International Dairy Journal 99 (2019) 104549

Contents lists available at ScienceDirect

International Dairy Journal

journal homepage: www.elsevier.com/locate/idairyj

Utilisation of salty whey ultrafiltration permeate with electrodialysis

Sahar Talebi [a], Esther Kee [b], George Q. Chen [a], Karren Bathurst [c], Sandra E. Kentish [a, *]

[a] The ARC Dairy Innovation Hub, Department of Chemical Engineering, University of Melbourne, Parkville, Victoria 3010, Australia
[b] Department of Chemical Engineering, University of Melbourne, Parkville, Victoria 3010, Australia
[c] Bega Cheese Pty. Ltd., 1 Vegemite Way, Port Melbourne, Victoria 3207, Australia

ARTICLE INFO

Article history:
Received 29 April 2019
Received in revised form
29 July 2019
Accepted 5 August 2019
Available online 17 August 2019

ABSTRACT

Salty whey is a waste by-product that incurs increasingly high disposal costs for the dairy industry. This study investigated electrodialysis of the ultrafiltration permeate of salty whey as either a concentrate for the treatment of sweet whey or as a source of lactose and salt. The type of concentrate (0.1 M NaCl or salty whey permeate) did not affect the rate of sweet whey demineralisation or the energy consumed per tonne of whey, but less sodium and more divalent cations were removed when salty whey permeate was used as the concentrate. Salty whey permeate could be effectively demineralised using either 0.1 M NaCl or a second stream of salty whey permeate as the concentrate. The concentrate purity could be enhanced using monovalent selective membranes without increasing the energy consumption of the process (3.2 ± 0.3 kWh per kg of NaCl removed from the diluate at 15 V).

1. Introduction

Salty whey is produced during the cheese salting process, with 50–60% of the added salt ending up in this stream (Chen, Gras, & Kentish, 2018). The protein content of this stream can be recovered through an ultrafiltration (UF) step. However, the high mineral content of the permeate stream from the UF process limits its utilisation in human and animal feed (Diblíková, Čurda, & Kinčl, 2013) and presents a major environmental concern if disposed of without treatment (Nishanthi, Chandrapala, & Vasiljevic, 2017). As the costs of salt disposal to the sewer are increasing in Australia, this is also becoming an economic burden to the dairy industry. Hence this paper examines different applications for salty whey permeate that might add value to dairy processing.

Firstly, the use of salty whey permeate as the concentrate stream during the electrodialysis (ED) of sweet whey (Fig. 1a) was examined. ED refers to the process in which ions are transferred through ion-exchange membranes (IEMs) under the application of an electric field. An ED module contains three major processing streams: (i) a diluate stream that is demineralised; (ii) a concentrate stream that uptakes the ions that are removed from the diluate stream; and (iii) an electrolyte solution that conducts the current through the system and protects the electrodes. The process has been commercialised for mineral removal from sweet whey prior to whey powder production. The use of salty whey permeate as the concentrate stream in such a process could eliminate the use of a concentrate stream prepared from fresh water and salt, thus reducing the overall water consumption and effluent salt load for the dairy factory.

Secondly, the potential to use salty whey permeate as the feed to the ED process (Fig. 1b) is considered. This allows the recovery of a demineralised lactose-rich stream that can be utilised for the production of lactose powders, while also producing a concentrated salt solution that could be either processed to produce salts for the chlor-alkali industry (Liu et al., 2016; Reig et al., 2014) or re-used in the cheese salting process. Using ED for salty whey processing has been investigated by Diblíková, Čurda, and Kinčl (2013). In this work the authors used fresh whey and reconstituted cheese whey with different dry matter and sodium chloride concentrations to simulate salty whey. ED has also been studied widely for concentrating salty brines produced from seawater desalination using reverse osmosis (RO). Such solutions are known for their high conductivity and sodium content (Liu et al., 2016; Reig et al., 2014). However, the sodium levels found in salty whey permeate are almost double the levels reported for these salty brines.

Furthermore, the use of monovalent selective IEMs in this application was investigated. These membranes can selectively pass sodium and chloride, generating a purer salt concentrate, while retaining the more nutritionally useful calcium within the lactose-rich stream. However, they tend to have a more limited pH

* Corresponding author. Tel.: +61 3 83446682.
E-mail address: sandraek@unimelb.edu.au (S.E. Kentish).

https://doi.org/10.1016/j.idairyj.2019.104549
0958-6946/

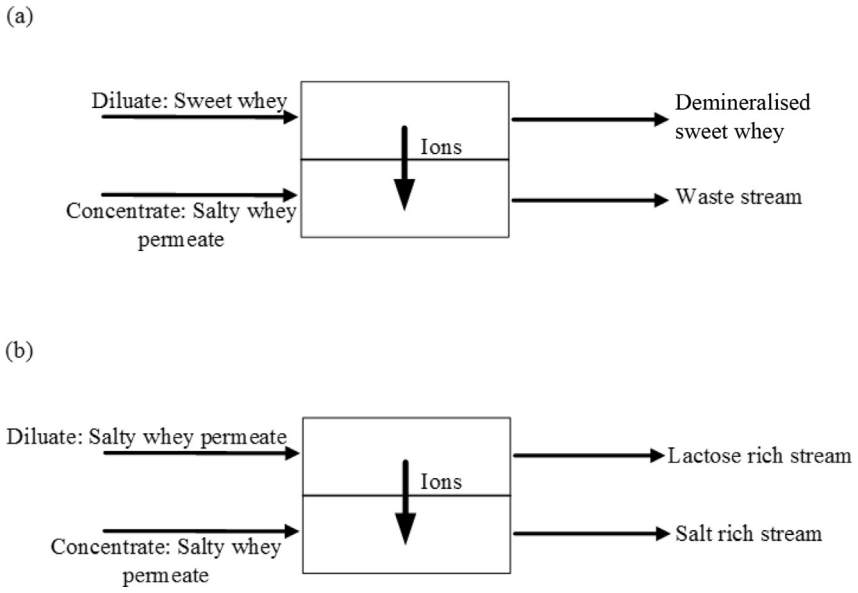

Fig. 1. Proposed applications for salty whey permeate: ED processes with (a) sweet whey as the diluate stream and salty whey permeate as the concentrate stream and (b) salty whey permeate as both diluate and concentrate streams.

and temperature tolerance range, thus making them more challenging to clean in a dairy processing environment. Andrés, Riera, and Alvarez (1995) examined the use of these membranes in the electrodialysis of skimmed milk intended for infant formula. The work presented in this manuscript is believed to be among the first that examines the use of monovalent-selective membranes in whey treatment.

