Desalination and Water Treatment
www.deswater.com

1944-3994/1944-3986 © 2013 Desalination Publications. All rights reserved
doi: 10.1080/19443994.2012.693713

51 (2013) 786–799
January

Taylor & Francis
Taylor & Francis Group

A novel implementation of water recovery from whey: "forward–reverse osmosis" integrated membrane system

Coskun Aydiner*, Semra Topcu, Caner Tortop, Ferihan Kuvvet, Didem Ekinci, Nadir Dizge, Bulent Keskinler

Department of Environmental Engineering, Faculty of Engineering, Gebze Institute of Technology, 41400 Kocaeli, Turkey
Tel. +90 262 6053220; Fax: +90 262 6053205; email: aydiner@gyte.edu.tr

Received 28 February 2012; Accepted 10 May 2012

ABSTRACT

As a result of its emerging contribution to water recovery and clean water production, forward osmosis (FO) in integrated membrane system has recently especially been preferred by research communities on membrane science and desalination technology. In this study, the effectiveness of FO reverse osmosis (RO) integrated membrane system in whey dewatering was investigated in laboratory scale experiments in which FO and RO were utilized for whey concentration and water recovery, respectively. FO experiments were carried out at different conditions of cross-flow rate, temperature, membrane kind, membrane orientation mode, and microfiltration (MF) pretreatment. A single-step RO system was applied for water recovery from the FO draw solution. In the FO process, about 1.6 L water of 3 L whey was withdrawn into 3 M NaCl draw solution during 6 h operating time, and a sufficiently high performance in whey concentration was obtained, with the solid content being increased from 6.8 to 14.3%. However, the process resulted in a high salt permeation into the whey, in addition to some soluble organics being permeated into the draw. RO process are operated with relatively low performances due to excessive salt concentration of the FO draw solutions, which indicates that there is a need for RO implementation in two or three sequential levels for achieving an absolute success in the water recovery from whey. Despite the fact that MF pretreatment to some extent decreased the FO performance, it could be used for directly productive activities intended to recover fats from whey. Results have proved that prior to whey powder production, the integrated system could be effectively employed for whey concentrations up to a solid content of 25–35%. Accordingly, FO–RO system can be utilized as a novel alternative in concentrating whey compared to ultrafiltration-RO combined system widely used worldwide. However, before practical implementation of the system, an optimization between alleviating the salt concentration in FO draw and multi-step RO implementation should have to be considered concurrently with the economics of the investment.

Keywords: Forward osmosis; Reverse osmosis; Integrated membrane system; Whey; Water recovery

1. Introduction

Forward osmosis (FO) is an osmotically-driven membrane process that works by spontaneous osmosis between two solution phases of separation and purification of waste streams [1]. In this process, water

---

*Corresponding author.

Presented at the International Conference on Desalination for the Environment, Clean Water and Energy, European Desalination Society, 23–26 April 2012, Barcelona, Spain

flows across a semi-permeable membrane from the solution having low concentration (feed solution) to the solution having high concentration (draw solution), due to the osmotic pressure difference. This phenomenon brings about the use of a concentrated draw solution in a controlled FO process so as to provide a high osmotic pressure difference, enabling water transport through the membrane. The process results in a diluted draw solution, which needs to be concentrated both for reuse in FO system and to produce clean water, so that it necessitates a complementary treatment with another process featuring an integrated treatment technology. Hence, FO process as a pretreatment step in the combined treatment for the disposal or recovery of waste content is accompanied by processes such as reverse osmosis (RO), membrane distillation, membrane osmotic distillation, decomposition with heating, and magnetic separation [2–6]. The water gained from the whole system is of high quality and with nearly complete retention of microorganisms, particulates, and organic matter as well as significant retention of dissolved salts [7–9].

FO is superior to the pressure-driven membrane process. One of its main advantages is the use of limited external energy to separate solutes and solvent from each other. Another advantage is that a draw solution having relatively high osmotic pressure can be readily supplied from renewable sources such as seawater or concentrated brackish water, etc. [1]. The cost of the process is remarkably lower as there is no hydraulic pressure operation involved, nearly complete rejection of contaminants, potentially low membrane fouling tendency, and more environmentally friendly desalination technology [10,11]. However, there are still a number of technical barriers in using this in large-scale industrial applications. One of the major drawbacks is the lack of an optimized membrane that can produce a high flux comparable to commercial RO membranes; research is being undertaken on this subject [10]. Moreover, internal concentration polarization which significantly limits flux efficiency, and finding easily separable draw solutes are the other major challenges. In addition, the membrane flux can be adversely affected by membrane fouling [12,13]. In spite of all these constraints, the process pursues its development depending on desalination needs, as it is one of the extensively studied processes in separation science and technology in the recent years. The process can be successfully utilized in many applications such as electricity production [14–17], power generation [18–21], water or wastewater reclamation [7,8,22,23], seawater desalination or brine concentration [12,24], concentration of liquid foods [25–27], protein enrichment and concentration [28], and even for water purification and reuse in space [29,30]. Additionally, it may also be used as a promising alternative to decrease the energy usage of a dewatering process such as desalination and concentration processes [4,26,31]. In a detailed perspective, its combined application with RO process has drawn the attention of research communities towards membrane science and technology. Up to now, FO–RO integrated membrane system has been successfully tested for direct potable reuse of wastewater [2,32]. Some of the studies have proved that FO–RO is capable of treating low-quality waste streams such as landfill leachate and industrial wastes [33–35]. Taking into account all these features, one can conclude that FO process not implemented for concentrating whey liquors may be applied as an economical and viable pretreatment or dewatering method prior to whey powder production, concurrently with water recovery from whey by RO process.

Cheese whey is a by-product of the dairy industry and its production is estimated to be over 10^8 tonnes per year, yielding an important source of environmental pollution [36]. Approximately, 47% of the whey produced world-wide every year is disposed of in the environment, which leads to both serious environmental pollution problems and a significant loss of valuable products [37–40]. Due to its rich nutrient content, cheese whey has been used for the production of different chemicals such as organic acids (lactic, acetic), alcohol (ethanol), single cell protein, methane, and cheese whey powder [41]. About 50% of the cheese whey produced worldwide is now treated using different methods and transformed into various food products. It has been reported that the European Union uses about 45% of cheese whey directly in liquid form, 30% in the form of powdered cheese whey or cheese whey protein concentrates, and 15% as lactose [42].

Based on literature survey, no study has presented on whey concentration using a FO-included membrane system. In that sense, the main objective of this study was to investigate, using laboratory-scale research, the capabilities of both FO process in whey concentration and complementary RO process in clean water production from whey. For whey concentration, the influences of cross-flow rate, temperature, membrane kind, membrane orientation mode and microfiltration (MF) pretreatment on FO performance were particularly examined. The performance of the FO process was evaluated based on parameters such as the net osmotic pressure difference, volumetric water permeation, water and salt fluxes, and whey solid content. Besides, the performance of RO process in water recovery from FO draw solutions was determined based on water flux and rejections. Finally, the capability and competence of FO–RO integrated system on water recovery from whey concurrently with whey concentration were straightforwardly exposed with an intent directed to its industrial-scale implementation in dairy industry.

2. Materials and methods

2.1. Materials

FO experiments were conducted using flat sheet cellulose triacetate (CTA) FO membrane (Hydration Technologies Inc., OR). Four different membrane kinds with flat sheet form were tested in RO experiments as well as FO experiments of which CPA-3, LFC-3 and ESPA-2 membranes (Hydranautics Inc., CA) were made from composite polyamide, whereas GE-PA membrane was manufactured from polyamide (GE Osmonics, MN). PVDF membrane having a pore size of 0.45 μm (GE Osmonics, MN) was used in MF pretreatment of whey. Cheese whey was obtained from industrial facilities of Cayirova Milk&Milk Products Inc., located at Kocaeli, Turkey. The characteristics of raw whey and its liquor concentrated at the best conditions of FO experiments were given in Table 1.

