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Use of Nanofiltration for Concentration and Demineralization in the Dairy Industry

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Abstract: In laboratory-scale experiments, rejection-flux curves of four different commercial membranes were established for four different model solutions (NaCl, $CaCl_2$) and (NaCl + $CaCl_2$) and for Ultrafiltration (UF) whey-permeate (pH = 4.6). The results indicated that the salt transport through all the NF membranes investigated depends on the flux. At low flux, when the contribution of diffusive transport is the most important, permeability of (specially monovalent) cations is high. At high flux, when transport by convection is the most important, rejection reaches a maximum (constant) value. From this it follows that the salt transport can be controlled by the flux. The parameters derived from the results with UF-whey permeate can be used to predict the salt rejection for similar multi-component systems like whey and UF-permeate in industrial systems.

Key words: Nanofiltration, whey, flux, rejection, demineralization, membrane

INTRODUCTION

Nanofiltration (NF) is characterized by a membrane pore size between 0.5 and 2 nm and operating pressures between 5 and 40 bars. The pore size corresponds to a molecular weight cut off value of approximately 300-500 g mol^{-1}. NF membranes have a slightly charged surface and this can be used to separate ions with different valences (Rautenbach and Groschi, 1990).

Mass transfer in NF is described by different theories, in Extended Nernst Planck (ENP) model transfer in boundary layer between solution and membrane and the body of membrane is discussed separately. A representation of the mass transfer process occurring in NF is given in Fig. 1.

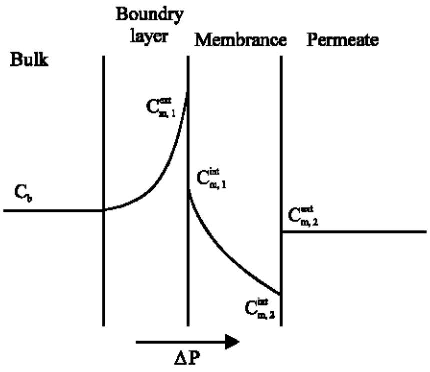

Fig. 1: Mass transfer in nanofiltration

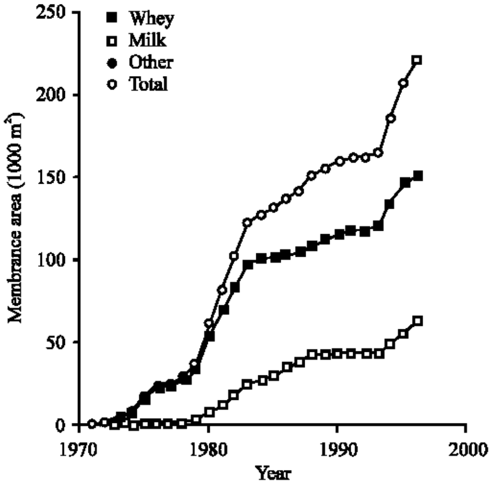

Fig. 2: Total installed membrane area world-wide for NF of dairy liquids

World-wide an increasing amount of whey is industrially processed to whey powders and other high-quality, protein-rich products meant for nutritional use (Clark, 1987; Zadow, 1987). Whey intended for human or animal consumption will increase in value if it is demineralised (Kelly *et al*., 1991; Horton, 1987). Nanofiltration (NF) is an alternative for the concentration and demineralization of whey by Evaporation (EV) + by Electrodialysis (ED) which is now in practice (Gregory, 1987). NF-membranes have a high permeability for (monovalent) salts (NaCl, KCl) and have a very low permeability for organic compounds (lactose, proteins, urea). The use of NF instead of EV+ED has the advantage of simultaneous concentration and demineralization of whey (Horton, 1987). This will lead to a considerable reduction of the costs for energy consumption, waste

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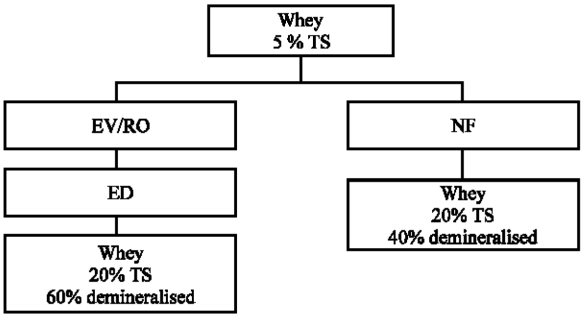

Fig. 3: Concentration and demineralization of whey in two steps using Evaporation (EV) or Reverse Osmosis (RO) followed by electrodialysis and in one step using Nanofiltration (NF)

water disposal and total costs (Kelly *et al.*, 1991; Gregory, 1987). This way the increasing use of NF technology in dairy industry is well justified. Figure 2 shows the membrane area for NF that has been installed in the dairy industry (Timmer and Van der Horst, 1998).

