Investigation of ultra- and nanofiltration for utilization of whey protein and lactose

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Received 25 March 2003; accepted 26 April 2004

Abstract

Ultrafiltration is one of the most fascinating technologies, which has been introduced for application in the dairy industry. Ultrafiltration makes it possible to improve the quality of traditional dairy products, to create new food staffs, to utilize dairy by-products (such as whey) to a much greater extent for human nutrition and to prepare milk ingredients to be used in the entire food industry.

In this study the application of ultrafiltration for milk and whey protein concentration, and research on nanofiltration for lactose concentration of the ultrafiltration permeate are detailed.

The performance of ultra- and nanofiltration membranes can be characterized in terms of permeate flux, membrane retention and yield, which parameters are determined by pressure, recycle flow rate and temperature. The influence of these parameters on milk- and whey protein and lactose concentration was measured. The experiments were carried out using laboratory scale ultra- and nanofiltration units. The permeate flux, protein and lactose content in the permeate and in the concentrate fractions were measured during the experimental runs. Comparing the separation behavior of the membranes it was found that the investigated membranes are suitable for concentration of the milk- and whey proteins and lactose with high flux and retention. The filtration characteristics were obviously influenced by the process parameters.

A new combination of membrane based cheese production procedure is proposed, which makes possible a significant increase in the cheese yield by incorporating the whey proteins.

Keywords: Concentration of milk and whey; Ultrafiltration; Nanofiltration

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1. Introduction

Developments in ultrafiltration technology have created the opportunity for an entirely new approach to cheese technology. The particular application of the ultrafiltration technique to the treatment of milk for making cheese was covered in a French patent in 1969. The general approach was to concentrate skim milk by ultrafiltration and than combine cream with the unfermented or partially fermented retentate to form a pre-cheese (Ernstrom, Sutherland, & Jameson, 1980).

The basic difference between ultrafiltration technology and traditional cheese making is that one is dealing with concentrated milk, which may also have been extensively heat-treated or diafiltered (Lawrence, 1989). The principle of making cheese with ultrafiltered milk is to retain or to reintegrate whey proteins into the cheese to increase the yield of the product (Glover, 1986; Hinrichs, 2001; Kessler, 2002; Lawrence, 1993).

Whey protein concentrates, which are obtained by whey ultrafiltration are available in great variety according to protein content and functional properties. Another aspect which must not be neglected is the utilization of the by-product which is the permeate of ultrafiltration. In this study new technologies have been developed (using nanofiltration or reverse osmosis) for concentration of the lactose which can be applied in the sweets industry or in pharmaceutical fermentation procedures (Balint & Okos, 1995; Georgiev, Pashova, Veselka, Toncheva, & Ventislava, 1996; Renner & Abd El-Salam, 1991).

Ultra- and nanofiltration can be defined as pressure-driven membrane processes for the separation and concentration of substances having a molecular weight between 10^3 and 10^6 Da for ultrafiltration and between 100 and 500 Da for nanofiltration. In both processes a solution flows under pressure along the surface of a suitably supported membrane (cross-flow filtration). The solvent and certain dissolved components pass through the membrane and are collected as permeate. Depending on the characteristics of the applied membrane some other components from the solution are retained by the membrane and concentrated, as the retentate fraction (Fonyo & Fabry, 1998; Porter, 1990).

The dairy industry is one of the pioneers in the development of ultrafiltration equipment and techniques (Elyas, 1990). Ultrafiltration can be used in the cheese industry to fractionate the proteins from whey and to make cheese from ultrafiltered milk. Application of ultrafiltration in the dairy industry started with the separation and concentration of whey proteins from whey in 1972. Whey is the liquid remaining after the recovery of cheese. Whey contains more than the half of the solids of the original whole milk, including whey protein (20% of total protein) and most of the lactose, minerals and water-soluble vitamins. The principal aim of ultrafiltration of whey is to concentrate the native or pre-denatured whey proteins in order to obtain a whey protein powder with varying protein content and reduced lactose and ash content (Da Costa, Fane, & Wiley, 1993; Huffman, 1996; Marshall, 1982).

