Figure 3 also illustrates that not only peptides but also two native proteins (β-lactoglobulin and α-lactoalbumin) pass in the permeate during ultrafiltration. This is confirmed by the retention coefficients of these proteins, reported in Table 3.

Diafiltration

Whey diafiltration has been performed in a two-stage batch countercurrent operation, in order save both water and membrane surface. In fact, water is used

Table 3. Whey Proteins Retention Coefficients after Different Times of Processing (Calculated by Eq. (5) through HPLC Determinations)

	σ			
Protein	30 min	60 min	90 min	120 min
Casein	1.0	1.0	1.0	1.0
IgG	1.0	1.0	1.0	1.0
BSA	1.0	1.0	1.0	1.0
β-lactoglobulin	0.92	0.94	0.89	0.90
α-lactoalbumin	0.78	0.78	0.77	0.79

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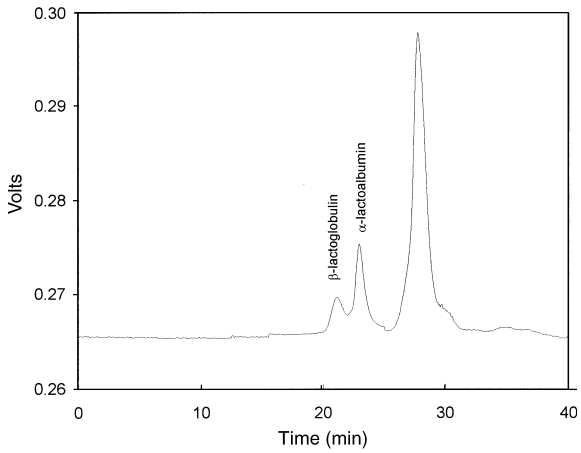

Figure 3. UF permeate chromatogram after 30 min of processing (determined spectrophotometrically at 280 nm).

just in the last stage, whereas the permeate of the previous cycle is used as washing solution for the first stage (see the "Theory" section for details on the operation). Furthermore, a saving in filtration surface is also achieved, with respect to a continuous countercurrent operation.

The treated solution was the retentate of a preliminary whey ultrafiltration (VCR 2), aimed at the reduction of volumes (and consequently of necessary water). Figure 4 shows the profiles of lactose concentration in the permeate vs. permeated volumes during each stage. It is evident that the most significant washing is achieved in the first stage, where a solution containing 1.8 g/L protein, 1.6 g/L lactose, and 0.34 g/L ash is used as washing system. This solu-tion was permeated during the second stage washing (with water) of a previous cycle. Continuous lines have been calculated by Eq. (7) in the case of washing with water (second stage) and by Eq. (9) in the case of washing with a solution containing whey components (first stage). It is evident there is good agreement between experimental (points) and calculated data in both stages.

Figure 5 shows protein concentration on a dry basis, W_P (%), vs. permeated volumes profiles during the two diafiltration stages. W_P has been calculated according to the following equation:

$$W_P = \frac{C_{proteins}}{C_{proteins} + C_{lactose} + C_{ashes} + C_{fats}} \cdot 100 \tag{11}$$

where the components' concentrations have been either experimentally determined (points) or calculated by Eq. (10) (lines). A first W_P enhancement from

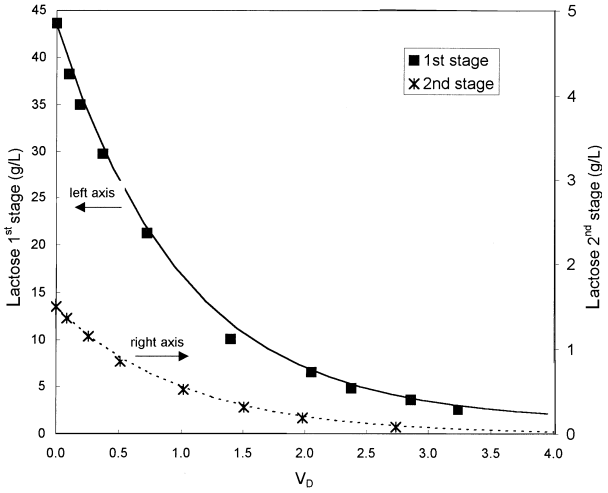

Figure 4. Lactose concentration in the permeate as a function of permeated volumes of washing solution, V_D, during the two diafiltration stages. Lines have been calculated by Eqs. (9) and (7) for the first and the second stage, respectively.