Figure 3 shows the advantage of nanofiltration, in comparison with current methods, through shorter route towards demineralization.

MATERIALS AND METHODS

To observe the practical use of NF-membranes in whey processing a pilot experiment is undergone in laboratory of faculty of environment, university of Tehran during winter and spring of the year 2005. The samples used in this research are as follows:

- A 17 mM Solution of NaCl (pro analysis, E. Merck. Darmstadt, FRG) in demineralised water.
- A 13.2 mM Solution of $CaCl_2$ (pro analysis, E. Merck, Darmstadt, FRG) in demineralised water.
- A 17 m M Solution of NaCl and 13.2 mM $CaCl_2$ in demineralised water.
- UF-whey permeate, pH 4.6. UF-whey permeate powder (NIZO, Nether lands) was dissolved in demineralised water (5% m m^{-1}) and pH was adjusted to 4.6 (5 N HCl) in order to obtain a clear solution. A typical composition of the UF-whey permeate at pH 4.6 is given in Table 1.

Four different makes of commercial membranes were used. Names and properties are given in Table 2.

Table 1: Composition of UF-whey permeate pH 4.6

Concentration	Component	Concentration	Component
Sodium (mg g^{-1})	361	Sulfate (mg g^{-1})	117.00
Potassium (mg g^{-1})	2208	nitrate (mg g^{-1})	49.00
Calcium (mg g^{-1})	197	lactose (%)	4.00
Phosphate (mg g^{-1})	2030	Protein (%)	0.18
Phosphate (mg g^{-1})	1647	total solids (%)	5.00
Citrate (mg g^{-1})	1089		

Table 2: NF-membranes used in experiments

Membrane	Manufacturer/Source	pH	Temperature (C)	Material
FE700-004	Filtration Engineering	2-11	50	PA/PS*1
NF40	Film Tec	2-11	45	PA/PS*1
HC50	Dow Chemical	2-10	60	PA/PS*1
CA960PP	Dow Chemical	2-8	30	CA*2

*1 PA/PS = Ployamide on Polysulfone Support, *2 CA = Cellulose acetate

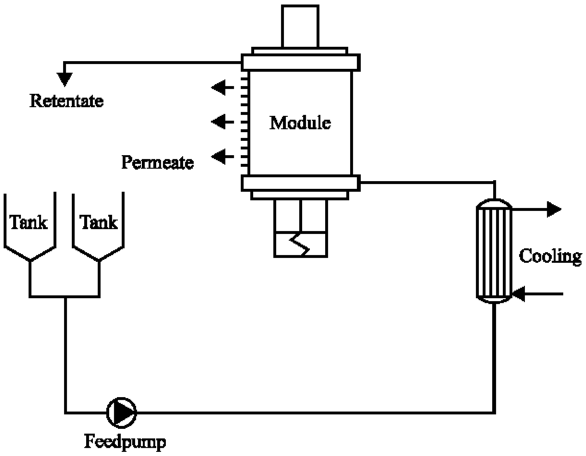

Fig. 4: Laboratory membrane unit for nanofiltration

Schematic illustration of the pilot used in this research in shown in Fig. 4. Experiments with the different solutions were performed in recirculation mode. Both permeate and retentate were returned to the feed vessel. The experiments were carried out with maximum circulation flow of 10 l min^{-1} at 20°C at pressures ranging from 2.5 to 40 bar. The composition of retentate and permeate was determined at each pressure level.

The cleaning procedure of the laboratory unit was as follows. After removing the feed from the module, the system was rinsed with water. After rinsing, cleaning took place (30-35°C, 5 bar) with 0.5% Ultrasil 50. After 45 min of cleaning the module was rinsed again with water and water flux was measured to verify cleaning efficiency. In all cases original water flux was restored.

