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MODELLING OF A DIRECT OSMOTIC CONCENTRATION MEMBRANE SYSTEM

A thesis presented in partial fulfilment of the requirements for the degree of

Doctor of Philosophy

in Food Engineering

at Massey University

Marie Wong

1997
ABSTRACT

Direct osmotic concentration (DOC) is a novel continuous membrane process. Two co-current streams, separated by a semi-permeable membrane, are recycled through a DOC module. The turbulent-flow dilute juice stream is concentrated by osmotically extracting water across the membrane into a laminar-flow, concentrated osmotic agent (OA) stream. The semi-permeable membrane is asymmetric, with a non-porous active layer (15 μm) and a porous support layer (150 μm). Membrane solute rejection was greater than 99%. Normal operation orients the active layer towards the juice stream.

For this study, water (osmotic pressure = 0) was used in the juice channel. The relationship between water flux rate and the osmotic pressure of the bulk OA stream was asymptotic, reaching a maximum flux of $1.75 \times 10^{-3} \text{kg m}^{-2} \text{s}^{-1}$, when using fructose OA at 15 MPa osmotic pressure and 20°C.

Flux rates doubled when NaCl replaced fructose as OA. A doubling in temperature to 40°C resulted in a 50% increase in flux rate. OA solution properties, particularly viscosity and factors affecting diffusion coefficients had a strong influence on flux rates.

When the membrane was reversed, with the active layer facing the OA channel and the support layer filled only with water, flux rates were 40 to 60% higher than the normal orientation.

There were three resistances to water flow associated with: osmosis across the membrane active layer ($R_1$); diffusion and porous flow across the support layer ($R_2$), and, diffusion across the boundary layer in the OA channel ($R_3$). For fructose OA at 0.50 g (g solution)$^{-1}$ (osmotic pressure = 15 MPa), $R_1$ contributed 9% of the total resistance to water flux in the DOC module, $R_2$ contributed 64% and $R_1$ contributed 27%. For an iso-osmotic concentration of NaCl OA (0.15 g (g solution)$^{-1}$) the relative resistances were: $R_1 = 17\%$, $R_2 = 44\%$ and $R_3 = 39\%$. It was clear that the water flux from the dilute to concentrated stream was more strongly influenced by the support membrane and OA solution properties than the active semi-permeable membrane itself. This accounted for the asymptotic relationship between bulk OA stream properties and flux rate.

The mathematical model successfully incorporated these resistances and solution properties. Data calculated using this model agreed well with experimental results.
ACKNOWLEDGEMENTS

I would like to express my sincere thanks and appreciation to all my supervisors Professors Ray Winger, Robert McKibbin and Ronald Wrolstad, for all their help, support and guidance throughout this research. Each supervisor helped me in very different ways towards completing this research and I have gained a lot from each.

I would like to thank Osmotek Inc. for supplying the DOC equipment and membranes. Thanks to Robert Salter, Keith Lampi, Dr Ed Beaudry, Dr Jack Herron and Carl Jochums for their help and generosity.

I would like to thank The Horticulture & Food Research Institute of NZ Ltd (HortResearch) for supporting me and providing financial assistance. I would also like to thank FRST for funding the programme through which this research was funded. I would like to thank Frucor Processors Ltd for supplying apple juice for trials.

Thanks to all members of the Food Science Group who were formally from the Food Science & Technology Group, HortResearch for their support. Special thanks to Jeff Keene for assistance on the HPLC.

Thanks to Paul Sutherland and Dr Ian Hallet for microscopy work. Thanks to the following people for letting me use their equipment: Dr Ron Wong, Crop & Food Research; Dr Don Stanton, ESR; Stephanie Parkes and Dr Ross Beever, LandCare.

Thanks to the Department of Food Science & Technology, Oregon State University (OSU), especially to those who helped me during my two visits to OSU and made the work more enjoyable and successful. Special thanks to Bob Durst and Ken Stewart.

I would like to thank John Maindonald, University of Newcastle (formally HortResearch), for his help and advice on the statistical analyses completed. I would also like to thank Dr Alan Easteal, University of Auckland, for his advice and discussions on diffusion coefficients.

Special thanks to Dr Ian Boag, Alicia Boag, Catriona Jenkinson, Dr Sam Peterson, Jane La Riviere and Jason Wallis who I am indebted to for their generosity and friendship. Special thanks to my parents for their continual support and encouragement.