2.2. Methods

2.2.1. Experimental procedures

FO experiments. A laboratory scale FO system shown in Fig. 1(a) was employed in the experiments. The cross-flow membrane module was a custom-made cell with equivalent flow channel at both sides of the membrane. Hydrodynamic flow at the channels was concurrently operated. The membrane module made from Delrin acetal resin material (DuPont, Delaware) has an effective membrane area of 140 cm^2. Two speed-controllable peristaltic pumps (EW 77111-67, Cole Parmer, IL) were used to pump the solutions. Two flow meters with a maximum flow rate of 10 L min^{-1} were separately placed on the feed and draw lines of the setup to enable the desired same flow rates on each line. The setup was also equipped with a constant-temperature water bath (462-7028, VWR Scientific, IL) to maintain the same temperature (25 ± 0.5 °C) at both the feed and draw solutions during FO tests.

In FO experiments, the influences of five operational parameters–cross-flow rate (150, 300, 450, and 600 L h^{-1}) (0.25, 0.50, 0.75, and 1.00 m s^{-1}), temperature (20, 25, 30, 35, and 40 °C), membrane kinds (CTA-FO, CPA-3, LFC-3, ESPA-2, and GE-PA), membrane orientation modes (normal and reverse), and MF pretreatment of whey–were explored to evaluate the process effectiveness. Normal orientation mode means that the active or selective layer of the membrane faced the draw solution while its support layer faced whey or feed; in reverse mode, the active and support layers of the membrane were in contact with the feed and the draw, respectively. The experiments including cross-flow rate and temperature parameters were executed in reverse mode of CTA-FO membrane for 2 h using 2 M NaCl draw solution. While temperature was kept constant as 25 ± 0.5 °C in the cross-flow rate experiments, the cross-flow rates at both flow channels were maintained at 300 L h^{-1} (0.50 m s^{-1}) in the temperature experiments. In the subsequent three experiments including membrane kinds, membrane orientation modes and MF pretreatment, 3 M NaCl as the draw was used to increase whey dewatering performance of FO system. These experiments were carried out at a cross-flow rate of 300 L h^{-1}, temperature of 25 ± 0.5 °C, and duration of 6 h. In all FO experiments, the system was employed with 3 L volumes for both the feed and draw sides. The process was conducted in the normal mode for the membrane kind experiments and in reverse mode of CTA-FO membrane for the MF pretreatment experiments.

MF and RO experiments. MF and RO experiments were executed using laboratory scale pressure-driven membrane system (Fig. 1(b)). The system had a flat-sheet cross-flow membrane module having an effective membrane area of 140 cm^2 (GE Osmonics, MN). It was equipped with a feed tank, high-pressure pump of 100 bar with a flow volume of 330 L h^{-1} (Bosch, Germany), flow splitter, digital flow meter (max 720 L h^{-1}) (Honsberg, Germany), and manual oil pump for clamping the module. In the experiments, the processes were employed in the concentration mode, which means that permeate solutions were not returned into the feed tank while the retentate was. The permeate solutions were collected in a beaker and flow rates were measured by an electronic balance (Precisa XT2220M-DR) and recorded by a computer. MF pretreatment of raw whey were carried out for 4.5 L feed volume along 180 min filtration time by maintaining a transmembrane pressure of 2 bar, cross-flow rate of 0.60 m s^{-1} and temperature of 25 ± 0.5 °C. The draw solutions obtained from the membrane kind experiments were filtrated by the reverse osmosis membranes. RO filtration experiments were executed under the conditions of 3 L draw solution, 120 min filtration time, 0.30 m s^{-1} cross-flow rate, 25 ± 0.5 °C temperature, and 25 bar transmembrane pressure.

Table 1
Characteristics of raw and FO concentrated whey

Parameter	Unit	Raw whey	FO[a] concentrated whey
pH	–	4.65 ± 0.19	4.62
Conductivity	$mS\,cm^{-1}$	6.91 ± 0.40	13.43
Cl^-	$mg\,L^{-1}$	932 ± 34	3,386
Osmolality	$mmol\,kg^{-1}$	334 ± 33	671
Density	$g\,cm^{-3}$	1.0242 ± 0.0016	1.0440
Freezing point	℃	−0.529 ± 0.039	−1.248
SCOD	$mg\,L^{-1}$	61,855 ± 6,792	125,151
STOC	$mg\,L^{-1}$	38,561 ± 2,242	67,350
NH_4–N	$mg\,L^{-1}$	154 ± 21	202
NO_2–N	$mg\,L^{-1}$	0.1 ± 0.1	0.5
NO_3–N	$mg\,L^{-1}$	257 ± 16	319
TKN	$mg\,L^{-1}$	1,362 ± 151	2,145
Org-N	$mg\,L^{-1}$	1,208 ± 162	1,943
TN	$mg\,L^{-1}$	1,619 ± 149	2,465
PO_4–P	$mg\,L^{-1}$	363 ± 23	635
TP	$mg\,L^{-1}$	436 ± 75	769
Total protein	%	2.46 ± 0.16	5.08
Fat	%	0.42 ± 0.15	0.79
SNF (fat-free dry matter)	%	6.69 ± 0.39	13.49
Total solid content	%	7.12 ± 0.52	14.28
Lactose	%	3.37 ± 0.22	6.52
Minerals	%	1.02 ± 0.09	1.84

[a] At the best experimental conditions–membrane type: FO membrane, operation mode: reverse mode, draw solution: 3 M NaCl, temperature: 25 ± 0.5℃, cross-flow rate: 300 $L\,h^{-1}$ (0.5 $m\,s^{-1}$), process time: 360 min, each volume of feed and draw solutions: 3 L.

2.2.2. Analytical procedure

Total protein, fat, fat-free dry matter (solids-not-fat [SNF]), total solids, lactose and minerals contents of whey samples were measured by Lactostar instrument equipped with thermal and optic sensors (Funke Gerber Company, Germany). The pH, conductivity, and temperature measurements were taken using WTW Multi 340i Meter (WTW, Weilheim, Germany). For density measurements in the samples, DA-130N density meter (KEM Co., Ltd., Kyoto, Japan) was used. Osmolalities of the samples were determined in duplicate for each data point by Advanced Osmometer instrument (Model 3250-Advanced Instruments Inc., USA) in accordance with freezing point depression method after completing the entire experiment. Water qualities in whey and draw solution were analyzed in accordance with standard methods [43]. Besides, nitrite, nitrate, and TOC, TN parameters were analyzed using GBC UV–visible Cintra 20 spectrometer (Cintra, Australia), and HACH IL 550 TOC–TN (Hach Lange Ltd., Germany) instruments, respectively.