The application of ultrafiltration in cheese processing started in 1983 for production of cottage cheese and soft cheese varieties. In ultrafiltration the constituents of milk are fractionated according to molecular size. Depending on the retention characteristics of the membranes there can be a significant difference in the nutritive power of the retentate and permeate. The protein and fat fractions are retained very well (virtually completely) in the retentate, while the lactose, minerals and vitamins are divided between the retentate and the permeate (Hinrichs, 2001; Kessler, 2002).

Nanofiltration can be used for concentration of the permeate which penetrates the membrane during ultrafiltration processing of milk or whey and which contains lactose in the same concentration as in the water phase of the original fluid. Ultrafiltration permeate has a biological oxygen demand of 30,000–45,000 mg O_2/l and cannot, therefore, be directly discharged into sewage as wastewater (Puhan & Gallmann, 1981; Qureshi & Manderson, 1995).

The major purpose of this study is utilization of whey and to decrease the amount of waste in the cheese industry. The main steps of the investigations are: pre-concentration of milk using different pore size membranes; concentration of whey for incorporation of whey proteins for increasing cheese yield; concentration of lactose for use in the sweets industry or as a raw material for fermentation processes by using different membrane filtration processes.

2. Experimental

2.1. Materials and methods

The average composition of light milk and whey, which were ultrafiltered in this study are shown in Table 1.

In our study two different ultrafiltration membranes and one nanofiltration membrane have been investigated. SP015 and FS10 membranes were used for pre-concentration of the light milk, FS10 membrane for ultrafiltration of the whey and RA55 membrane for nanofiltration of the ultrafiltation permeate. The characteristics of the investigated membranes are shown in Table 2.

The measured filtration characteristics were defined as follows:

- Solute (protein and lactose) rejection (R):

$$R = \left(1 - \frac{c_P}{c_R}\right) 100\%$$

Table 1
The average composition of feed solutions

Feed solutions	Protein	Lactose	Fat	Ash	Calcium
Light milk [g/l]	33	48	15	7	1.2
Whey [g/l]	8.5	48.5	0.75	9.2	4.6
UF permeate [g/l]	4.2	48	1	8.5	3.3

Table 2
The characteristics of the investigated membranes

Membrane type	Membrane materials	Producer company	PWF ($T=25$ °C) [l/m^{2}h]	Membrane cut-off	Process parameters		
					Pressure [bar]	Temperature [°C]	pH
FS10 (UF)	Polyvinil-difluoride	Zoltek Rt MAVIBRAN	~150–200 [4 bar]	6–8 kDa	8	0–60	1–13
SP015 (UF)	Polyethersulfone	Zoltek Rt MAVIBRAN	~200–350 [4 bar]	15–20 kDa	6	0–60	1–13
RA55 (NF)	Polyamide	MILLIPORE	~58–60 [35 bar]	400 Da; $R_{NaCl}=80\%$	41	60	3–10

where c_P—solute concentration in the permeate (g/l), c_R—solute concentration in the retentate (g/l).

- Concentration factor (F):

$$F = V_F/V_R$$

where V_F—volume of feed (L); V_R—volume of retentate (L).

- Permeation time was the filtration time in batch mode.
- Solute (protein and lactose) yield (Y):

$$Y = \frac{V_R \cdot c_R}{V_F \cdot c_F}$$

where c_F—solute concentration in the feed (g/l).

2.2. Ultrafiltration of milk and whey (UF/M and UF/W)

The ultrafiltration experiments were carried out in a laboratory unit designed by the Department of Food Engineering and produced by the Hidrofilt Company (details are explained elsewhere, Atra, 2000). The effective area of the flat membrane in the ultrafiltration cell was 470 cm^2. The solution was circulated in contact with the membrane from a feed tank. The constant temperature of feed (30–50 °C) was maintained by using a thermostat bath, and controlled at the inlet of the membrane cell. The volume of permeate was measured during the experimental runs in the collector. The pressure (1–5 bar) and the recycle flow rate (100–400 l/h, or the tangential velocity 0.43–1.74 m/s) were controlled by regulation valves. The protein content in the permeate and in the concentrate fractions were determined by taking samples from this fractions during the experimental runs, which were analyzed by SORENSEN method (Elyas, 1990). The method is based on the oxidation of the proteins with an excess amount of formaldehyde, and the titration of the residue with sodium hydroxide.