Last but not least thanks to Phil for his patience, support and encouragement.
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LIST OF ABBREVIATIONS

am - active membrane layer
bl - boundary layer in OA channel
Case 1 - active layer facing the juice circuit, support layer facing the OA circuit and a fully-developed velocity boundary layer in OA flow channel
Case 2 - active layer facing the juice circuit, support layer facing the OA circuit, no velocity boundary layers in OA channel
Case 3 - active layer facing the OA circuit, support layer facing the juice circuit and a fully-developed velocity boundary layer in OA flow channel

DOC - direct osmotic concentration
ED - electrodialysis
fdbl - fully-developed boundary layer
Hg - mercury
HPLC - high performance liquid chromatography
GS - gas separation
KCl - potassium chloride
MD - membrane distillation
MF - microfiltration
NaCl - sodium chloride
NF - nanofiltration
NZ - New Zealand
OA - osmotic agent
OA\textsubscript{IN} - flow rate of OA into module
OA\textsubscript{OUT} - flow rate of OA out of module
OD - osmotic distillation
p - level of significance of a statistical test
PRO - pressure retarded osmosis
PV - pervaporation
PVC - polyvinyl chloride
RO - reverse osmosis
sm - support membrane layer
UF - ultrafiltration
USA - United States of America
vs - versus
# LIST OF NOMENCLATURE