RESULTS AND DISCUSSION

In Fig. 5 the effect of temperature on the water permeability of the four different membranes is shown. Theoretically temperature should hardly affect the product of permeability (Aw) and viscosity (μ) in this temperature range. Figure 5 it can be seen that this is indeed the case for all membranes. The results from the NF 40 and the HC50 membranes are virtually the same, which is expected because both membranes were produced by the same method.

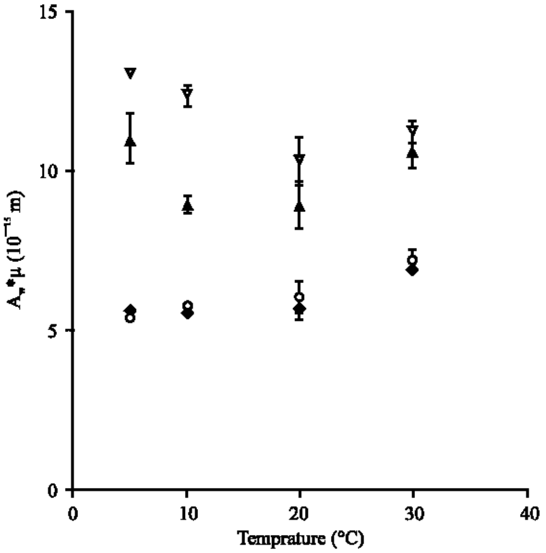

Fig. 5: Effect of temperature on the solvent permeability for four different NF-membranes

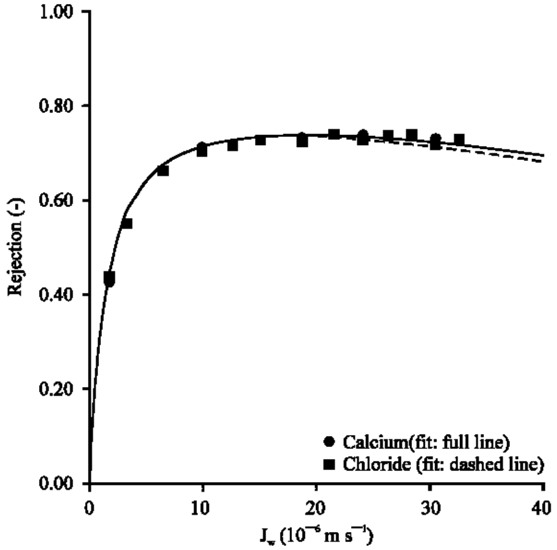

Fig. 7: Rejection-flux curve for calcium and chloride (feed 2)

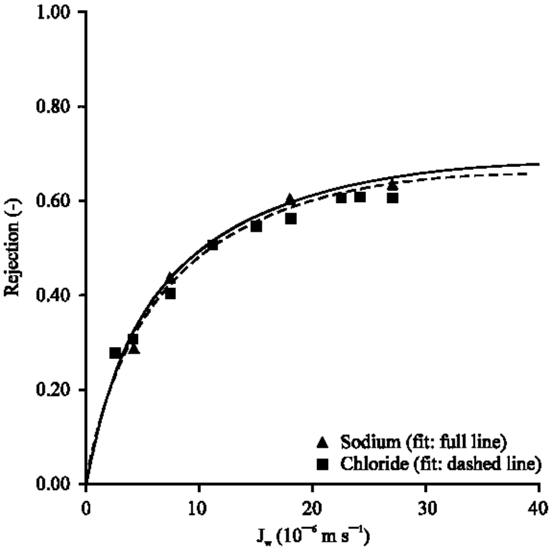

Fig. 6: Rejection-flux curve for sodium and chloride (feed 1)

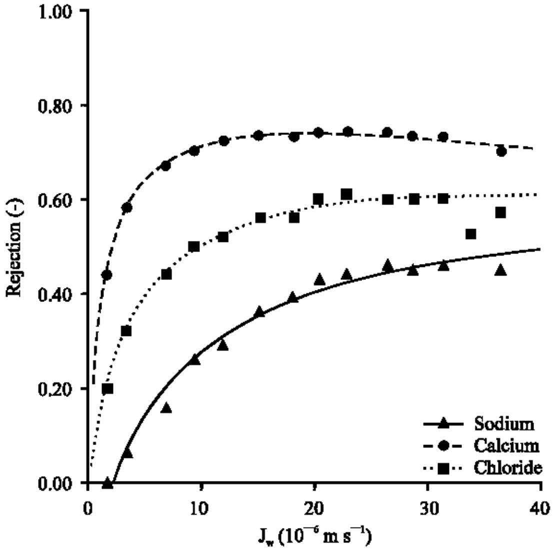

Fig. 8: Rejection-flux curve for sodium, calcium and chloride (feed 3)

In Fig. 6-9 the rejection-flux curves for different samples are given.