2.2.3. Performances of processes

FO performance. The permeated water volume, V was determined from osmolality differences between the feed and draw solutions in the FO system. First, the osmolalities at both sides were individually measured at definite time intervals along the experiments, and thereafter V was determined from the differences in sequential time points in accordance with osmolarity-based mass balance calculation. The results were also made valid by verifying them with whey solid content (W_{SC}). The water flux, J_w was determined from the volume increase in the draw solution using Eq. (1) [44].

$$J_w = \frac{1}{A_m} \cdot \frac{\Delta V_t}{\Delta t} \tag{1}$$

where A_m is the membrane area, t the time, and V_t the volumetric water permeation at any time. The salt flux, J_s that flew in reverse direction from the water

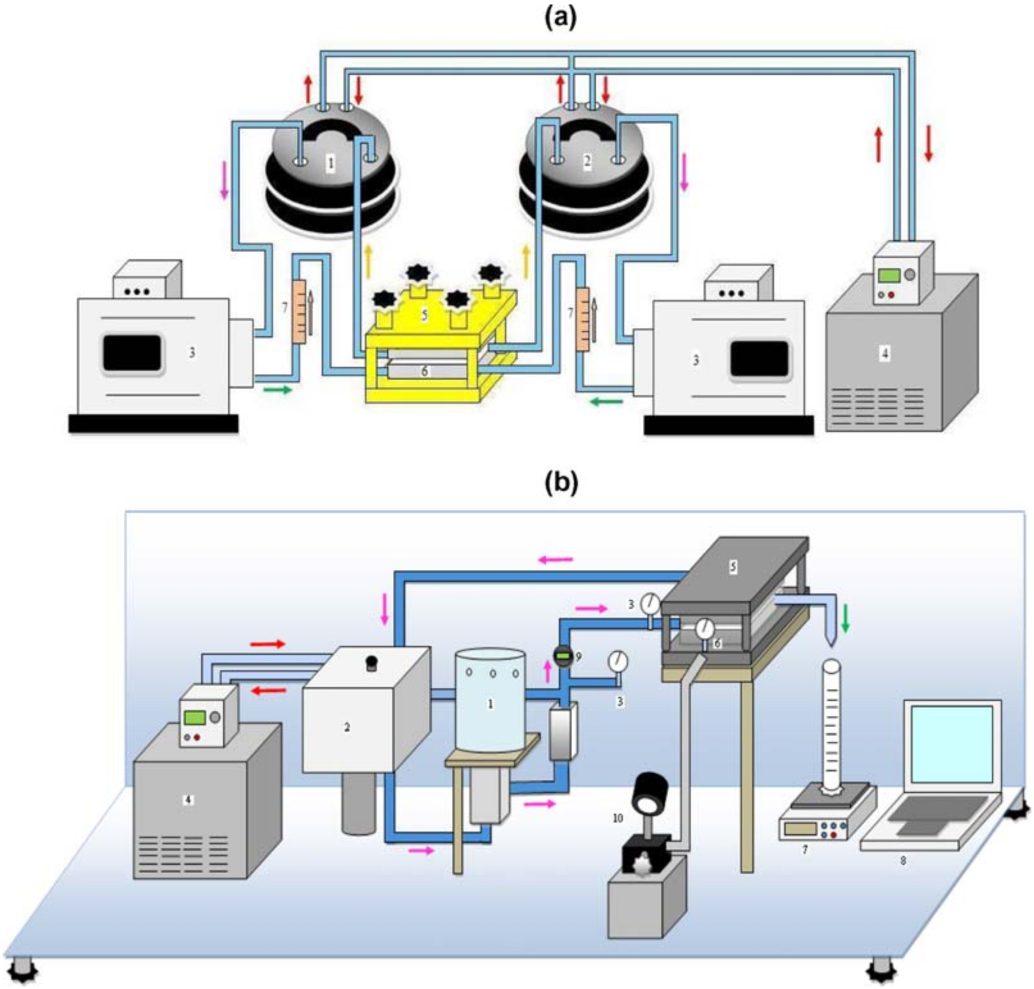

Fig. 1. Experimental membrane system setups (a) FO system (1: feed tank, 2: draw tank, 3: peristaltic pumps, 4: water bath, 5: clamp, 6: membrane module, 7: flow meters) and (b) RO system (1: high pressure pump, 2: feed tank, 3: pressure gauges, 4: water bath, 5: clamp, 6: membrane module, 7: balance, 8: computer, 9: digital flow meter, 10: clamp arm).

flux between both solutions was calculated using the following equation [44]:

$$J_s = \frac{1}{A_m} \times \frac{\Delta(C_t \cdot V_t)}{\Delta t} \tag{2}$$

where C_t is the salt concentration at any time. Net osmotic pressure difference ($\Delta\pi_{net}$), which refers to the osmotic pressure difference between the draw and the feed was determined from the difference in the osmotic pressures determined individually at both sides [44,45]:

$$\Delta\pi_{net} = \pi_{d,b} - \pi_{f,b} \tag{3}$$

where $\pi_{d,b}$ and $\pi_{f,b}$ are the osmotic pressures of the draw and the feed, respectively. The osmotic pressures of the solutions were calculated in accordance with the van't Hoff equation.

$$\pi = R \cdot T \cdot [m \cdot d] \tag{4}$$

where R is the ideal gas constant (8.314 J K^{-1}mol^{-1}), T the absolute temperature (K), m the osmolality (mosm kg^{-1}), and d the solution density (kg L^{-1}).

MF and RO performances. In pressure-driven membrane filtration experiments, the permeate fluxes were determined from the water permeation performance of each membrane process using Eq. (1). The percentage rejection rates of contaminants, R_i were calculated by Eq. (5).

$$R_i\ (\%) = 100 \times \left(1 - \frac{C_{p,i}}{C_{f,i}}\right) \tag{5}$$

where $C_{p,i}$ and $C_{f,i}$ are the values of contaminant i measured in the permeate and the feed, respectively. It should be noted that whey as the feed is used for MF process; while the draw solution from the FO system is the feed solution for RO process.

3. Results and discussion

3.1. FO experiments influences of FO operating parameters

In FO experiments, whey concentration by FO was broadly investigated in light of four system parameters: cross-flow rate, temperature, membrane kinds, and operation modes. The performance results comprising of net osmotic pressure difference, volumetric water permeation, water and salt fluxes, and total solid content of whey are presented in the subsequent subsections.

3.1.1. Cross-flow rate

The water permeation and whey concentration performances for cross-flow rates are depicted in Fig. 2. The net driven force is a function of water volume passed into the draw, whose influence on water passage from the feed into the draw decreased as the permeated volume increased due to dilutive operation preferred for the draw solution in FO process. The pressure differences of 95–96 bar at the beginning of the experiments reduced to 74–83 bar due to the transferred water of 385–480 mL.

Fig. 2 also indicated that water volume that passed into the draw somewhat increased as the cross-flow rate was increased. In the process, despite varying flow rates, water fluxese ranged between 10.6 and 14.3 $\mathrm{L\,m^{-2}\,h^{-1}}$. The highest water flux at the end of 2 h process time was observed at cross-flow rate of 600 $\mathrm{L\,h^{-1}}$. A similar trend was observed in salt flux variations among 150 and 450 $\mathrm{L\,h^{-1}}$, a distinctive difference appeared at 600 $\mathrm{L\,h^{-1}}$. After the turning point of about 0.25 h, salt flux at 600 $\mathrm{L\,h^{-1}}$ reached a steady state at the end of the process, while the others were decreasing after their turning points at around 1 and 1.5 h. The lowest and highest final salt fluxes were determined at 150 and 450 $\mathrm{L\,h^{-1}}$ flow rates with 0.52 and 5.38 $\mathrm{g\,m^{-2}\,h^{-1}}$, respectively. Whey was concentrated depending on water permeation, which increased with cross-flow rate, and its solid content rose to the levels of 7.23, 8.30, 8.50 and 8.61% in an order of the increasing flow rate.

3.1.2. Temperature

The influences of varying temperatures at both solutions on the performance of FO system can be seen from Fig. 3.