2. Methods and materials

2.1. Materials

Skimmed sweet whey and salty whey permeate were obtained from dairy companies in Victoria, Australia. The salty whey permeate samples were produced using ultrafiltration to remove whey protein. The composition of the solutions is given in Table 1. The samples were stored at 4 ± 1 °C. Purified water (>8.6 MΩ cm; Merck Millipore KGaA, Darmstadt, Germany) was used in the preparation of all laboratory made solutions. Sodium sulphate (Na_2SO_4; >99%; Thermo Fisher Scientific Australia, Scoresby, Australia) with a concentration of 20 g L^{-1} was used as the electrolyte solution. Cleaning chemicals were prepared from hydrochloric acid (HCl; 36%; Thermo Fisher Scientific Australia), sodium chloride (NaCl; >99.5%; Merck KGaA, Darmstadt Germany) and 5 M sodium hydroxide (NaOH; Chem-Supply, Port Adelaide, Australia).

Table 1
The composition of skimmed sweet and salty whey permeate used in this work.[a]

Component	Unit	Sweet whey	Salty whey permeate
pH	–	6.3 ± 0.05	5.3 ± 0.1
Conductivity	mS cm^{-1}	6.4 ± 1	130 ± 4
Total solids	% (m/v)	6.5 ± 0.3	14.5 ± 0.5
Total protein	% (m/v)	1.04 ± 0.05	0.3 ± 0.05
K	mg 100 mL^{-1}	82.5 ± 4	160 ± 33
Na	mg 100 mL^{-1}	32 ± 1	5000 ± 500 (2.2 M)
Ca	mg 100 mL^{-1}	39 ± 7	130 ± 8
Mg	mg 100 mL^{-1}	9 ± 1	20 ± 2
Lactose	mg 100 mL^{-1}	Not measured	2600 ± 200
Lactic acid	mg 100 mL^{-1}	Not measured	0.72 ± 0.2

[a] Total protein = Total nitrogen × 6.38.

2.2. Electrodialysis unit

The experiments were carried out using an FTED-40 unit manufactured by FuMA-Tech GmbH (Bietigheim-Bissingen, Germany). Detailed information of the unit can be found elsewhere (Chen, Eschbach, Weeks, Gras, & Kentish, 2016). The module was arranged with three cation-exchange membranes (CEMs) separated by two anion-exchange membrane (AEMs) purchased from Astom (Tokyo, Japan) (see Table 2) and alternating diluate and concentrate spacers. The effective area per IEM was 36 cm^2. All membranes were preconditioned by soaking in 5% NaCl solution to allow for membrane hydration and expansion. After each trial the CMB and AHA membranes were removed from the ED unit and cleaned by soaking in HCl (pH 1.0 ± 0.15), followed by 3% NaCl (pH 9.2 ± 0.15) and then 5% NaCl (neutral pH) for two days each. This was done to ensure that the membranes were clean and had returned to their original ionic state. The alkali soaking step was eliminated for the monovalent selective membranes due to the low pH tolerance of the ACS membrane (Table 2).

2.3. Experimental protocol

The experiments were carried out in batch mode, with all solutions continuously recycled to their supply tanks. The diluate and concentrate flowrates were maintained at 500 mL min^{-1}, while the electrolyte flowrate was kept at 1000 mL min^{-1} as recommended by the manufacturer to minimise concentration polarisation effects. Samples were collected (5–10 mL) on an hourly basis from the diluate and concentrate tanks for analysis.

For the demineralisation of sweet whey, a total volume of 2 L was used for both diluate and concentrate streams and the experiments were terminated when 75% sweet whey demineralisation was achieved. Both salty whey permeate and 0.1 M NaCl solution were used as the concentrate solution. These experiments were performed at 44 ± 2 °C to minimise microbial growth.

For the demineralisation of salty whey permeate, experiments were run for 8 h using either salty whey permeate or 0.1 M NaCl as the concentrate solution. The unit was operated either at a constant current density of 55 mA cm^{-2}; or a constant voltage of 5, 10 or

Table 2
Properties of Neosepta ion-exchange membranes used in this work (provided by the manufacturer).

Property	Unit	Cation exchange membrane		Anion exchange membranes	
		CMB	CIMS	AHA	ACS
Type	–	Strong acid (Na type)		Strong base (Cl type)	
Characteristics	–	Alkaline resistance	Monovalent cation selective	Alkaline resistance	Monovalent anion selective
Thickness	mm	0.21	0.15	0.22	0.13
Temperature range	°C	≤60	≤40	≤60	≤40
pH range	–	0–14	0–10	0–14	0–8
Electrical resistance	Ω cm^2	4.5	1.8	4.1	3.8

15 V. The experiments were performed at 26 ± 2 °C. Ambient temperatures can be used with salty whey, since the high salinity limits bacterial growth. The volume ratio between the concentrate and diluate tanks was maintained at 1:4 (actual volumes were 1 L and 4 L for concentrate and diluate tanks, respectively).

While it was not possible to experimentally determine the limiting current density of this system, operation was likely to be below this value for the salty whey permeate/salty whey permeate system, based on measurements in comparable systems (Dlugolęcki, Anet, Metz, Nijmeijer, & Wessling, 2010). The initial current density was 56, 167, and 278 mA cm^{-2} at 5, 10 and 15 V, respectively. Conversely, for the salty whey permeate/0.1 M NaCl system, the initial current density was 41, 125, and 194 mA cm^{-2} at 5, 10 and 15 V, respectively, which is likely to be above the limiting current density at the two higher voltages, given the lower ionic strength of the concentrate solution.