The differences between rejection of Na^+ and Cl^- in Fig. 6. and Ca^{++} and Cl^- in Fig. 7 are marginal, which is according to expectations, because from the electroneutrality condition. The permeability of Ca^{++} is lower than the permeability of Na^+, while the reflection of Ca^{++} is higher than for Na^+.

Comparing Fig. 8 and 9, there is an unexpected increase in Ca^{++} rejection in the UF-permeate. The major reason for this increase is the availability of free Ca^{++} in the retentate. One of the striking features in all the rejection flux curves is the low solute rejection at low flux levels. In this range of all curves the correlation between rejection and flux is almost linear. This indicates that diffusive transport is the most important transport mechanism at low flux. At higher flux, rejection reaches a constant value, indicating that convective transport is then the major transport mechanism. Not taking in to account the large membrane area that is needed, the best removal of solutes (in our case salts) takes place at a low

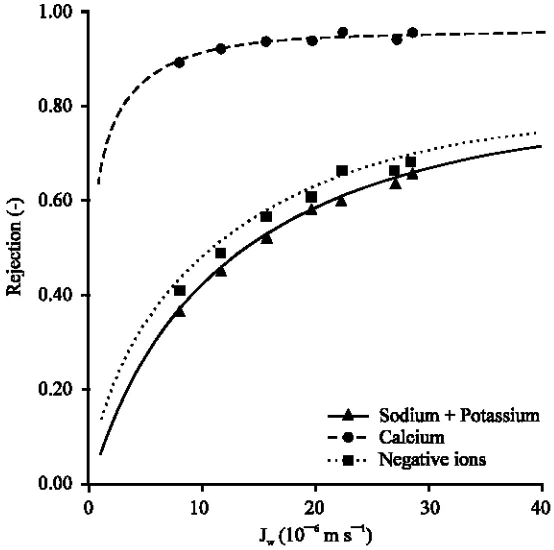

Fig. 9: Rejection versus flux of cations and anions during NF of UF permeate at pH 4.6

Table 3: Composition of concentration NF-permeate of UF-whey permeate

	Permeate concentration			
Component	NF40	HC50	FE700-004	CA960
Sodium (mg g^{-1})	154.00	117.00	176.00	53.00
Potassium (mg g^{-1})	1012.00	781.00	1002.00	406.00
Calcium (mg g^{-1})	11.00	5.00	6.00	10.00
Phosphate (mg g^{-1})	315.00	360.00	91.00	119.00
Chloride (mg g^{-1})	1008.00	771.00	1105.00	436.00
Citrate (mg g^{-1})	46.00	40.00	0.00	57.00
Sulfate (mg g^{-1})	0.00	0.00	0.00	7.00
Nitrate (mg g^{-1})	28.00	22.00	35.00	13.00
Lactose (%)	0.11	0.03	0.00	0.17

flux level (thus at a low effective trans membrane pressure).

The characteristics of NF-treated permeate is shown in Table 3.

CONCLUSIONS

The salt transport during NF of binary salt solutions like NaCl and $CaCl_2$, ternary salt solutions like (NaCl + $CaCl_2$) and UF-permeate were monitored in this experiment. The results can be described by a model derived from the extended Nernst-Planck equation.

Regarding a complex multi-component salt system like UF-whey permeate as a three-component system with monovalent and divalent cations and anion equivalent charges seems to be a useful approach for evaluating membrane performance and predicting salt transport for similar systems. The salt rejections increase with increasing flux, indicating the importance of the contribution of diffusive transport at low flux and convective transport at high flux. This finding may help to increase demineralization during NF dairy systems. As a suggestion for further work, this study could be done in different pHs and be compared with the current results. Theoretically, different models like, Theorell Meyers Siever (TMS), Space Charge (SC) and Generalised Maxwell Stefan (GMS) could be used instead of Extended Nernst Planck (ENP) model.

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