As the temperature increased from 20 to 35°C, the net pressure difference decreased from the levels of 95–100 to 72–84 bar. However, its highest decline was seen at 40°C. The permeated water increased to some extent from 435 to 546 mL with the increase in temperature from 20 to 35°C; it resulted in 535 mL at 40°C. This meant that the increased temperature from 35 to 40°C accounted for a fall in effective osmotic pressure difference inside the membrane as an inherent result of higher internal concentration polarization. The variations in final water fluxes with temperature which was determined between 10.0 and 16.9 $\mathrm{L\,m^{-2}\,h^{-1}}$ seemed to be the same as those of the permeated water volumes. Despite similar variation trends at

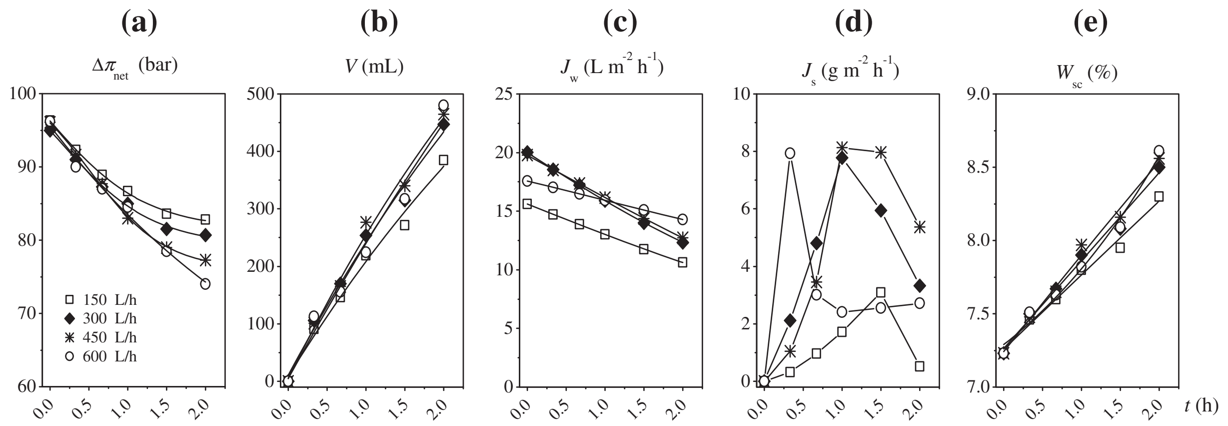

Fig. 2. Water permeation and whey concentration performances in reverse mode FO process for various cross-flow rates: (a) net osmotic pressure difference; (b) volumetric water permeation; (c) water flux; (d) salt flux; and (e) total solid content of whey.

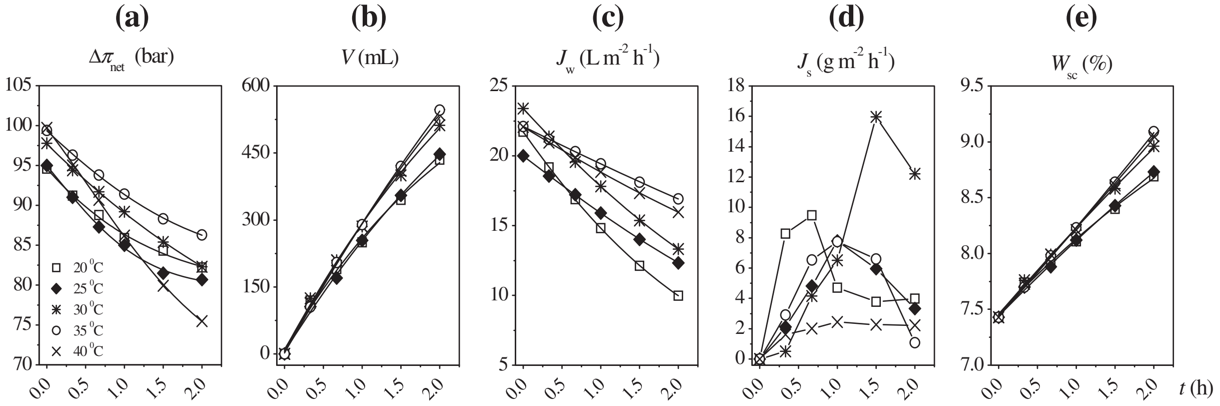

Fig. 3. Water permeation and whey concentration performances in reverse mode FO process under various temperatures: (a) net osmotic pressure difference; (b) volumetric water permeation; (c) water flux; (d) salt flux; and (e) total solid content of whey.

turning points 0.5 and 1.5 h for salt fluxes, high salt passage of 12.2 g m^{-2} h^{-1} into whey was observed at 30°C. The process exhibited a good performance at 35°C, with a salt flux of 1.1 g m^{-2} h^{-1} compared to other temperatures. The proportions of 14.5 and 18.2 of water in whey were withdrawn into the draw solution at 20 and 35°C, respectively. Whey was concentrated to the levels of 8.7–9.1%, which were as low as in the cross-flow rate experiments. The results so far show that the performances of water and salt fluxes needs to improve concurrently, keeping the flow rate and temperature within reasonable limits so as to prevent further energy consumptions, a practical implementation of FO. In this regard, for all subsequent experiments, FO was employed at a cross-flow rate of 300 L h^{-1} and temperature of 25 ± 0.5°C. In addition, the process time was increased to 6 h so as increase the whey solid content, and 3 M NaCl draw solution was used instead of 2 M used up to now to increase the water flux performance.

3.1.3. Membrane kind

In the membrane kind experiments, the process was operated in normal orientation mode so as to take into account its possible effects on increasing the performance. The results are depicted in Fig. 4.

FO process revealed discrete behaviors of membrane kinds for all performance parameters in which FO membrane definitely exhibited a clear superiority over all the other RO membranes. Despite operating with higher driven forces compared to 2 M NaCl from about 165 bar in the beginning to 138–162 bar in the end, the process could be employed with an inadequate performance even for FO membrane that 382 mL of water in whey was transferred to the draw with the final water and salt fluxes of 1.23 L m^{-2} h^{-1} and 0.08 g m^{-2} h^{-1}, respectively. The lowest water permeation of 108 mL resulted from ESPA-2 membrane, which is nearly a quarter of that of FO membrane. Although the evolutions for the permeation and flux rates of water were similar in characteristics for the various membrane kinds, distinctive variations in salt fluxes of the membranes were observed. However, final salt fluxes of the membranes were determined in a quite low range of 0.02–0.1 g m^{-2} h^{-1}. While about 12.7% of water in whey was withdrawn into the draw solution at the end of 6 h by FO membrane, this value was designated as 9.1, 5.3, 3.6, and 5.0% for RO membranes comprising CPA-3, LFC-3, ESPA-2, and GE-PA, respectively. On the other hand, a sufficient concentration of whey was not ensured as the solid content reached 8.4% after treatment.

3.1.4. Operation mode

The operation mode experiments were carried out using FO and CPA-3 membranes with both reverse and normal orientationsas they performed better than others. Similar experiments were conducted by Aydiner et al. [46], except that they considered the interrelationship between the performance and membrane fouling behaviors to be independent of membrane type and operating mode in organic-inorganic binary FO system [46]. In that study, two different approaches grounded on modeling- and *ex-situ* membrane characterization-based performance analysis were applied for the evaluation of performance-fouling relationships depending on a general performance evaluation. On the other hand, by a thorough evaluation of the results, this study intended to assess the