2.4. Analysis methods

Inductively coupled plasma optical emission spectroscopy (ICP-OES 720ES, Varian, Santa Clara, CA, USA) was used to measure the concentration of sodium (Na), potassium (K), calcium (Ca), and magnesium (Mg) in both the diluate and concentrate tanks. Total solid analysis was performed by weighing 2 mL of samples before drying under vacuum at 100 °C for more than 20 h in a fan forced oven to evaporate all the moisture.

During each experiment, the conductivity and pH of the concentrate tank were measured continuously using a pH probe (Mettler-Toledo, Greifensee, Switzerland) and a conductivity probe (Crison Instruments, Greifensee, Switzerland), while the diluate conductivity was measured using a second conductivity probe (Mettler-Toledo). At the start and end of each experiment, the conductivity and pH of the electrode solution was also recorded. The recorded conductivity value can be used to calculate the diluate demineralisation rate (DR) according to Eq. (1) (Cifuentes-Araya, Pourcelly, & Bazinet, 2013; Jiang, Wang, Zhang, & Xu, 2014):

$$DR = \left(\frac{\chi_i - \chi_f}{\chi_i}\right) \times 100 = \left(1 - \frac{\chi_f}{\chi_i}\right) \times 100 \quad (1)$$

where χ_i and χ_f are the initial and final conductivity of the diluate solution in mS cm^{-1}, respectively.

The current and voltage were read directly from the current supply and used to calculate the system resistance using Ohm's law. The energy consumed (kWh kg^{-1} of NaCl removed) during the batch process can be estimated using Eq. (2) (Reig et al., 2014). The diluate concentration was used as the calculation basis. The sodium chloride concentration was estimated from the sodium concentration, assuming that chloride is the dominant anion present in this system.

$$E_{NaCl} = \frac{V_{cell} \cdot I \cdot t}{\left(v_{D,i} \cdot C_{Na,i} - v_{D,f} \cdot C_{Na,f}\right) \cdot \left(MW_{NaCl}/MW_{Na}\right)} \quad (2)$$

where V_{cell} is the mean stack potential (V), t is the time (h), I is the current (A), MW is the molecular weight (g mol^{-1}), v and C_{Na} refer to the volume (L) and concentration of Na (g L^{-1}) in the diluate tank, i and f indicate the beginning and end of the experiment, respectively.

Purity was defined by (Zhang et al., 2017) as the NaCl concentration in the final concentrate solution according to Eq. (3):

$$P_{NaCl} = \frac{v_{C,f} \cdot c_{Na,f} \cdot \left(MW_{NaCl}/MW_{Na}\right)}{m_{C,dry}} \times 100\% \quad (3)$$

where $c_{Na,f}$ is the final concentration of Na ion in the concentrate tank (g L^{-1}), and $m_{C,dry}$ is the final solid weight in the concentrate tank (g). However, using this equation in the present work resulted in values greater than 100% as the salty whey permeate contains anions other than chloride. Therefore, purity in this work is used only as a qualitative parameter.

3. Results and discussion

3.1. Use of salty whey as concentrate stream for sweet whey demineralisation

Initial experiments used a constant voltage of 7 V with fresh sweet whey as the diluate stream and either salty whey permeate (2.2 M Na) or 0.1 M NaCl as the concentrate. The reduction in diluate conductivity was identical for both types of concentrate solutions, as illustrated in Fig. 2a. However, the concentrate conductivity only increased by 1 mS cm^{-1} for salty whey permeate while it increased by 5 mS cm^{-1} for the 0.1 M NaCl solution. This more limited change in conductivity partly reflects the nonlinear relationship between conductivity and salt concentration, but also probably reflects an osmotic flow of water from the diluate to the salty whey permeate (concentrate tank), given their very different concentrations (Jiang et al., 2014; Liu et al., 2016; Reig et al., 2014). Analysis of ion concentrations (Fig. 2b), shows that fewer Na ions were removed from the sweet whey when salty whey permeate was used as the concentrate stream. Conversely, a greater quantity of all other ions (K, Ca and Mg ions) were removed. Such a behaviour is a result of the high concentration of Na in the salty whey permeate thus resulting in diffusional backflow of Na from the concentrate to the diluate. However, the final Na concentration in the diluate tank (2.8 ± 0.3 g Na per kg of dry mass) falls in the reported range for demineralised whey powders.

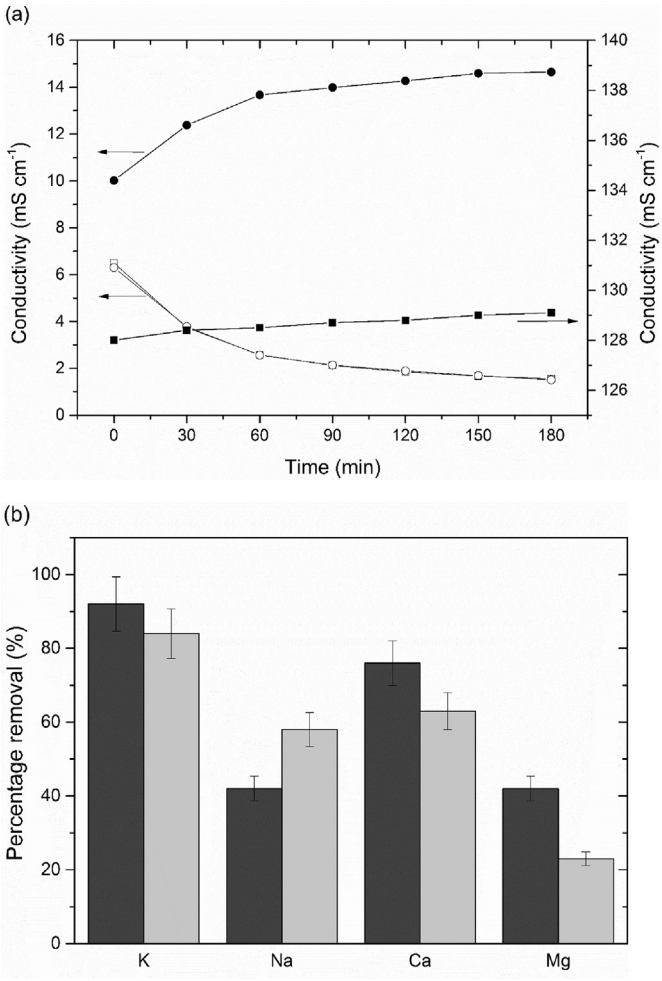

Fig. 2. Demineralisation of sweet whey under a constant voltage of 7 V using two different concentrate streams: (a) change in the conductivity of the diluate (□, sweet whey/salty whey permeate; ○, sweet whey/0.1 M NaCl) and concentrate (■, sweet whey/salty whey permeate; ●, sweet whey/0.1 M NaCl) and (b) percentage removal of cations from sweet whey (■, salty whey permeate; ■, 0.1 M NaCl).