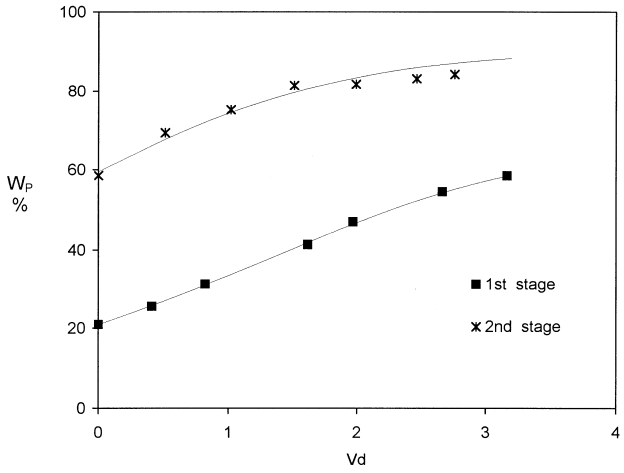

Figure 5. Protein concentration on a dry basis, W_P, profiles as a function of the permeated volumes of washing solution, during the two-stage diafiltration. Lines have been calculated by Eq. (10).

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about 20 to about 60% is realized in the first washing stage; a further increase to >80% is obtained in the second step, where washing is realized with water. A very good agreement between experimental and calculated data confirms the ability of Eq. (10) to predict the course of diafiltration.

Process Analysis

Figure 6 shows the block diagram of the pilot plant process realized for the WPC powder production. The streams' properties are reported in Table 4, and the process time diagram is shown in Fig. 7a. A first whey acidification (aimed at whey preservation) was followed by an ultrafiltration in order to reduce the volumes to be treated (and subsequently water consumption). This operation was carried out in 2 h for a volume concentration factor (VCR) of about two. The concentrated whey (stream 5) contained 13.6 g/L protein, 42.8 g/L lactose, 5.8 g/L ash, and 0.9 g/L fats. Then, the two-stage countercurrent diafiltration process took place. Even though it is represented in Fig. 6 as a continuous operation for clarity, it operated batchwise, as described in Fig. 1. First, a washing was realized with a solution coming from a previous diafiltration (precisely, it was the second stage washing with water, where the permeate was stored—stream 10). About 3 volumes (per volume of feed) of washing solution were introduced and the process was carried out for 5 h. After this first stage, a first whey protein concentration from 22 (stream 5) to 63% (stream 6) was realized, calculated as mass of proteins per total solute mass. Then a second-stage washing (with water) took place, and the protein concentration was brought to 83% (stream 9). This stage lasted about 3 h, for 3 volumes of water permeated (per volume of feed). The second-stage lasting time is obviously lower with respect to the first-stage time, as permeate fluxes are higher. This is probably associated

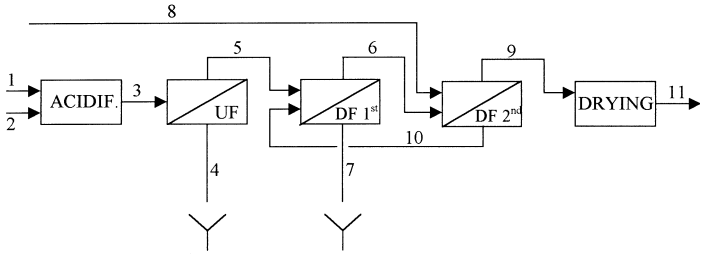

Figure 6. Block diagram of the process for WPC production (see Table 4 for stream properties).