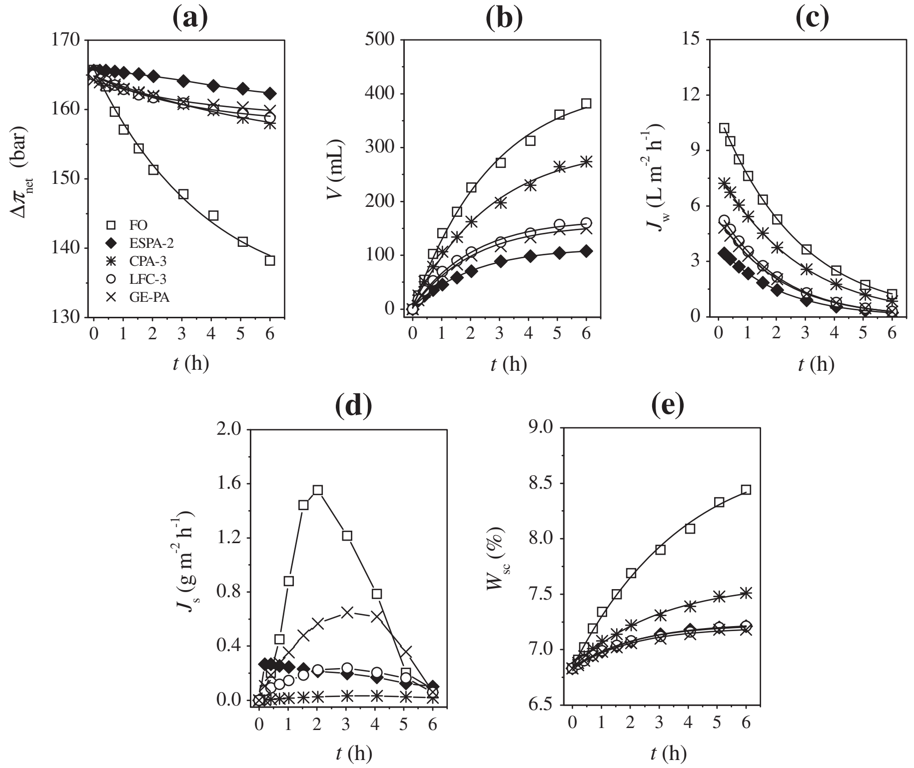

Fig. 4. Water permeation and whey concentration performances in normal mode FO process for different membranes: (a) net osmotic pressure difference; (b) volumetric water permeation; (c) water flux; (d) salt flux; and (e) total solid content of whey.

feasibility of FO process as a novel implementation combined with RO process in whey concentration.

As can be seen from Fig. 5, the FO membrane presented too high a water transport performance in reverse operation mode obtained so far. In spite of decrease in net driven force from 165 to about 84 bar, a high final water flux of 14.34 L m^{-2} h^{-1} was established. About 1.6 L water of 3 L whey was taken into 3 M NaCl draw solution within 6 h operating time, and a sufficiently high performance for whey concentration was obtained with a noticeable increase in whey solid content from about 6.8 to 14.3%. However, the process resulted in a high final salt permeation of 4.8 g/m^2 h into the whey; moreover, it did not reach a steady state at the end of 6 h whey processing in FO reverse mode. Against higher salt passages less internal membrane fouling that originated from both organic and inorganic solutes had taken place than that of RO membrane [46]. Neither FO membrane in normal mode nor CPA-3 membrane in both modes yielded enough good performance when compared to reverse operation of FO membrane. Besides, as indicated in Table 2 for the water qualities in the draw solution after FO processing, any organic carbon leakages into the draw were not observed in reverse operation modes of both membranes. In addition, the passages in small varying amounts of nitrogen and phosphorous were determined for all operations of the membranes, except for TP in normal mode of CPA-3. Low contamination of whey constituents in the draw in reverse mode provided meant that FO process can be executed for higher concentrations until the desired higher solid content is achieved.

3.2. Water recovery by RO

A single-step RO process was implemented for water recovery from the draw solutions at operation-mode experiments. Depending on the predominant effect of high salt concentration on RO membrane, the process could be operated only with a permeate flux under 5 L/m^2 h, and rejection rates of 63–81, 75–90, 47–96, and 19–27% for KOI, N, P, and Cl$^-$, respectively (Table 2). These results put forward that an additional RO operation is needed at least in second-step implementation, for achieving an absolute success on the basis of water recovery, especially in terms of the removal of dissolved salt ions. Very low permeate fluxes from the first step RO of FO draw solutions prevented to maintain sequencing lab-scale RO opera-

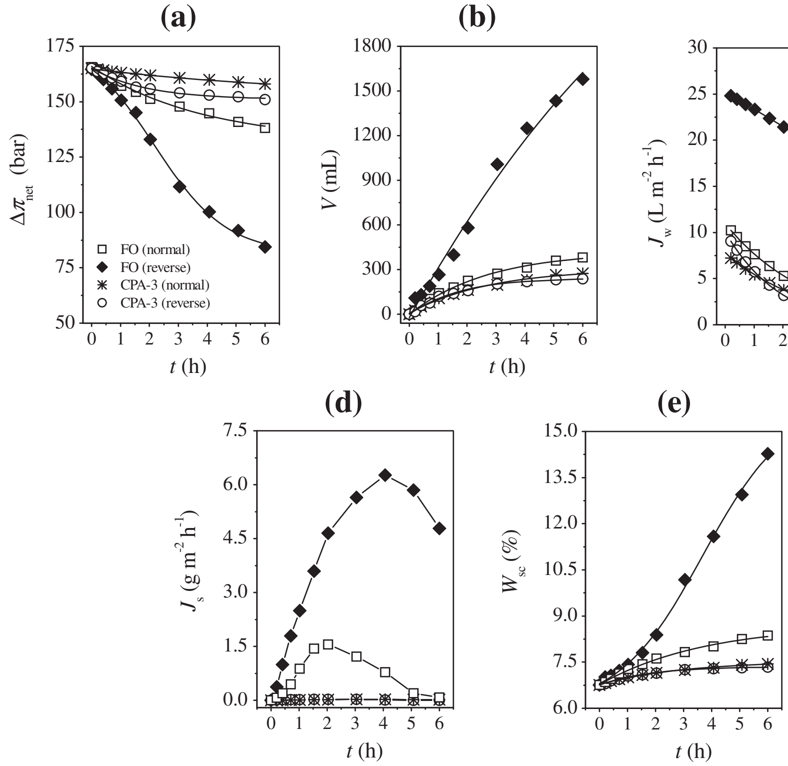

Fig. 5. Performance comparisons of FO processes operated at normal and reverse modes: (a) net osmotic pressure difference; (b) volumetric water permeation; (c) water flux; (d) salt flux; and (e) total solid content of whey.

tions, which means that the first step RO should have a higher scale such as a pilot plant to obtain the permeate volume required for the last-step RO in terms of completely testing the quality of product water. On the other hand it can be suggested that to remove dissolved salt ions more effectively, single-step membrane distillation (MD) can be utilized instead of multi-step RO processes for water recovery from the draw solutions. At this point, it should be emphasized that the assessment of capabilities of both membrane systems (FO–RO and FO–MD) and the selection of their appropriate one in whey concentration should be comprehensively taken into consideration on the basis of total economics including capital and operating costs, concurrently with the efficacy of whey powder production.

3.3. *MF pretreatment of whey*

Prior to FO run, MF processing of raw whey was conducted to compare the effectiveness of MF pretreatment with direct whey processing by FO. The performance results of the MF pretreatment and the direct FO processing and MF-FO system are shown in Table 3 and Fig. 6, respectively. In addition, time evolutions of whey contents concentrated by MF, FO and MF-FO are presented in Fig. 7.

Fig. 6 indicates that the direct FO processing resulted in higher water permeation than MF–FO system. FO water flux was determined as 7.3 and 14.3 L/m^2 h with or without MF pretreatment, respectively. However, any difference in salt flux variation for both processing events was not observed. In MF pretreatment, fat content of whey was completely not considered, while protein and lactose content was taken as about 16% (Table 3). It was ascertained that MF process somewhat decreased the FO performance. Also the whey concentration using both FO and MF–FO treatments did not reach to steady-state at the end of 6 h. The direct FO was more effectively employed to concentrate whey in spite of relatively lower net osmotic pressure difference in which whey was concentrated to a solid content of 14.3%. The solid content of whey pre-concentrated by MF pretreatment from a starting value of 6.8% to the final value of 7.9% could be raised to a lower level of 12.9% than the direct FO. The FO processing proved that whey concentration using the direct FO could be effectively improved to an order of 25–35% of solid content by a continuous operation. It was comprehended as another choice

Table 2
Water qualities for draw solutions obtained from different FO operation modes and their RO desalination performances