The initial system resistance for the sweet whey/salty whey permeate system was lower compared with the sweet whey/0.1 M NaCl system (see Fig. 3), due to the higher conductivity of the salty whey. The resistance increased with time for both systems due to diluate demineralisation and possibly membrane fouling (Bazinet & Araya-Farias, 2005; Casademont, Farias, Pourcelly, & Bazinet, 2008). Since both systems achieved 75% demineralisation in 3 h, the greater increase in resistance for the sweet whey/0.1 M NaCl system suggests that membrane fouling was greater comparing to that for the sweet whey/salty whey permeate system. There was indeed some visual evidence of greater scaling for this system on the sides of the membranes facing the concentrate (see Supplementary material; Figs. S1 and S2).

Although salty whey permeate contains more salts that might cause membrane scaling, the solution pH played a significant role. As observed from Fig. 4, the pH increased for both concentrate solutions, however, the increase was greater for the 0.1 M NaCl solution due to its lower buffering capacity (see Supplementary material, Fig. S3). It is well known that the solubility of calcium salts reduces with an increase in pH, thus resulting in their precipitation on the membrane surface. When 0.1 M NaCl solution is used as the concentrate stream, membrane fouling could be avoided or reduced by adjusting the pH of the solution (Talebi et al., 2019). However, adjusting the pH for salty whey permeate is less readily achieved because of the system buffering (see Supplementary material, Fig. S3). Fouling in such systems could be avoided or minimised using a pulsed electrical field, electrodialysis reversal, using higher liquid flowrates or by acid cleaning.

The use of the salty whey permeate as the concentrate may lead to lower power consumption, because of the lower resistance as shown in Fig. 3. However as noted, more energy is wasted in this system due to the diffusional backflow of sodium and the greater transfer of ions such as calcium and magnesium, so the total energy consumption per kg of NaCl removed, is indeed higher when this stream is used (Table 3). However, sodium ions are not the dominant ions in sweet whey (Table 1), and therefore calculating the energy per kg of NaCl removed is overestimating the energy cost of the process. The energy consumed per tonne of sweet whey treated is more comparable for both systems (Table 3) and falls within the range reported by Chen et al. (2016) for 75% demineralisation.

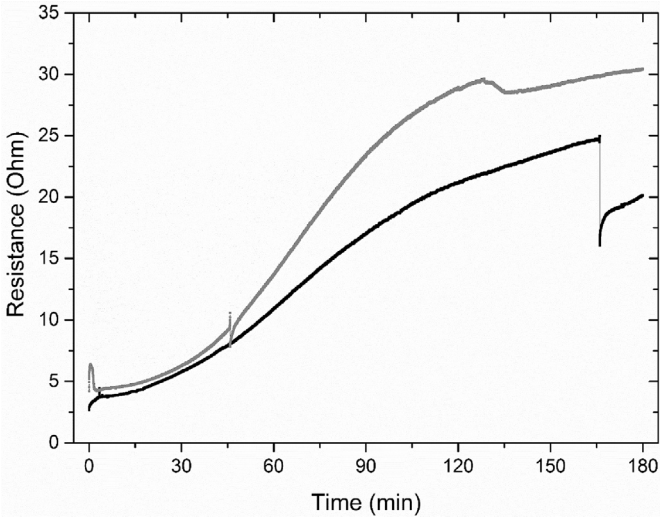

Fig. 3. Change in system resistance during the demineralisation of sweet whey under a constant voltage of 7 V using two different concentrate streams: sweet whey/salty whey permeate system (dark grey trace) and sweet whey/0.1 M NaCl system (pale grey trace).

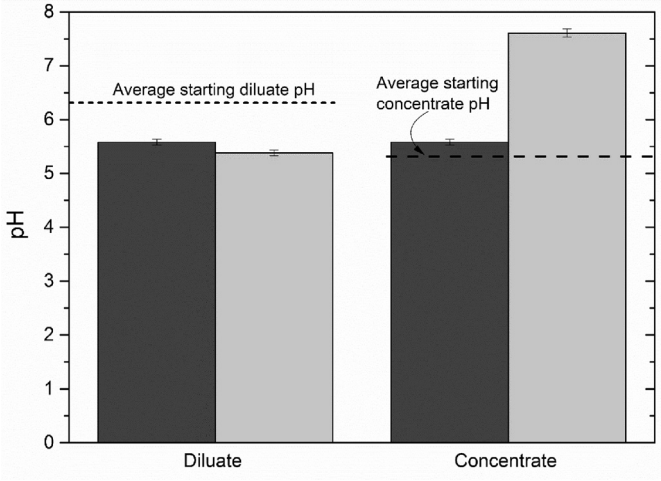

Fig. 4. pH of the diluate and concentrate tanks after 75% demineralisation of sweet whey under a constant voltage of 7 V using two different concentrate streams: salty whey permeate (■) and 0.1 M NaCl (■).

Table 3
Energy consumption for 75% demineralisation of sweet whey at 7 V and using a concentrate of either 0.1 M NaCl or salty whey permeate (2.2 M Na).

