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# Direct contact membrane distillation for the concentration of saline dairy effluent



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## ABSTRACT

The ability of direct contact membrane distillation to concentrate the waste effluent from salty whey, a by-product from the cheese making industry has been investigated. The effect of trace protein in the feed, cross-flow velocity and feed acidity were the factors examined. Flat Sheet PTFE membranes of nominal pore sizes 0.05, 0.22 and 0.45  $\mu$ m were utilised. A decline in feed flux in the presence of trace protein in the feed was observed, but liquid penetration through the membrane could still be prevented by utilization of a membrane of smaller pore size, to achieve a final total solids concentration of  $\pm 30\%$  w/ w with water recovery from 37 to 83 %. The pressure-drop across the channel length was also predicted accounting for the feed spacer. To increase the channel length up to 1 m will require operation using the smallest pore size of 0.05  $\mu$ m, unless very low cross-flow velocities are used. The fouling of the membrane is primarily governed by precipitation of a calcium phosphate salt. However, operation at low pH does not improve the flux or the final salt concentration significantly.

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## 1. Introduction

The increasing salinity of inland waterways is a major environmental issue in Australia and the generation of saline waste in dairy processing adds to this problem. The dairy industry is one of the largest rural industries in Australia and produces 10.3 GL/y of high salinity effluent (Wilkinson et al., 2004). Salty whey, a byproduct of hard cheese manufacture, contributes to this effluent load. The salt concentration ranges from 3 to 19% w/w, while the protein and fat concentration is commonly lower than 1% w/w (Blaschek et al., 2007). Through the use of low energy ultrafiltration (UF), the traces of fat and protein can be completely removed, yet a large volume of saline waste is still generated. This is a paramount issue since direct discharge of this salty effluent to the environment would disrupt the ecological balance. As a consequence, the effluent must either be diluted with less salty streams before discharge, or alternatively concentrated and then evaporated to dry solids within a waste treatment pond.

Reverse osmosis (RO) is widely utilized for the concentration step. Nonetheless, as the osmotic pressure increase, the efficiency

\* Corresponding author. E-mail address: sandraek@unimelb.edu.au (S. Kentish). of RO decreases. An emerging alternative is membrane distillation (MD) (Alkhudhiri et al., 2012a, 2012b; Alklaibi and Lior, 2005; Angela et al., 2011; Bandini et al., 1992; Bandini and Sarti, 1999; Curcio and Drioli, 2005; Fane et al., 1987; Findley, 1967; Gostoli et al., 1987; Gryta, 2012; Gryta et al., 2013; Hausmann et al., 2012, 2013; Hsu et al., 2002; Khayet et al., 2007; Khayet Souhaimi and Matsuura, 2011, Schofield et al., 1990; Tomaszewska, 2000; Van der Bruggen and Vandecasteele, 2002; Yun et al., 2006). The use of membrane distillation at a large scale is still rare; however it has the potential to concentrate the waste stream to higher solid levels than an RO process.

An advantage of this process compared to conventional pressure-driven filtration is that it requires only low pressures and moderate temperatures and thus can utilize a membrane with lower mechanical strength (Burgoyne and Vahdati, 2000; Fritzmann et al., 2007; Khawaji et al., 2008). Furthermore, MD can theoretically achieve 100% rejection of salts and particulates i.e. only volatile solvents in the vapour form are able to travel through the membrane, Finally, as the process operates at only moderate temperatures it can be readily driven by waste heat or solar energy.

Unlike pressure-driven filtration, the mass flux across the membrane (N) in MD is driven by the water vapour partial pressure difference across the membrane ( $\Delta P_m$ ), which is essentially influenced by the temperature gradient on the membrane surfaces





Nomenclature		u	Tangential velocity m/s
		V	Tank Volume L
В	Pore geometric factor -	μ	Viscosity Pa.s
С	Molar concentration mol/m <sup>3</sup>	γ	Liquid surface tension N m $^{-1}$
с	Salt concentration g/L NaCl equivalent	$\Delta P_m$	Pressure drop across the membrane Pa
Ctd	Total drag	θ	Liquid-surface contact angle $^\circ$
d <sub>H</sub>	Hydraulic diameter m	ρ	Density Kg $m^{-3}$
f	Fanning friction factor		
k <sub>cp</sub>	Mass transfer coefficient ms <sup>-1</sup>	Subscripts	
К <sub>т</sub>	Overall mass transfer coefficient ms <sup>-1</sup>	b	Bulk
LEP	Liquid entry pressure Pa	d	Permeate tank
Lmesh	Mesh element length m	f	Final
Ν	Mass flux kg m <sup>-2</sup> s <sup>-1</sup>	F	Feed
Re	Reynolds number -	i	Initial
r <sub>pore</sub>	Pore radius m	m	Membrane surface
Sc	Schmidt number -	р	Permeate
Sh	Sherwood number -		

(Equation (1)).

$$N = K_m \Delta P_m \tag{1}$$

where  $K_m$  is the mass transfer coefficient of water vapour through the membrane itself. The vapour pressure of water on the feed side is also affected by the solute concentration. With a significant increase in solute (salt) concentration, the water activity will fall resulting in a reduction of this partial pressure and thus a loss in partial pressure driving force.