Parameter[a]	Unit	FO normal mode						FO reverse mode			
		FO membrane			CPA-3 membrane			FO membrane			CPA-3[c]
		FO draw	RO permeate[b]	*R* (%)	FO draw	RO permeate	*R* (%)	FO draw	RO permeate	*R* (%)	FO draw
Water flux[a]	$L\,m^{-2}\,h^{-1}$	–	0.00[b]	–	–	4.47	–	–	2.09	–	–
pH	–	5.4	6.1	–	6.8	7.2	–	4.6	4.2	–	6.1
Conductivity[a]	$mS\,cm^{-1}$	178.1	141.0	20.9	193.7	153.4	20.8	155.6	134.0	13.9	191.6
Cl^-[a]	$mg\,L^{-1}$	77,276	56,901	26.4	84,574	61,531	27.2	64,880	52,443	19.2	88,373
Osmolality[a]	$mmol\,kg^{-1}$	5,530	4,663	15.7	6,033	5,200	13.8	3,792	2,948	22.3	5,831
COD	$mg\,L^{-1}$	1,038	195	81.2	1,869	699	62.6	0.0	0.00	–	0.0
TOC	$mg\,L^{-1}$	69.2	56.2	18.8	50.5	31.6	37.4	0.0	0.0	–	0.0
TKN	$mg\,L^{-1}$	3.7	0.9	74.6	5.0	0.5	90.3	14.2	2.4	83.0	1.6
TN	$mg\,L^{-1}$	3.7	0.9	74.6	5.0	0.5	90.3	14.2	2.4	83.0	1.6
TP	$mg\,L^{-1}$	0.6	0.3	54.0	0.0	0.0	-	47.0	1.7	96.4	2.8

[a] 0.5 M NaCl solution was also purified by RO process to compare the effectiveness of RO filtrations of FO draw solutions. Water flux of the solution was determined as 43.62 $L\,m^{-2}\,h^{-1}$, while conductivity, Cl, and osmolality in the feed and permeate were determined as 42,900 and 132 $mS\,cm^{-1}$, 17,725 and 55 $mg\,L^{-1}$, and 882 and 16 $mmol\,kg^{-1}$, respectively, of which rejection rates were about 99.7, 99.7 and 98.2%, respectively.

[B] As the draw solution of FO process in normal operation mode used in the FO operation completely clogged the pores of the RO membrane at about 45 min of the experiment, the results are the values measured in the sample taken at that time.

[C] As there was no water permeation in the RO experiment, data for the draw solution of FO process in reverse operation mode which used CPA-3 membrane could not be obtained.

Table 3
Performance of MF process in whey pretreatment

Parameter	Unit	Raw whey	Concentrated whey	Permeate stream	R (%)
Water flux	$\mathrm{L\,m^{-2}\,h^{-1}}$	–	–	9.12	–
Density	$\mathrm{g\,cm^{-3}}$	1.0267	1.0272	1.0231	0.4
Freezing point	℃	−0.643	−0.652	−0.580	9.8
Osmolality	$\mathrm{mmol\,kg^{-1}}$	361	366	326	9.8
Total protein	%	2.38	2.67	2.25	15.8
Fat	%	0.37	0.49	0.00	100.0
Fat-free dry matter	%	6.39	7.42	6.27	15.4
Total solid content	%	6.76	7.91	6.27	20.7
Lactose	%	3. 05	3.32	2.80	15.8
Minerals	%	0.99	1.02	0.97	5.0

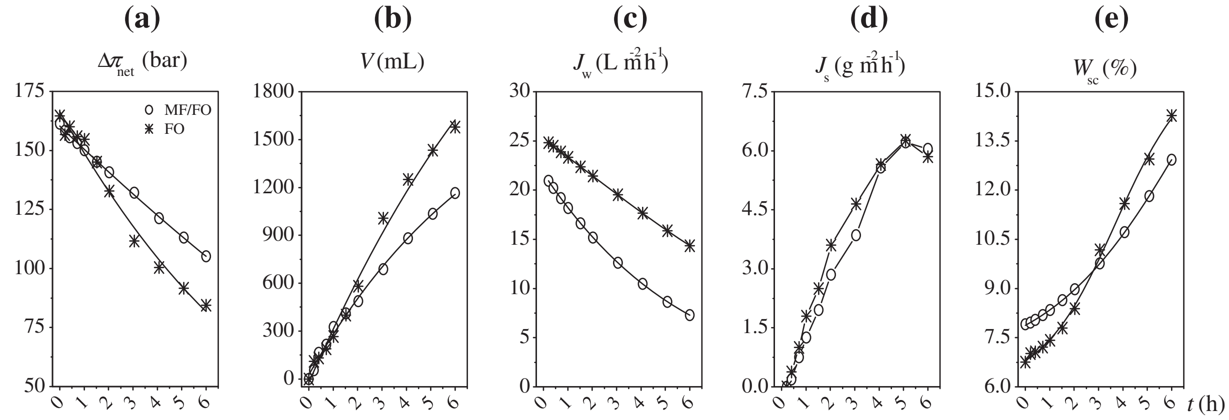

Fig. 6. Influence of MF pretreatment on FO performance in whey concentration: (a) net osmotic pressure difference; (b) volumetric water permeation; (c) water flux; (d) salt flux; and (e) total solid content of whey.

that MF pretreatment could be directly used for additional productive activities intended to recover fats from raw whey by producing cream or butter foods, as well as whey powder production by subsequent treatments including FO processing and thereafter using various drying technologies such as spray drying.

3.4. *Proposed integrated membrane system*

Whey consists of rather valuable food sources such as fats, proteins, lactose, and minerals. So, a lot of researchhas been ongoing on the fractionation of whey contents that are widely used as food additives and nutrition materials. From the results obtained in this study, an integrated membrane system can be proposed for the recycling and reuse of water from whey (Fig. 8). The integrated system involves two or three sequential processing steps: (1) preferentially pre-concentration of whey by MF to recover fats, (2) concentration of the pre-concentrated whey by FO process, and (3) clean water production from the water withdrawn into the FO draw solution by RO process.

According to the results, FO–RO system could be successfully utilized for whey concentration with a performance up to the whey solid content of 25–35%. However, full-scale adaptation to the conditions of continuous operation needs to use sequencing RO treatments both to properly recover the whey water and to permanently concentrate the diluted draw solution in the closed loop in the system. Moreover, as in the practices commonly applied in a conventional production system on whey products, the proposed system can be readily incorporated into a more comprehensive application oriented to both water and valuable product recovery from whey by which the

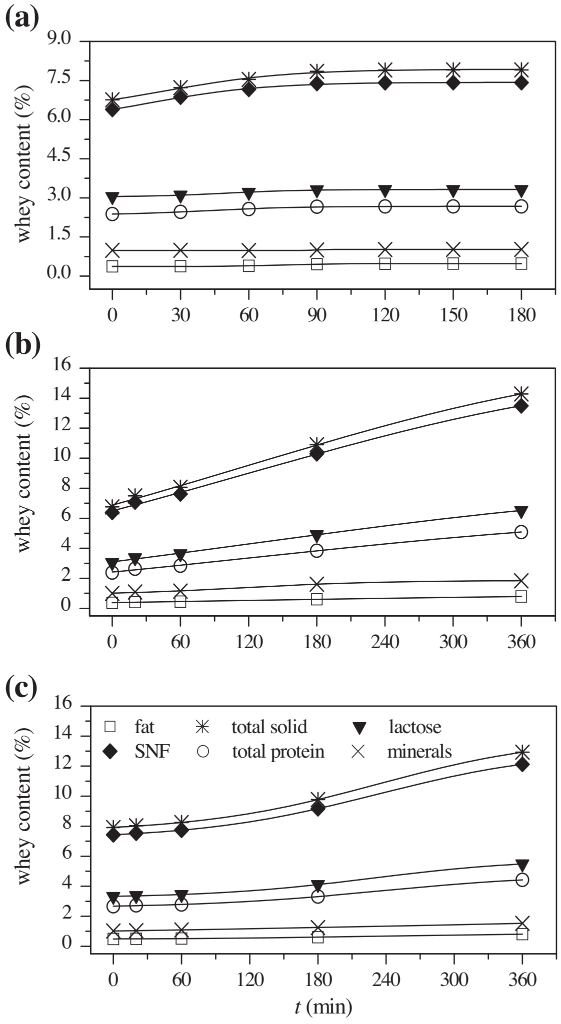

Fig. 7. Time evolutions of MF (a), FO (b), and MF-FO (c) concentrated whey contents (total protein, fat, SNF, total solid content, lactose, and minerals).