Energy calculation basis	Salty whey permeate	0.1 M NaCl
Removal of sodium (kWh kg^{-1} NaCl)	18 ± 0.5	11.4 ± 0.5
Removal of monovalent cations (kWh kg^{-1} NaCl and KCl)	3.7 ± 0.5	3.2 ± 0.5
Whey treated (kWh ton^{-1} whey)	7.4 ± 0.5	5.9 ± 0.5

3.2. Demineralisation of salty whey using ED

3.2.1. Constant current mode

The use of salty whey permeate as the diluate was next examined, with the objective of creating a purified lactose stream and a salt stream suitable for re-use. Initial experiments were run in constant current mode with salty whey permeate acting as diluate, and either salty whey permeate or 0.1 M NaCl as the concentrate.

As observed for the sweet whey case, the change in diluate conductivity over a period of 8 h was identical regardless of the salinity of the concentrate solution (Fig. 5a). This was consistent with the measured change in concentrations of the major ions in the diluate stream, with the sodium ion concentration falling by 6.5 ± 0.5 g L^{-1} regardless of the concentrate type. Again, the change in concentrate conductivity was lower for the salty whey permeate, reflecting the osmotic flows of water from diluate to concentrate. The concentrate solution volume changed by 160 ± 10 mL when 0.1 M NaCl was used versus 195 ± 10 mL when salty whey permeate was used. The resistance was lower for the system with salty whey permeate as concentrate due to its higher concentration. (Fig. 5b).

During the 8 h operation, changes in the concentration of the divalent ions in the diluate tank were within the experimental error margins. The main reason behind such an observation is the high concentration of monovalent ions found in salty whey permeate (Table 1). No significant changes were noticed in the diluate pH but the concentrate pH fell from 5.5 ± 0.25 to 3.4 ± 0.1 for 0.1 M NaCl solution and to 5.1 ± 0.1 for salty whey permeate. The more limited drop in the salty whey permeate pH was again a result of the greater buffering capacity of this system (Supplementary material, Fig. S3). This is in contrast to the sweet whey system, where the pH increased in the concentrate (Fig. 4).

3.2.2. Constant voltage mode

Similar to constant current experiments, no major differences were observed in the final diluate conductivity or monovalent ion removal rates between the salty whey permeate and 0.1 M NaCl concentrates at applied voltages of 5 V and 10 V (see Table 4, Figs. 6 and 7). However, when the applied voltage was increased to 15 V, more sodium was removed when the 0.1 M NaCl solution was used (Fig. 7). This was again the result of less back diffusion in this concentrate, relative to that of salty whey permeate. Due to the absence of other ions or lactose in the starting 0.1 M NaCl solution, the final NaCl purity in this concentrate will be higher. The energy consumption is less than that obtained for the sweet whey system due to the lower solution resistance as salty whey permeate has a much higher conductivity than sweet whey (Fig. 8). These energy consumptions are still relatively high compared with the values reported for RO brine by Reig et al. (2014) (<1.2 kWh kg^{-1} of NaCl) and Liu et al. (2016) (1.4 kWh kg^{-1} of NaCl at 15 V). However, such values reflect the complexity of the whey solution due to the presence of lactose, residual protein and larger ions. Furthermore, the consumed energy is overestimated as all Na will not be present as NaCl.

3.2.3. Monovalent-selective membranes

Experiments were finally conducted to determine whether monovalent selective IEMs (CIMS/ACS) could be used to improve the quality of the final concentrate stream so that it can more readily be re-used; and to retain nutritionally important calcium in

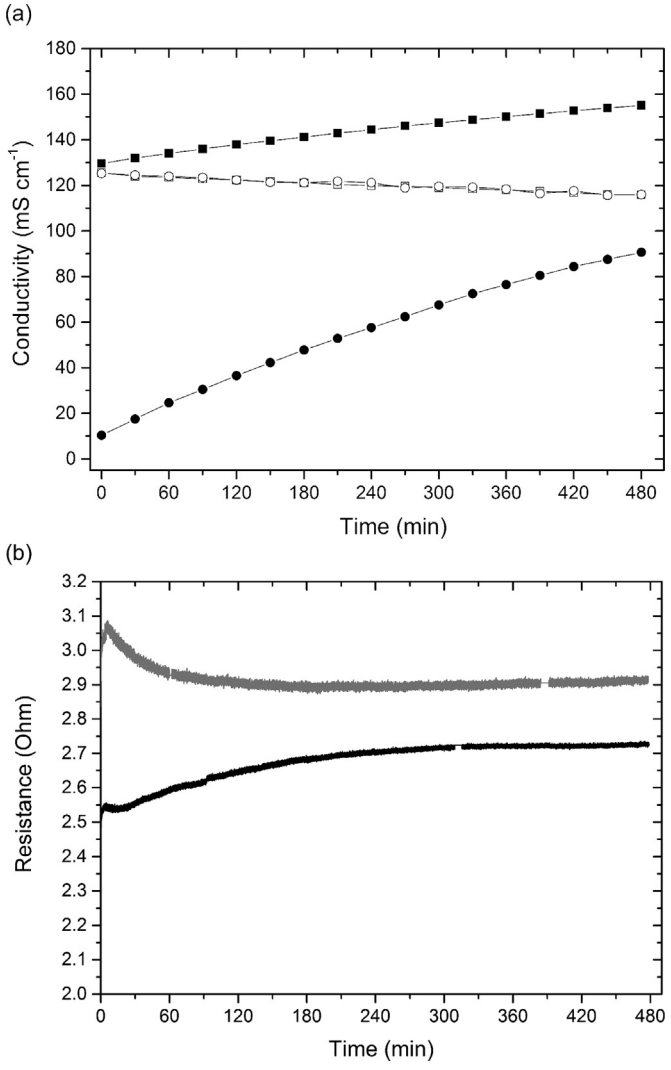

Fig. 5. Demineralisation of salty whey over a period of 8 h under a constant current density of 55 A m^{-2}: (a) change in the conductivity of the diluate (□, salty whey permeate/salty whey permeate system; ○, and salty whey permeate/0.1 M NaCl system) and concentrate (■, salty whey permeate/salty whey permeate system; ●, salty whey permeate/0.1 M NaCl system) and (b) system resistance (dark grey trace, salty whey permeate/salty whey permeate system; pale grey trace, salty whey permeate/0.1 M NaCl system).