Table 4. Characteristics of WPC Production Process Stream (see Fig. 6)

Stream	Fluid	Volume (L)	Proteins		Lactose		Ashes		Fats		Proteins[a] (%)
			g/L	g	g/L	g	g/L	g	g/L	g	
1	Whey	47	8.6	405	42.8	2000	5.8	270	0.5	23	15
2	H_2SO_4 0.1 N	0.05	—	—	—	—	—	—	—	—	—
3	Acid whey	47	8.6	405	42.8	2000	5.8	270	0.5	23	15
4	Permeate UF	21	2.4	49	42.8	900	5.8	120	0	0	—
5	Retentate UF	26	13.6	356	42.8	1100	5.8	150	0.9	23	22
6	Ret. DF first stage	26	11	290	4.7	123	1.0	26	0.9	23	63
7	Per. DF first stage	73	2.7	198	15	1100	2	150	0	0	—
8	Water	73	—	—	—	—	—	—	—	—	—
9	Ret. DF second stage	26	6	160	0.35	9	0.03	0.82	0.9	23	83
10	Per. DF second stage	73	1.8	135	1.6	114	0.34	25	0	0	—
11	WPC powder			160		9		0.82		23	83

[a] Calculated as mass of proteins per total solute mass.

to a lower protein concentration when washing is realized with water, due to the protein loss that takes place during the process (see Table 4, protein concentration expressed as g/L). A further concentration of proteins could not be realized, because of the presence of fat (whose retention coefficient for the membrane under test was 1). These may be eliminated by a preliminary whey microfiltration, and further work is in progress aimed at the production of higher concentration

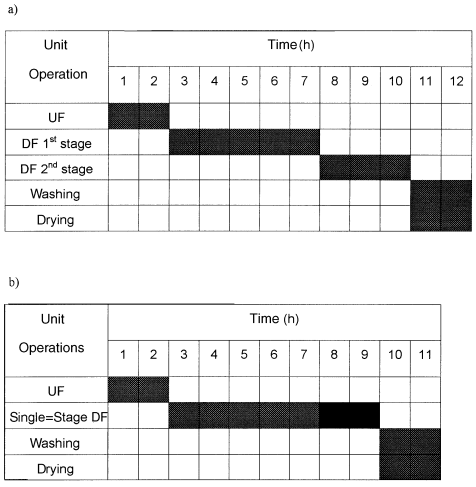

Figure 7. a) time diagram of the realized process for WPC production. b) of a control process based on a conventional approach (single diafiltration step).

WPC. Finally, the powdered WPC was produced in a spray dryer, where about 200 g of product were produced in two hours. Figure 7b also shows the time chart of a control process, based on a conventional approach (single-step diafiltration, washing with water), for comparative purposes. The only difference is obviously in the DF operation, which would last about 7 h. This value was calculated considering the water consumption (about 135 L for 26 L of whey) and a permeate flux evaluated at an average protein concentration. A comparison between the two processes evidences that the conventional process takes place in about 1 h less. Consequently, more batches may be carried out during the same period of time. This is another aspect that has to be taken into account during optimization in the process design step.

CONCLUSIONS

A process for the production of WPC has been evaluated in a pilot scale. The first two unit operations of the process (ultrafiltration, diafiltration) have been studied in detail. With respect to ultrafiltration, permeability tests enabled the calculation of the main mass-transfer resistances through the membrane: the intrinsic membrane resistance, R_M, the one due to fouling, R_F, and the one due to polarization. A further study performed on the membrane retention properties showed that not only peptides but also two native proteins (β-lactoglobulin and α-lactoalbumin) pass in the permeate during ultrafiltration. This aspect has to be taken into account during the process design step, as it influences significantly protein yields. Diafiltration was performed in a two-stage countercurrent batch mode, in order to have two main advantages: a significant water saving with respect to a single-stage diafiltration process and a membrane surface saving with respect to a continuous multistage process. A comparison between experimental data and a previously developed model (Eq. 10) indicated the usefulness of the model in predicting the components' concentration during diafiltration. Drying results in a WPC powder with a protein concentration of 83% (on dry basis) and a residual moisture of 4%. A further concentration of proteins could not be realized, because of the presence of fat (whose retention coefficient for the used membrane was 1). These may be eliminated by a preliminary whey microfiltration, and further work is in progress aimed at the production of higher concentration WPC.