FO concentrated whey is thereafter handled for whey powder production using various drying technologies such as spray drying.

4. Conclusion

The technological efficacy of FO–RO integrated membrane system in water recovery from whey has been explored in which the capabilities and competences of FO and RO processes were elaborately investigated for whey concentration and clean water production, respectively. The prominent findings obtained from the study are presented below:

(1) For whey concentration, FO process was operated with a sufficiently high performance that resulted in an increase in whey solid content from 6.8 to 14.3% by employing FO membrane in reverse mode. However, it was observed that the process performance did not reach steady-state at the end of 6 h operating time. This meant that whey could be concentrated up to 25–35% of solid content, with longer operating times of 15–20 h. However, FO processing should be carried out using a draw solution including NaCl concentration less than 3 M to prevent high salt leakage into whey.

(2) For water recovery, RO process was used to produce clean water by withdrawing the whey water from FO draw solution. However, it was ascertained that multi-step implementation of RO process is required for clean water production by which high amounts of dissolved salt ions led to relatively low performance. It was also concluded that by lowering NaCl concentration of the draw in FO step, the required order in RO step can be thoroughly minimized, improving the RO performance.

(3) MF pretreatment somewhat decreased the FO performance. However, after MF processing, MF concentrate could be centrifuged to recover fats from whey to produce cream or butter foods, as well as whey fractionating and lactose recovery by ultrafiltration and nanofiltration membrane processes from MF permeate. In addition to its use with preferentially MF pre-concentration of whey, FO–RO integrated membrane system could be employed by first concentrating the pre-concentrated or raw whey using FO and subsequently obtaining clean water from the FO draw solution using RO. Besides, after FO processing, the concentrated whey will be directly used for whey powder production by an appropriate process such as spray drying.

In light of the results obtained, FO–RO system seems to have an adequate technological base for recovering water from whey. In this respect, future work will become relevant to the investigation the technical and economic aspects of water recovery from whey by continuous operation of FO–RO system. By this means, together with the economics of whey powder production from the concentrated whey, a thorough assessment of FO–RO system in whey processing will be carried out for its real-scale implementation.

Acknowledgments

This study was financially supported by a national project (No: 109Y300) by the TUBITAK, the Scientific and Technological Research Council of Turkey.The

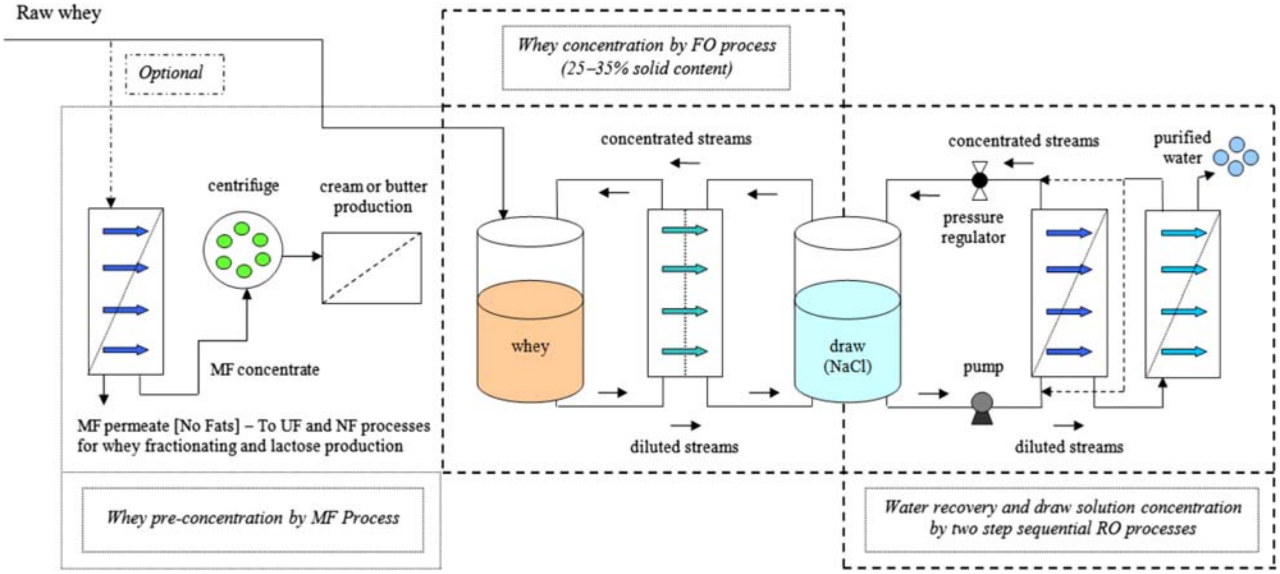

Fig. 8. FO–RO integrated membrane system proposed for water recovery and whey concentration from whey wastewater.

authors would like to thank the Cayirova Milk&Milk Products Inc., especially to Mr Seyhmuz Aslan and Ms Neslihan Genal, for the cheese whey supplement. Thanks are also due to Hydration Technologies Inc., and Hydranautics Inc., for the membrane supplements.

References

[1] T.Y. Cath, A.E. Childress, M. Elimelech, Forward osmosis: Principles, applications, and recent developments, J. Membrane Sci. 281 (2006) 70–87.

[2] T.Y. Cath, S. Gormly, E.G. Beaudry, M.T. Flynn, V.D. Adams, A.E. Childress, Membrane contactor processes for wastewater reclamation in space. Part I. Direct osmotic concentration as pretreatment for reverse osmosis, J. Membrane Sci. 257 (2005) 85–98.

[3] T.Y. Cath, V.D. Adams, A.E. Childress, Membrane contactor processes for wastewater reclamation in space II. Combined direct osmosis, osmotic distillation, and membrane distillation for treatment of metabolic wastewater, J. Membrane Sci. 257 (1–2) (2005) 111–119.

[4] J.R. McCutcheon, R.L. McGinnis, M. Elimelech, A novel ammonia–carbon dioxide forward (direct) osmosis desalination process, Desalination 174 (2005) 1–11.

[5] M.M. Ling, K.Y. Wang, T.S. Chung, Highly water–soluble magnetic nanoparticles as novel draw solutes in forward osmosis for water reuse, Ind. Eng. Chem. Res. 49 (2010) 5869–5876.

[6] C.R. Martinetti, A.E. Childress, T.Y. Cath, High recovery of concentrated RO brines using forward osmosis and membrane distillation, J. Membrane Sci. 331 (2009) 31–39.

[7] R.W. Holloway, A.E. Childress, K.E. Dennett, T.Y. Cath, Forward osmosis for concentration of anaerobic digester centrate, Water Res. 41 (2007) 4005–4014.

[8] E.R. Cornelissen, D. Harmsen, K.F. de Korte, C.J. Ruiken, J.J. Qin, H. Oo, L.P. Wessels, Membrane fouling and process performance of forward osmosis membranes on activated sludge, J. Membrane Sci. 319 (2008) 158–168.

[9] Y. Xu, X. Peng, C.Y. Tang, Q.S. Fu, S. Nie, Effect of draw solution concentration and operating conditions on forward osmosis and pressure retarded osmosis performance in a spiral wound module, J. Membrane Sci. 348 (2010) 298–309.