Table 4
Process outcomes for constant voltage experiments conducted over 8 h with salty whey permeate as the diluate and either salty whey permeate or 0.1 M NaCl as concentrate.

Parameter	Salty whey permeate			0.1 M NaCl		
	5 V	10 V	15 V	5 V	10 V	15 V
Change in concentrate volume (mL)	140 ± 5	540 ± 30	650 ± 30	100 ± 5	480 ± 30	750 ± 40
Demineralisation rate (DR)	6.5	22	33	6.5	22	38

the final diluate. These experiments were conducted at a constant voltage of 15 V using 0.1 M NaCl as the concentrate stream.

The concentrate conductivity increased further for the monovalent selective IEMs (CIMS/ACS) compared with the non-selective IEMs (CMB/AHA) (Fig. 9a). However, this only reflects the fact that the conductivity of monovalent ions is greater than that of divalent ions. As shown in Fig. 10, the monovalent selective IEMs allow less divalent ions to pass through the membranes and hence more monovalent ones are transferred (see Supplementary material, Fig. S4). The lower the concentration of multivalent ions in the concentrate stream, the higher will be the purity of the NaCl produced (Zhang et al., 2017). The energy consumed per kg of NaCl removed from the diluate tank was comparable for both types of membranes (3.2 ± 0.3 kWh kg^{-1} of NaCl for CIMS/ACS, and 3.4 ± 0.3 kWh kg^{-1} of NaCl for CMB/AHA). While these results would tend to suggest that these monovalent selective IEMs can indeed produce a cleaner salt solution for the same energy demand, this needs to be balanced against the higher cost of these membranes and their more limited tolerance of both process temperature and pH (Table 2), making them less attractive for industrial applications.

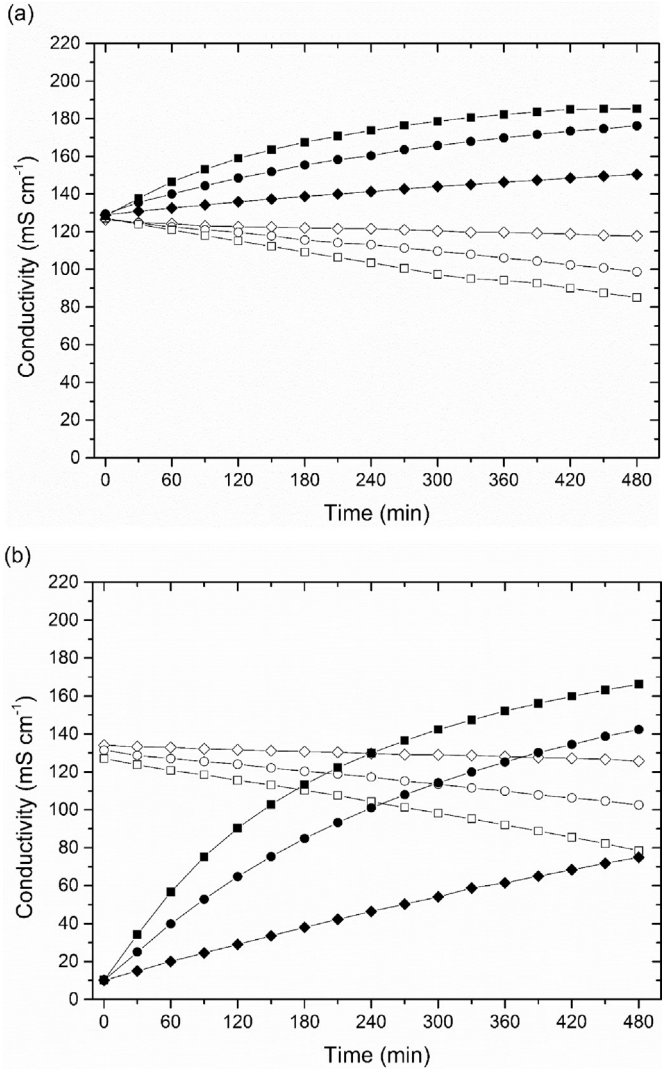

Fig. 6. Demineralisation of salty whey permeate over a period of 8 h under different applied voltages showing the change in the conductivity of the diluate at 5 V (◇), 10 V (○) and 15 V (□) and concentrate at 5 V (◆), 10 V (●) and 15 V (■) for (a) salty whey permeate and (b) 0.1 M NaCl as the concentrate solution.

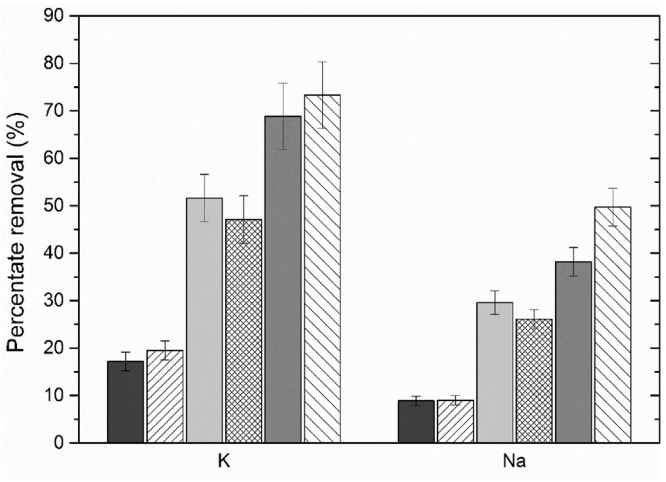

Fig. 7. Percentage removal of monovalent ions from salty whey permeate during demineralisation over an 8 h period with the concentrate solution being either salty whey permeate at an applied voltage of 5 V (■), 10 V (□) and 15 V (■) or 0.1 M NaCl at an applied voltage of 5 V (▨), 10 V (■) and 15 V (▧).