NOMENCLATURE

C_0	initial component concentration in the solution to be treated
C_{IN}	component concentration in the washing solution (1st stage of a two-stage diafiltration)

C_P [g/L]	component concentration in the permeate
C_R [g/L]	component concentration in the retentate
CSTR	continuous stirred tank reactor
D [L/h/m^2]	drop in flux in the time interval from the start of the experiment to the development of steady state (it is indicated in the following as *stationary flux decline*)
J_∞ [L/h/m^2]	steady-state flux
J_p [L/h/m^2]	permeate flux
R_F [kPa h m^2/L]	resistance due to membrane fouling, not dependent on pressure
R_G [kPa h m^2/L]	resistance due to concentration polarization and boundary layer
R_M [kPa h m^2/L]	membrane intrinsic resistance
TMP [kPa]	transmembrane pressure
VCR	volume concentration ratio
V_D	volumes of water per volume of solution to be diafiltrated
W_P (%)	protein concentration on a dry basis (Eq. 11)
WPC	whey protein concentrate
σ	retention coefficient of the component
τ [min]	decline time constant (it is the time where the 63% of the stationary flux decline is achieved)

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REFERENCES

1. Barba, D.; Beolchini, F.; Veglió, F. Minimising Water Use in Diafiltration of Whey Protein Concentrates. Sep. Sci. Technol. **2000,** *35* (7), 951–966.
2. Carlsson, R.; Hanczakowski, P. The Nutritive Value of Mixtures of White Leaf Protein and Food Proteins. J. Sci. Food Agric. **1985,** *36*, 10, 946–950.
3. Cheryan M. *Ultrafiltration and Microfiltration Handbook*. Technomic Publishing Company: Lancaster, PA, 1998.
4. Da Costa, A.R.; Fane, A.G.; Wiley, D.E. Ultrafiltration of Whey Protein Solutions in Spacer-Filled Flat Channels. J. Membr. Sci. **1993,** *76*, 245–254.

5. Daufin, G.; Merin, U.; Labbé, J.P.; Quémerais, A.; Kerhervé, F.L. Cleaning of Inorganic Membranes after Whey and Milk Ultrafiltration. Biotechnol. Bioeng. **1991,** *38*, 82–89.
6. Gonzalez, J.; Real Del Sol, E.; Perea, J.; Martinez, G.; Ortega, A. Obtaining Delactosed Milk for Nursing Infants. Alimentaria **1993,** *30* (239), 67–71.
7. Himmelblau, D.M. *Process Analysis by Statistical Methods*. John Wiley and Sons: New York, 1970.
8. Morr, C.V. Composition Physicochemical and Functional Properties of Reference Whey Protein Concentrates. J. Food Sci. **1985,** *50* (5), 1406–1411.
9. Pompei, C.; Peri, C.; Ariatta Olivani, M. Whey Proteins Purification by Ultrafiltration and Diafiltration. Scienza e tecnologia degli alimenti **1973,** 209–215.
10. Pouliot, Y.; Wijers, M.C.; Gauthier, S.F.; Nadeau, L. Fractionation of Whey Protein Hydrolysates Using Charged UF/NF Membranes. Journal of Membr. Sci. **1999,** *158*, 105–114.
11. Slack, A.W.; Amundson, C.H.; Hill, C.G., Jr. Production of Enriched Beta Lactoglobulin and Alpha Lactalbumin Whey Protein Fractions. Journal of Food Processing and Preservation **1986,** *10* (1), 19–30.
12. Taddei, C.; Daufin, G.; Aimar, P.; Sanchez, V. Role of Some Whey Components on Mass Transfer in Ultrafiltration. Biotechnol. Bioeng. **1989,** *34*, 171–179.
13. Tratnik, L.; Krsev, L. Production of Whey Protein Concentrates. Milchwissenschaft **1991,** *46* (2), 91–94.

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