2.3. Nanofiltration of ultrafiltration permeate (NF)

The nanofiltration apparatus used in the present study YPROLAB-2 was made by Millipore Co. (Atra, 2000). The area of the spiral wound membrane was 0.3 m^2. The solution was circulated in contact with the membrane from a feed tank. The constant temperature of feed (30–50 °C) was controlled using a thermostat bath. The permeate was measured during the experimental runs in the collector. The pressure (10–20 bar) was controlled by a regulation valve and the flow rate (100–200 l/h) was controlled by changing the rotation speed of the pump. The lactose content in the permeate and in the concentrate fractions were analyzed during the experimental runs using ZEISS-ABBE refractometer.

3. Results and discussion

3.1. Pre-concentration of light milk by ultrafiltration (UF/M)

The influence of the pressure at different flow rates on the permeate flux of milk and pure water flux (PWF) is shown in Fig. 1. Pressure and the recycle flow rate significantly influenced the permeate flux. Above a critical transmembrane pressure (2–4 bar) the flux becomes independent of pressure, because the protein molecules deposited on the surface of the membrane cause a concentration polarization controlled by two factors, the type of membrane and the recycle flow velocity. More open membranes give higher fluxes and hence are more susceptible to concentration polarization. The pressure independent region therefore occurs at lower pressures.

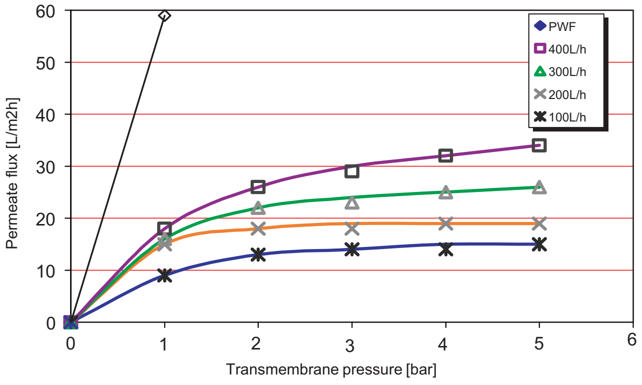

Fig. 1. Milk concentration by UF/M: influence of pressure on the permeate flux (FS10 membrane, 50 °C).

As flow velocity increases, concentration polarization decreases, hence the point of pressure independence advances to higher pressures. This phenomenon is in agreement with literature data (Kessler, 2002).

The effect of temperature on the permeate flux can be understood from its effect on the properties of the feed stream. Increasing the temperature results in a decrease in the viscosity of milk, resulting in an increase in permeate flow rate according to Hagen–Poisellue law. High temperature increases the solute diffusivity and the rate of transport of solutes from the membrane surface into the bulk stream. Generally, the permeate flux increases linearly with an increase in temperature; the rate in this study was about 0.5 l/(m^2 h °C), as it is illustrated in Fig. 2.

The influence of recycle flow rate on permeation time, for reaching certain protein concentration in retentate, is shown in Fig. 3. Higher flow rate at the membrane surface (for example 400 l/h, which means 1.74 m/s tangential velocity) is a very important factor in increasing the permeate flux. Using higher velocity the deposited molecules are continuously removed from the membrane surface and thus the hydraulic resistance of the fouling layer is reduced. The mass transfer of solutes through the boundary layer increases such that the required protein content in the permeate fraction can be achieved at shorter time. There are different methods which can be used to generate high turbulence: increasing the flow rate of the recycled stream, decreasing the flow channel dimensions or insertion of a static mixer.

The influence of pore size of the investigated membranes on the permeate flux is shown in Fig. 4. This diagram illustrates, that there is a significant influence of the membrane pore size on the permeate flux which increases with an increase in the cut-off of the membrane (see values in Table 2). Also, the protein content in the permeate fraction increases, because proteins with higher molecular size can be transported through the larger cut-off membrane. This phenomenon can be utilized for separation of the α-lacto albumin (molecular size 14 kDa) by diafiltration of milk using the SP015 membrane in the first step.

3.2. Concentration of whey by ultrafiltration (UF/W)

As in the case of ultrafiltration of milk the permeate flux of whey also increased with an increase in pressure. The influence of transmembrane pressure and concentration factor on permeate flux is shown in Fig. 5. At higher concentration factors a thicker and denser deposit layer is formed which reduces permeate flux until it reaches the steady-state condition.