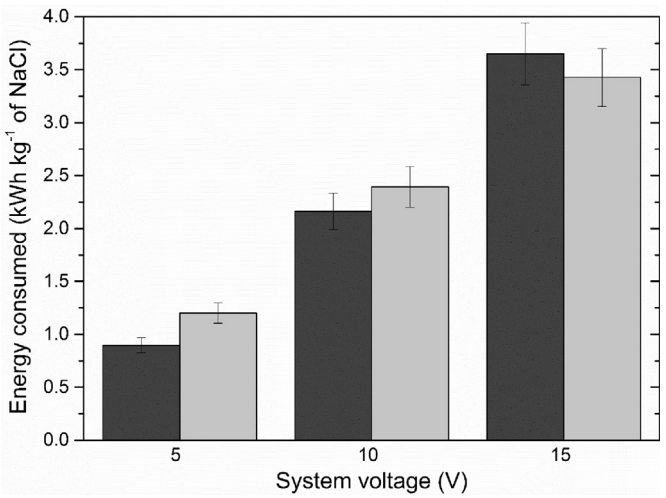

Fig. 8. Energy consumption during demineralisation of salty whey permeate over an 8 h period with either salty whey permeate (■) or 0.1 M NaCl (■) as the concentrate solution, as a function of the applied voltage.

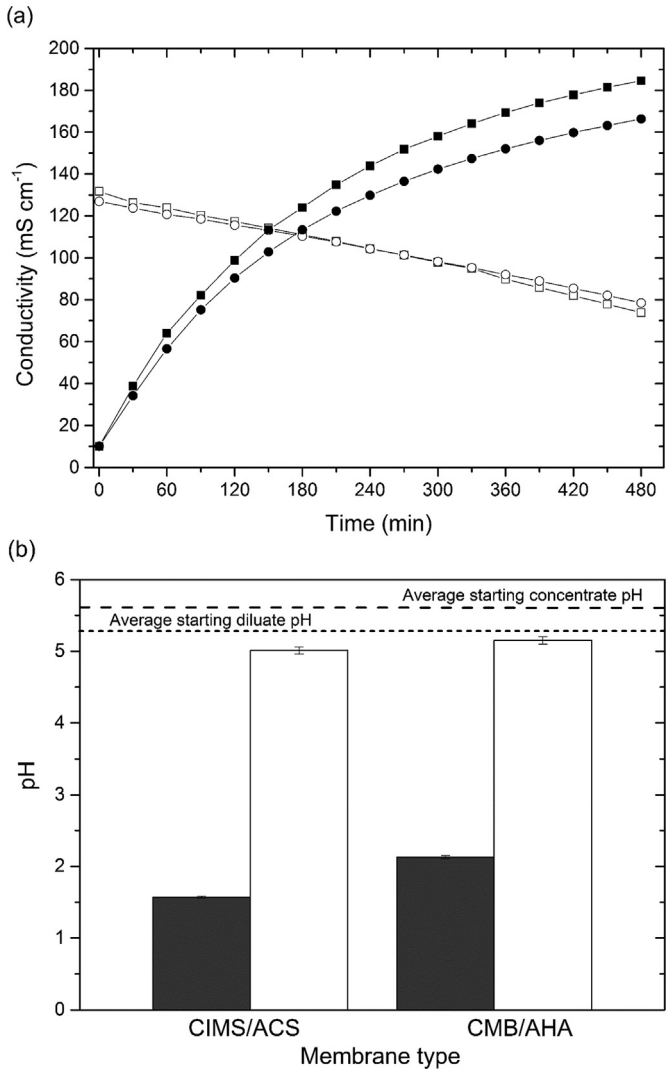

Fig. 9. Demineralisation of salty whey permeate over a period of 8 h under a constant voltage of 15 V with 0.1 M NaCl as concentrate: (a) the change in conductivity of the diluate (□) and concentrate (■) using monovalent selective (CIMS/ACS) membranes; and using the change in conductivity of the diluate (○) and concentrate (●) for non-selective (CMB/AHA) membranes; and (b) the final pH of the diluate (□) and concentrate (■).

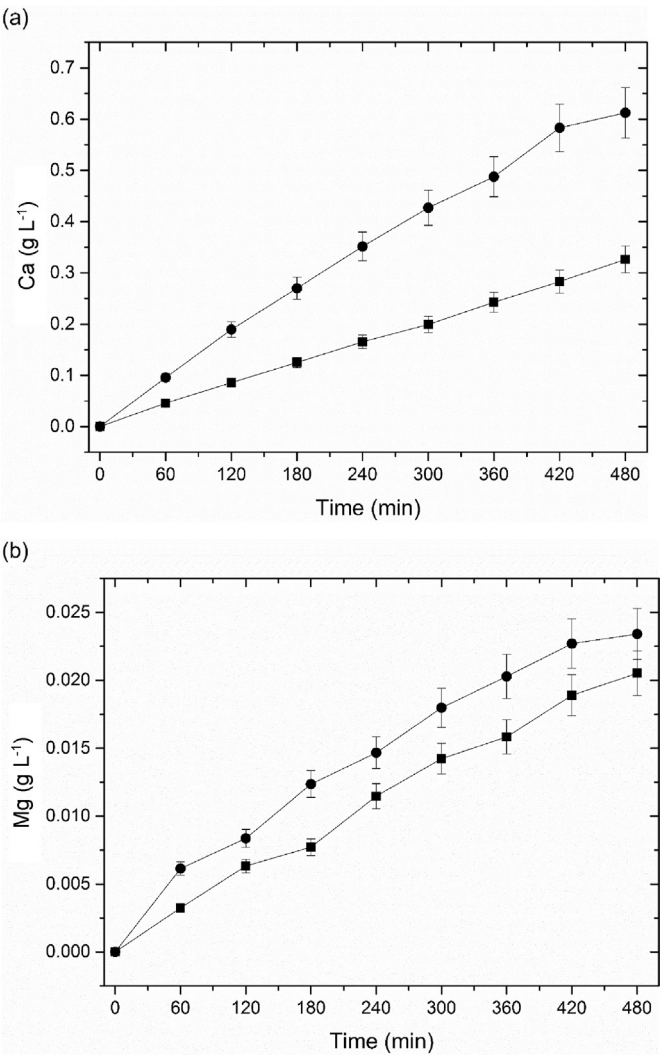

Fig. 10. Concentration of divalent ions in the concentrate compartment over 8 h of demineralisation of salty whey permeate with 0.1 M NaCl as concentrate at a system voltage of 15 V using monovalent selective (CIMS/ACS) (■) or non-selective (CMB/AHA) (●) membrane pairs: (a) calcium; (b) magnesium.