[10] R. Wang, L. Shi, C.Y. Tang, S. Chou, C. Qiu, A.G. Fane, Characterization of novel forward osmosis hollow fiber membranes, J. Membrane Sci. 355 (2010) 158–167.

[11] T.S. Chung, S. Zhang, K.Y. Wang, J. Su, M.M. Ling, Forward osmosis processes: Yesterday, today and tomorrow, Desalination 287 (2012) 78–81.

[12] J.R. McCutcheon, R.L. McGinnis, M. Elimelech, Desalination by ammonia–carbon dioxide forward osmosis: Influence of draw and feed solution concentrations on process performance, J. Membrane Sci. 278 (2006) 114–123.

[13] C.Y. Tang, Q. She, W.C.L. Lay, R. Wang, A.G. Fane, Coupled effects of internal concentration polarization and fouling on flux behavior of forward osmosis membranes during humic acid filtration, J. Membrane Sci. 354 (2010) 123–133.

[14] R.J. Aaberg, Osmotic power a new and powerful renewable energy source, ReFocus 4 (2003) 48–50.

[15] S. Loeb, Large-scale power production by pressure-retarded osmosis using river water and sea water passing through spiral modules, Desalination 143 (2002) 115–122.

[16] R.L. McGinnis, M. Elimelech, J. McCutcheon, Osmotic Heat Engine, Pub. No.: WO2008060435 (2008).

[17] L. Liu, M. Wang, D. Wang, C. Gao, Current patents of forward osmosis membrane process, Recent Patent Chem. Eng. 2 (2009) 76–82.

[18] A. Seppala, M.J. Lampinen, Thermodynamic optimizing of pressure retarded osmosis power generation systems, J. Membrane Sci. 161 (1999) 115–138.

[19] R.L. McGinnis, J.R. McCutcheon, M. Elimelech, A novel ammonia–carbon dioxide osmotic heat engine for power generation, J. Membrane Sci. 305 (2007) 13–19.

[20] S. Loeb, One hundred and thirty benign and renewable megawatts from Great Salt Lake? Desalination 141 (2001) 85–91.

[21] R.L. McGinnis, M. Elimelech, Global challenges in energy and water supply: The Promise of engineered osmosis, Environ. Sci. Technol. 42 (2008) 8625–8629.

[22] A. Achilli, T.Y. Cath, E.A. Marchand, A.E. Childress, The novel osmotic membrane bioreactor for wastewater treatment, in: Proc. of the 81st Annual Water Environment Federation Technical Exhibition and Conference, Water Environment Federation, Chicago, IL, USA, October 18–22, 2008, 12, pp. 6210–6221.

[23] K.Y. Wang, T.S. Chung, J.J. Qin, Polybenzimidazole (PBI) nanofiltration hollow fiber membranes applied in forward osmosis process, J. Membrane Sci. 300(1–2) (2007) 6–12.

[24] J.E. Miller, L.R. Evans, Sandia National Laboratories, Report. 2006.

[25] K.B. Petrotos, H.N. Lazarides, Osmotic concentration of liquid foods, J. Food Eng. 49 (2001) 201–206.

[26] B. Jiao, A. Cassano, E. Drioli, Recent advances on membrane processes for the concentration of fruit juices: A review, J. Food Eng. 63 (2004) 303–324.

[27] M.I. Dova, K.B. Petrotos, H.N. Lazarides, On the direct osmotic concentration of liquid foods. Part I. Impact of process parameters on process performance, J. Food Eng. 78 (2007) 422–430.

[28] Q. Yang, K.Y. Wang, T.S. Chung, A novel dual-layer forward osmosis membrane for protein enrichment and concentration, Sep. Purif. Tech. 69 (2009) 269–274.

[29] T.Y. Cath, S. Gormly, E.G. Beaudry, M.T. Flynn, V.D. Adams, A.E. Childress, Membrane contactor processes for wastewater reclamation in space. I. Direct osmotic concentration as pretreatment for reverse osmosis, J. Membrane Sci. 257 (2005) 85–98.

[30] T.Y. Cath, A.E. Childress, Membrane contactor processes for wastewater reclamation in space. II. Combined direct osmosis, osmotic distillation, and membrane distillation for treatment of metabolic wastewater, J. Membrane Sci. 257 (2005) 111–119.

[31] E.M. GarciaCastello, J.R. McCutcheon, M. Elimelech, Performance evaluation of sucrose concentration using forward osmosis, J. Membrane Sci. 338 (2009) 61–66.

[32] J.L. Cartinella, T.Y. Cath, M.T. Flynn, G.C. Miller, K.W. Hunter, A.E. Childress, Removal of natural steroid hormones from wastewater using membrane contactor processes, Environ. Sci. Technol. 40(23) (2006) 7381–7386.

[33] D.K. Anderson, Concentration of dilute industrial wastes by direct osmosis, University of Rhode Island, Providence, RI, 1977.

[34] E.G. Beaudry, J.R. Herron, Direct osmosis for concentrating wastewater, SAE Technical Paper (Paper Number: 972270) 14–17 (1997) 1–7.

[35] R.J. York, R.S. Thiel, E.G. Beaudry, Full-scale experience of direct osmosis concentration applied to leachate management, in: Proceedings of the Seventh International Waste Management and Landfill Symposium (Sardinia '99), S. Margherita di Pula, Cagliari, Sardinia, Italy, 1999.

[36] S. Grba, V.S. Tomas, D. Stanzer, N. Vahcic, A. Skrlin, Selection of yeast strain *Kluyveromyces marxianus* for alcohol and biomass production on whey, Chem. Biochem. Eng. Quart. 16 (2002) 13–16.

[37] A. Saddoud, I. Hassairi, S. Sayadi, Anaerobic membrane reactor with phase separation for the treatment of cheese whey, Bioresour. Technol. 98 (2007) 2102–2108.

[38] Q.H. Zhou, N. Kosaric, Effect of lactose and olive oil intra and extra cellular lipids of *Torulopsis bombicola*, Biotechnol. Lett. 15 (1993) 477–482.

[39] G.M.I. Siso, The biotechnological utilization of cheese whey: A review, Bioresour. Technol. 57 (1996) 1–11.

[40] B. Kavacik, B. Topaloglu, Biogas production from co-digestion of a mixture of cheese whey and dairy manure, Biomass Bioenerg. 34 (2010) 1321–1329.

[41] S. Ozmihci, F. Kargi, Kinetics of batch fermentation of cheese whey powder solution as function of the substrate and biomass concentrations, Bioresour. Technol. 98 (2007) 2978–2984.

[42] A. Kotoupas, F. Rigas, M. Chalaris, Computer-aided process design, economic evaluation and environmental impact assessment for treatment of cheese whey wastewater, Desalination 213 (2007) 238–252.

[43] APHA, AWWA, WEF, Standard methods for the examination of water and wastewater, in: A.D. Eaton, L. S. Clesceri, E. W. Rice and A. E. Greenberg (Eds.), 21st ed., Centennial Edition (2005).

[44] E.R. Cornelissen, D. Harmsen, K.F. De Korte, C.J. Ruiken, J.J. Qin, H. Oo, L.P. Wessels, Membrane fouling and process performance of forward osmosis membranes on activated sludge, J. Membrane Sci. 319 (2008) 158–168.

[45] C.H. Tan, H.Y. Ng, Modified models to predict flux behavior in forward osmosis in consideration of external and internal concentration polarizations, J. Membrane Sci. 324 (2008) 209–219.

[46] C. Aydiner, S. Topcu, C. Tortop, F. Kuvvet, D. Ekinci, N. Dizge, B. Keskinler, Interrelated analysis of performance and fouling behaviors in forward osmosis by ex-situ membrane characterizations, in: Viacheslav Kazmiruk (Ed.), The Scanning Electron Microscope, vol. 31, 2012, pp. 617–642, ISBN: 978-953-51-0092-8. Available from: http://www.intechopen.com.