It can be seen that the protein rejection slowly decreases with an increase in the pressure. The protein rejection of UF was higher: 93–98% (Fig. 6) at higher transmembrane pressures (3 and 5 bar) than at low pressure (1 bar).

The effect of the tangential crossflow velocity, i.e. recycle flow rate on permeate flux is shown in Fig. 7. It is obvious that the flux increases at higher crossflow velocities, that results in a decrease in the deposit layer resistance. Continued gain in flux is limited by energy, which can be afforded in pumping, and as well as will

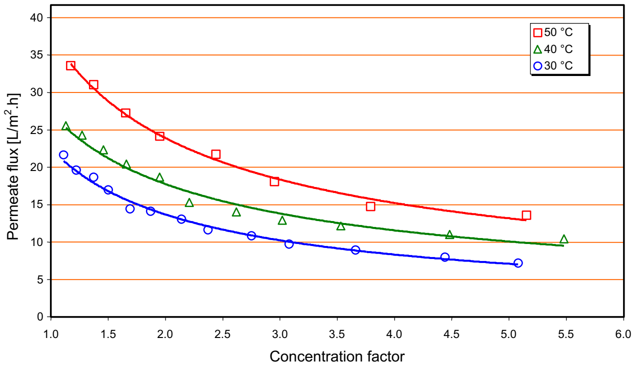

Fig. 2. Milk concentration by UF/M: influence of temperature and concentration factor on the permeate flux (FS10 membrane, 50 bar, 400 l/h flow rate).

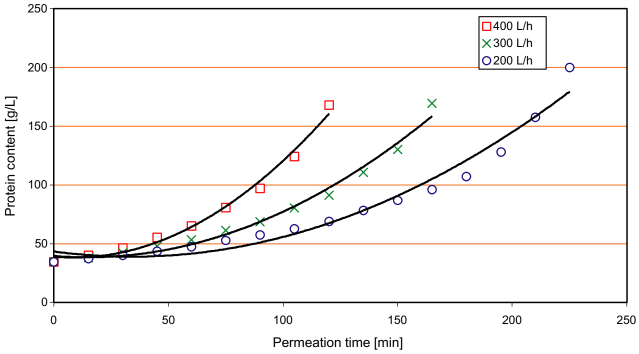

Fig. 3. Milk concentration by UF/M: influence of flow rate on the protein content in the retentate (FS10 membrane, 50 °C, 5 bar).

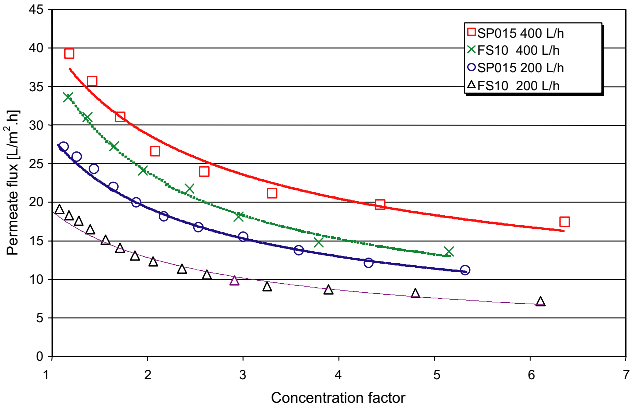

Fig. 4. Milk concentration by UF/M: influence of pore size of membranes on the permeate flux (5 bar, 50 °C).

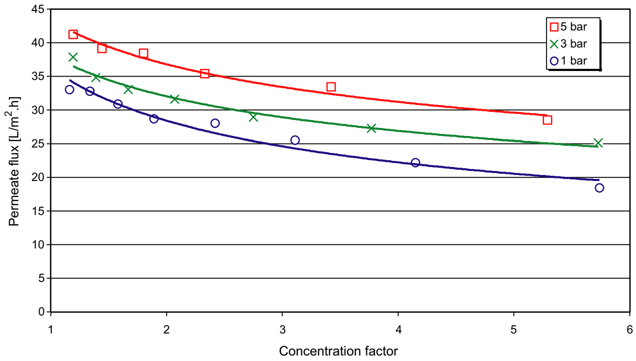

Fig. 5. Whey concentration by UF/W: influence of pressure and concentration factor on the permeate flux (FS10 membrane, 50 °C, 400 l/h flow rate).