Regardless of the type of ions transferred to the concentrate tank, the change in tank volume was 700 ± 35 mL for both cases. The monovalent selective membranes are known to have lower electrical resistance (see Table 2) and this was reflected in a lower system resistance calculated for these experiments (see Supplementary material, Fig. S5). The difference in the resistance between both systems is consistent with Table 2, thus suggesting the absence of greater water splitting for the CIMS/ACS system. The lower pH of the concentrate stream for the monovalent selective IEMs as shown in Fig. 9b may reflect a lower concentration of multivalent phosphate ions, which add buffering capacity.

The low pH values obtained for both concentrates after 8 h of processing at 15 V could limit the usefulness of these streams for re-use in cheese making. However, the decrease in pH is likely due to water splitting as a result of the low ionic strength of the concentrate solution and the high voltage used in these experiments. This could be eliminated at industrial scale by operating with a lower voltage per membrane pair or using a concentrate of higher ionic strength.

4. Conclusions

This work has shown that salty whey permeate can be used as the concentrate stream for sweet whey demineralisation, with 75% demineralisation achieved within 3 h at 7 V with two membrane pairs of 36 cm^2 each in size. This time could be reduced at industrial scale by increasing the membrane area. Furthermore, membrane fouling was less extensive when a salty whey permeate was used as the concentrate stream. However, while the overall demineralisation rates were comparable, the levels of sodium removed were lower with this concentrate (43% versus 58% for 0.1 M NaCl). While the energy demand was higher per kg of sodium removed from the diluate tank, this demand was comparable with that for a sweet whey/0.1 M NaCl system when the energy demand was calculated per tonne of treated whey. The quality of the concentrated salt solution produced from salty whey permeate demineralisation can be improved by utilising monovalent selective IEMs. The concentration of calcium in the concentrate solution dropped to half of the amount found when using non-selective IEMs. This, in turn, results in a concentrate of greater purity. The energy consumption is comparable for both types of membranes. However, the non-

selective IEMs have a higher pH tolerance range thus allowing the use of base cleaning agents within the dairy factory environment.

Acknowledgements

This research was supported under Australian Research Council's Industrial Transformation Research Program (ITRP) funding scheme (Project Number IH120100005). The ARC Dairy Innovation Hub is a collaboration between The University of Melbourne, The University of Queensland and Dairy Innovation Australia Ltd.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.idairyj.2019.104549.

References

Andrés, L. J., Riera, F. A., & Alvarez, R. (1995). Skimmed milk demineralization by electrodialysis: Conventional versus selective membranes. *Journal of Food Engineering, 26*, 57–66.

Bazinet, L., & Araya-Farias, M. (2005). Electrodialysis of calcium and carbonate high concentration solutions and impact on composition in cations of membrane fouling. *Journal of Colloid and Interface Science, 286*, 639–646.

Casademont, C., Farias, M. A., Pourcelly, G., & Bazinet, L. (2008). Impact of electrodialytic parameters on cation migration kinetics and fouling nature of ion-exchange membranes during treatment of solutions with different magnesium/calcium ratios. *Journal of Membrane Science, 325*, 570–579.

Chen, G. Q., Eschbach, F. I., Weeks, M., Gras, S. L., & Kentish, S. E. (2016). Removal of lactic acid from acid whey using electrodialysis. *Separation and Purification Technology, 158*, 230–237.

Chen, G. Q., Gras, S. L., & Kentish, S. E. (2018). Separation technologies for salty wastewater reduction in the dairy industry. *Separation and Purification Reviews*, 1–29.

Cifuentes-Araya, N., Pourcelly, G., & Bazinet, L. (2013). Water splitting proton-barriers for mineral membrane fouling control and their optimization by accurate pulsed modes of electrodialysis. *Journal of Membrane Science, 447*, 433–441.

Diblíková, L., Čurda, L., & Kinčl, J. (2013). The effect of dry matter and salt addition on cheese whey demineralisation. *International Dairy Journal, 31*, 29–33.

Dlugolęcki, P., Anet, B., Metz, S. J., Nijmeijer, K., & Wessling, M. (2010). Transport limitations in ion exchange membranes at low salt concentrations. *Journal of Membrane Science, 346*, 163–171.

Jiang, C., Wang, Y., Zhang, Z., & Xu, T. (2014). Electrodialysis of concentrated brine from RO plant to produce coarse salt and freshwater. *Journal of Membrane Science, 450*, 323–330.

Liu, J., Yuan, J., Ji, Z., Wang, B., Hao, Y., & Guo, X. (2016). Concentrating brine from seawater desalination process by nanofiltration–electrodialysis integrated membrane technology. *Desalination, 390*, 53–61.

Nishanthi, M., Chandrapala, J., & Vasiljevic, T. (2017). Compositional and structural properties of whey proteins of sweet, acid and salty whey concentrates and their respective spray dried powders. *International Dairy Journal, 74*, 49–56.

Reig, M., Casas, S., Aladjem, C., Valderrama, C., Gibert, O., Valero, F., et al. (2014). Concentration of NaCl from seawater reverse osmosis brines for the chlor-alkali industry by electrodialysis. *Desalination, 342*, 107–117.

Talebi, S., Chen, G. Q., Freeman, B., Suarez, F., Freckleton, A., Bathurst, K., et al. (2019). Fouling and in-situ cleaning of ion-exchange membranes during the electrodialysis of fresh acid and sweet whey. *Journal of Food Engineering, 246*, 192–199.

Zhang, W., Miao, M., Pan, J., Sotto, A., Shen, J., Gao, C., et al. (2017). Separation of divalent ions from seawater concentrate to enhance the purity of coarse salt by electrodialysis with monovalent-selective membranes. *Desalination, 411*, 28–37.