The MD process uses a porous membrane with pore sizes comparable to that of microfiltration or ultrafiltration (Laganà et al., 2000; Lawson and Lloyd, 1997; Martínez-Díez and Vázquez-González, 1999; Martínez-Díez et al., 1998; Zhang et al., 2010). The membrane pores must not be wetted by the feed solution. Pore wetting leads to deterioration of permeate quality as liquid crosses the interface. The susceptibility towards pore wetting can be characterised by the liquid entry pressure (LEP) of the membrane. This is the pressure where liquid first penetrates through the membrane pores. It can be measured experimentally, but also predicted by the Laplace—Kelvin equation as follows:

$$LEP = -\frac{2B\gamma_L \cos\theta}{r_{pore}}$$
(2)

Where B is a factor representing the pore geometry (0 < B < 1, unity for a cylindrical pore),  $\gamma_L$  is the liquid surface tension,  $\theta$  the liquid/ membrane contact angle and  $r_{pore}$  is the maximum pore radius (Khayet Souhaimi and Matsuura, 2011; Lawson and Lloyd, 1997). As indicated by Equation (2), pore wetting can be minimised by using a hydrophobic membrane material of a uniform, small pore size (Khayet and Matsuura, 2011; Lawson and Lloyd, 1997; Mulder, 1996; Simioni, 2010).

Furthermore, polarization phenomena (both temperature and concentration) can reduce the efficiency of heat and mass transfer. Temperature polarization causes a temperature reduction from the bulk feed (hot side) to the feed side membrane surface, while concentration polarization causes the salt concentration to build up within this boundary layer. The mass transfer coefficient within the concentration polarisation boundary layer ( $k_{cp}$ ) is governed by the cross-flow velocity (CFV). This mass transfer coefficient can be estimated using a dimensionless mass-transfer correlation (Equation (4)).

$$Sh = ARe^{x}Sc^{y}\left(\frac{d_{H}}{L_{mesh}}\right)^{z}$$
(3)

For a slit channel with net spacer equipped, A = 0.664, x = 0.55, y = 0.33, z = 0.5 (Da Costa et al., 1994). The Sherwood number,  $Sh = k_{cp}d_H/D$  where D is the diffusion coefficient and d<sub>H</sub> is the hydraulic mean diameter. The Reynolds number,  $Re = \rho u d_H/\mu$ , the Schmidt Number,  $Sc = \mu/\rho D$ , and u = CFV.  $L_{mesh}$  is the length of the mesh element.

This estimate of the mass transfer coefficient can be used to determine the polarisation ratio, which relates the bulk solute concentration ( $C_b$ ) to that at the surface of the membrane ( $C_m$ ), given the solution density ( $\rho$ ) and the permeate volumetric flux( $j_v$ ) is known:

$$\frac{C_m}{C_b} = \exp\left(\frac{j_\nu}{\rho k_{cp}}\right) \tag{4}$$

The membrane polymer matrix itself can also act to conduct heat from the hot to the cold side reducing energy efficiency. To reduce this, a membrane of high porosity, low tortuosity and low polymer thermal conductivity (Khayet Souhaimi and Matsuura, 2011; Souhaimi and Matsuura, 2011) should be used. To satisfy these characteristics, hydrophobic microporous materials such as polypropylene (PP), polydivinyldenedifluoride (PVDF) and polytetrafluoroethylene (PTFE) form the most adequate membranes.

The focus of this study is to assess the ability of direct contact membrane distillation to concentrate the saline waste stream generated as a by-product of cheese making and containing high concentrations of salt and lactose; and trace amounts of protein. The effect of trace protein on the permeate flux and pore breakthrough is investigated. As a hydrophobic membrane material is utilized, protein analyses are crucial since protein will be strongly attracted to the membrane surface and this could lead to liquid penetration into the pores, or fouling of the membrane surface. Previous work has shown that mineral scaling can also be a significant issue in concentrated salt solutions with respect to both pore breakthrough and membrane fouling (Edwie and Chung, 2012). We investigate this issue by observing the system performance at low pH, as this approach is known to eliminate calcium salt scaling in dairy systems (Rice et al., 2009a). Furthermore, the impact of cross-flow velocity on flux, pore breakthrough and pressure drop is considered.

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