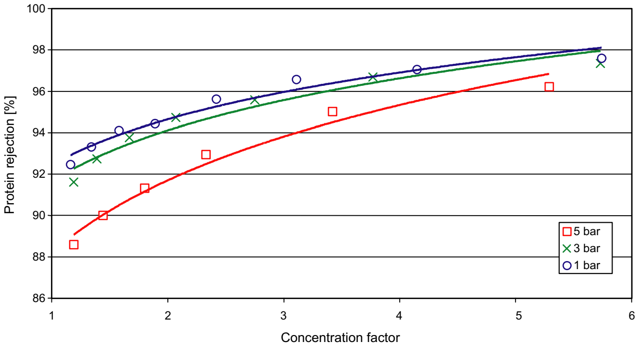

Fig. 6. Whey concentration by UF/W: influence of pressure and concentration factor on protein rejection (FS10 membrane, 50 °C, 400 l/h flow rate).

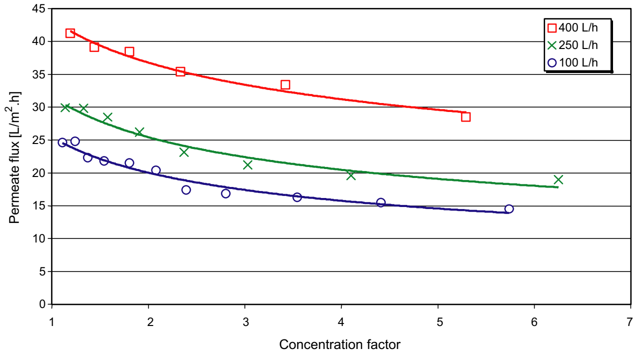

Fig. 7. Whey concentration by UF/W: influence of flow rate and concentration factor on the permeate flux (FS10 membrane, 50 °C, 400 l/h flow rate).

be demonstrated later in the case of the ultrafiltered milk, there is the danger of damaging the fat globules by excessive pumping.

The higher operating temperature resulted in an increase in the permeate flux (Fig. 8) until 50 °C, where the viscosity of the processed whey reaches its minimum value and hereafter temperature increase can cause heat denaturation of the whey proteins.

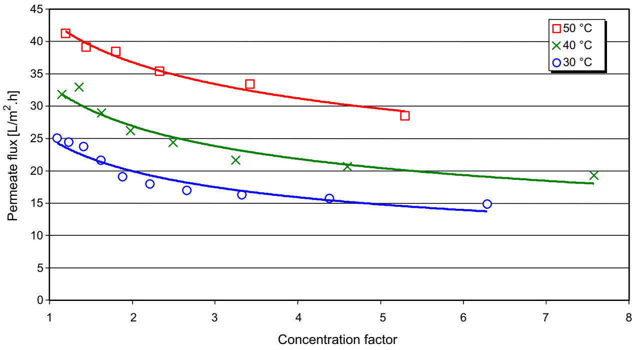

Fig. 8. Whey concentration by UF/W: influence of temperature and concentration factor on the permeate flux (FS10 membrane, 50 bar, 400 l/h flow rate).

3.3. *Concentration of ultrafiltration permeate by nanofiltration (NF)*

The nanofiltration membrane was used for the recovery of lactose from the milk permeate (UF/M) and whey permeate (UF/W). The nanofiltration membranes have lower molecular weight cut-off (Table 2), thus they reject the lactose molecules, which are smaller than proteins.

In the case of NF higher transmembrane pressure has to be applied, in our case it was 10–20 bar.

Changes in flow velocity and pressure affect the nanofiltration permeate in a similar manner to the ultrafiltration of light milk and whey. As the transmembrane pressure was increased from its lowest value, the NF permeate flux increased until a maximum, where further pressure increases had no advantage. Higher pressure increases the permeate flux; the concentration of the lactose on the membrane surface increases, nullifying the effect of the additional pressure. The point at which the flux becomes independent of pressure is related to the velocity of the feed stream over the membrane, the higher velocities making it profitable to use higher pressure and obtain higher fluxes (Atra, 2000).

The influences of the transmembrane pressure and the concentration factor on the permeate flux are shown in Fig. 9. From this diagram it is obvious, that there is a

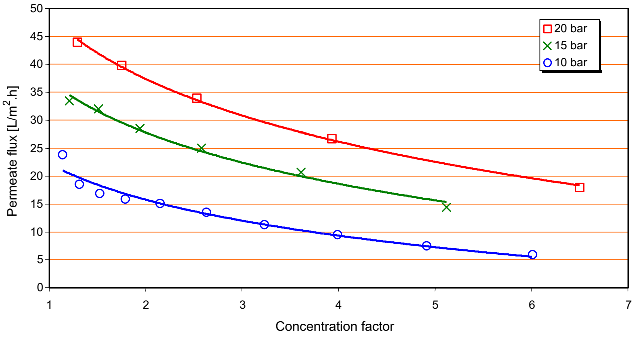

Fig. 9. UF permeates concentration by NF: influence of pressure and concentration factor on permeate flux of UF/M + UF/W permeates (RA55 membrane, 50 °C, 200 l/h flow rate).

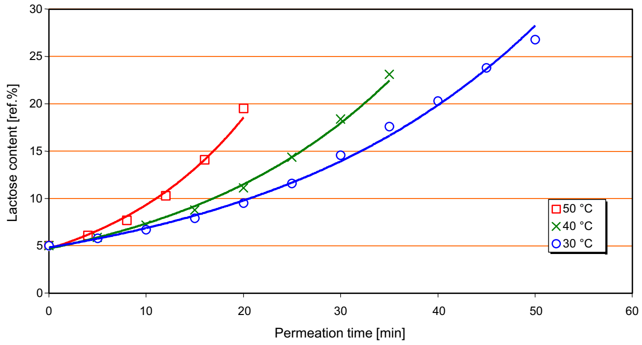

Fig. 10. UF permeates concentration by NF: influence of temperature on lactose content in the retentate (RA55 membrane, 20 bar, 200 l/h flow rate).

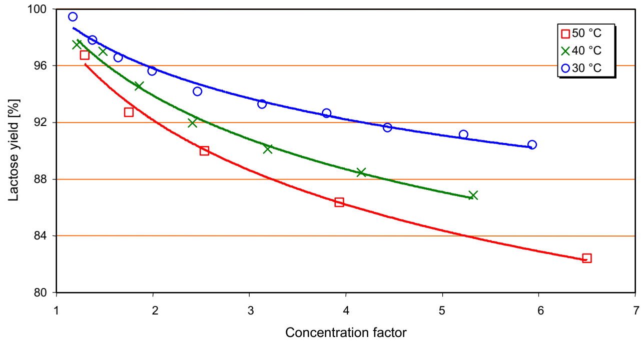

Fig. 11. UF permeates concentration by NF: influence of temperature and concentration factor on lactose yield (RA55 membrane, 20 bar, 200 l/h flow rate).

significant influence of the pressure on the permeate flux in the investigated range (10–20 bar), while the permeate flux decreases with an increase in the concentration factor, due to increase in the osmotic pressure of the retentate.

The lactose content increased with the operating temperature (Fig. 10), with rate of increase steeper at higher temperatures. Increase in temperature has a dual effect of lowering the viscosity, which assists permeate flow rate, and of increasing diffusivity, which assists

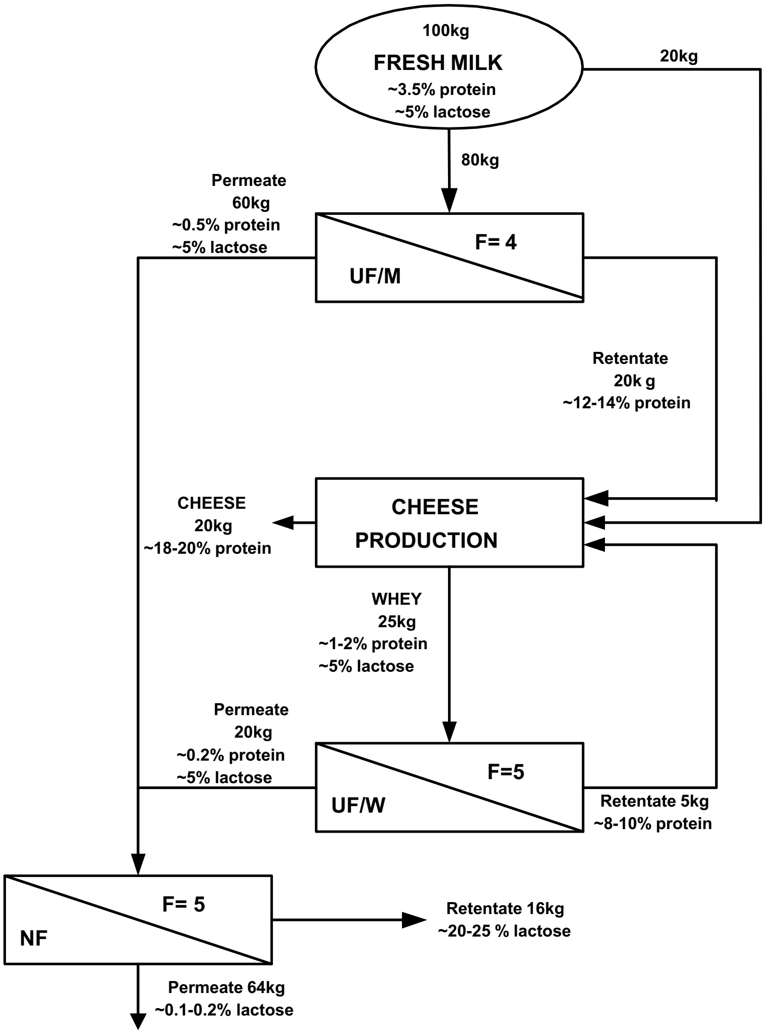

Fig. 12. Mass balance of the proposed process-combination.

dispersion of the polarized layer. It is always useful to quote fluxes at a standard temperature. Use of high temperature is limited by the properties of the nanofiltered solution and of the investigated membranes.

Fig. 11 shows the effect of operating temperature on the lactose yield which decreases with an increase in the temperature, as the result of lower lactose retention of the investigated membrane at higher temperature (Atra, 2000).

3.4. Combined process for a more economic cheese production

The concentration by membranes is a cold process, which preserves the nutritional value of the materials, and which is more economical than the traditional concentration processes (evaporation).

In this study the basic principles (flow diagram) of a combined cheese-making process is proposed (Fig. 12). The main target was the whey utilization. The UF-concentrated fresh milk with 12–14% protein and the UF-concentrated whey with 8–10% protein can be introduced into the cheese production, which should improve the nutritional value of the cheese and increase the economic effect. At the same time the UF permeates with ~0.1–0.5% protein and ~5% lactose can be fed to a NF.

The lactose concentrate (with 20–25% of lactose) can be applied in the sweets industry, while the permeate—with 0.1–0.3% lactose—can be reused in the production or for other purposes, representing a "cleaner technology".

The flow diagram was developed on the basis of the results of laboratory membrane filtration experiments, and the possibility or feasibility for real cheese-making process can be improved by further laboratory experiments including cheese production on laboratory scale. It will be the next step of our investigations.

4. Conclusions

From our experiments it can be concluded that the concentration of the light milk, concentration and incorporation of the whey protein by ultrafiltration, and also the concentration and utilization the ultrafiltration permeates by nanofiltration can be successfully achieved by the investigated membranes (FS10, SP015 and RA55) with a high efficiency.

- The protein rejection of UF membranes reached 92–98%, while the permeate flux was acceptable, 30 l/(m^2h), using low pressure (~3 bar).
- For milk and whey proteins the suitable temperature of UF is ~50 °C, where the viscosity of the solutes has a low value. Further increase in the temperatureis limited, because of decomposition of the proteins and damage of membrane material.
- The experimental results using NF membrane were positive, the lactose concentration was higher than 25 ref%, while the permeate flux reached $\geqslant 40$ l/(m^2h) using 20 bar pressure.
- Choosing the proper operation parameters for NF (30 °C, $F=5$ concentration factor) $Y>90\%$ lactose yield could be achieved.
- The permeate water of NF contains only 0.1–0.3% lactose, which makes possible the recycle and reuse of the permeate for other purposes (cleaning, irrigation) or at least can be directly discharged into sewer.
- In this study a flow diagram for combined cheese-making process was proposed with an increased cheese yield constituting the base of an environmental friendly cleaner technology.

Acknowledgment

This work was partially funded by the Hungarian Ministry of Culture and Education, the Syrian Embassy in Hungary and the Hungarian Scientific Foundation (OTKA T 037848 and T 031975).

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