- \( \alpha_i \) - activity of solvent \( i \) (always \( \leq 1.0 \))
- \( A_m \) - membrane area, \( \text{m}^2 \)
- \( c_i \) - mass concentration of component \( i \), \( \text{kg} \ \text{m}^{-3} \)
- \( C \) - membrane constant, \( \text{kg} \ \text{m}^{-2} \ \text{s}^{-1} \ \text{Pa}^{-1} \)
- \( d_s \) - thickness of membrane active layer, \( \text{m} \)
- \( d_e \) - thickness of membrane support layer, \( \text{m} \)
- \( D_{i,m} \) - diffusion coefficient of component \( i \) within the membrane, \( \text{m}^2 \ \text{s}^{-1} \)
- \( D_{i,A} \) - self diffusion coefficient, \( \text{m}^2 \ \text{s}^{-1} \)
- \( D_{i,t} \) - tracer diffusion coefficient, \( \text{m}^2 \ \text{s}^{-1} \)
- \( D_{i,B} \) - binary diffusion coefficient of component \( A \) in a mixture of \( A \) and \( B \), \( \text{m}^2 \ \text{s}^{-1} \)
- \( D_{i,B}^{\alpha} \) - binary diffusion coefficient of solute at infinite dilution at \( T^\alpha \ \text{C} \), \( \text{m}^2 \ \text{s}^{-1} \)
- \( D_{ij} \) - effective diffusion coefficient at concentration \( Y \), \( \text{m}^2 \ \text{s}^{-1} \)
- \( d_f \) - degrees of freedom
- \( E_a \) - activation energy, \( \text{J} \ \text{mol}^{-1} \)
- \( k[x] \) - expected value of \( x \)-coefficient
- \( F \) - force, \( \text{N} \)
- \( F(Y) \) - integral function of \( Y \)
- \( g \) - gravitational acceleration, \( \text{m} \ \text{s}^{-2} \)
- \( G \) - Gibbs free energy, \( \text{J} \)
- \( G_i \) - pressure gradient in the \( x \) direction, \( dP/dx \)
- \( h \) - equivalent flow channel height; distance between membrane and OA wall when fully deflected, \( \text{m} \)
- \( I \) - integral function describing velocity and concentration profiles across the OA channel
- \( l \) - correction factor for \( l \)
- \( j \) - molar diffusion flux, \( \text{mol} \ \text{m}^{-2} \ \text{s}^{-1} \)
- \( k \) - mass transfer coefficient, \( \text{m} \ \text{s}^{-1} \) [Section 2.4.2]
- \( k \) - Boltzmann constant, \( 1.38 \times 10^{-23} \ \text{J} \ \text{K}^{-1} \)
- \( k_f \) - permeability of porous media, \( \text{m}^2 \)
- \( l_c \) - characteristic length, \( \text{m} \)
- \( l_o \) - length of membrane arc between two membrane support bars, \( \text{m} \)
- \( l_m \) - length of OA flow channel, \( \text{m} \)
- \( l' \) - entry length before fully-developed flow, \( \text{m} \)
<table>
<thead>
<tr>
<th>Symbol</th>
<th>Definition</th>
</tr>
</thead>
<tbody>
<tr>
<td>$m$</td>
<td>solution molality, mol (kg solvent)$^{-1}$</td>
</tr>
<tr>
<td>$m, n$</td>
<td>number of periods in Fourier series</td>
</tr>
<tr>
<td>$m_i$</td>
<td>mass diffusive flux of component $i$, kg m$^{-2}$ s$^{-1}$</td>
</tr>
<tr>
<td>$m_w$</td>
<td>water mass flux rate, kg m$^{-2}$ s$^{-1}$</td>
</tr>
<tr>
<td>$m_{w}(x,y,z)$</td>
<td>water mass flux rate at position $(x,y,z)$, kg m$^{-2}$ s$^{-1}$</td>
</tr>
<tr>
<td>$M$</td>
<td>solution molarity, mol l$^{-1}$</td>
</tr>
<tr>
<td>$M_s$</td>
<td>molecular weight of solvent, g mol$^{-1}$</td>
</tr>
<tr>
<td>$M_j$</td>
<td>molecular weight of solute, g mol$^{-1}$</td>
</tr>
<tr>
<td>$n$</td>
<td>number of samples for each mean</td>
</tr>
<tr>
<td>$n$</td>
<td>number of horizontal flow channels in OA plate [Section 5.4.3]</td>
</tr>
<tr>
<td>$N_i$</td>
<td>number of moles of component $i$ (normally the solvent)</td>
</tr>
<tr>
<td>$N_j$</td>
<td>number of moles of component $j$ (normally the solute)</td>
</tr>
<tr>
<td>$Nu$</td>
<td>Nusselt number</td>
</tr>
<tr>
<td>$p$</td>
<td>pressure, Pa</td>
</tr>
<tr>
<td>$p_i^*$</td>
<td>vapour pressure of pure solvent $i$, Pa</td>
</tr>
<tr>
<td>$p_i$</td>
<td>partial vapour pressure of solvent $i$, in solution, Pa</td>
</tr>
<tr>
<td>$\Delta p$</td>
<td>hydraulic pressure difference, Pa</td>
</tr>
<tr>
<td>$p, q$</td>
<td>number of periods in Fourier series</td>
</tr>
<tr>
<td>$q_{aw}$</td>
<td>water mass flux rate per unit area along OA flow channel, kg m$^{-2}$ s$^{-1}$</td>
</tr>
<tr>
<td>$q_{af}$</td>
<td>fructose mass flux rate per unit area along OA flow channel, kg m$^{-2}$ s$^{-1}$</td>
</tr>
<tr>
<td>$Q_m$</td>
<td>total mass flow along OA flow channel, kg s$^{-1}$</td>
</tr>
<tr>
<td>$Q_{mc}$</td>
<td>total mass flow at channel entry, kg s$^{-1}$</td>
</tr>
<tr>
<td>$Q_{mf}$</td>
<td>total mass flow of fructose along flow channel, kg s$^{-1}$</td>
</tr>
<tr>
<td>$r$</td>
<td>pore radius, m</td>
</tr>
<tr>
<td>$R$</td>
<td>gas constant, 8.314 J K$^{-1}$ mol$^{-1}$ $\equiv$ 8.314 m$^3$ Pa K$^{-1}$ mol$^{-1}$</td>
</tr>
<tr>
<td>$R$</td>
<td>resistance, m$^2$ s Pa kg$^{-1}$</td>
</tr>
<tr>
<td>$R_1$</td>
<td>resistance in the active membrane layer, m$^2$ s Pa kg$^{-1}$</td>
</tr>
<tr>
<td>$R_2$</td>
<td>resistance in the porous support layer, m$^2$ s Pa kg$^{-1}$</td>
</tr>
<tr>
<td>$R_3$</td>
<td>resistance in the velocity boundary layer, m$^2$ s Pa kg$^{-1}$</td>
</tr>
<tr>
<td>$% R$</td>
<td>percentage rejection</td>
</tr>
<tr>
<td>$Re$</td>
<td>Reynolds number</td>
</tr>
<tr>
<td>$RSE$</td>
<td>residual standard error</td>
</tr>
<tr>
<td>$RSS$</td>
<td>residual sum of squares</td>
</tr>
<tr>
<td>$s$</td>
<td>pooled estimate of standard deviation</td>
</tr>
<tr>
<td>$S$</td>
<td>osmosity, molar concentration of NaCl, mol l$^{-1}$</td>
</tr>
<tr>
<td>$Sc$</td>
<td>Schmidt number</td>
</tr>
<tr>
<td>$sd$</td>
<td>standard deviation</td>
</tr>
<tr>
<td>$SE$</td>
<td>standard error</td>
</tr>
</tbody>
</table>
\( SE[x] \) - standard error of x-coefficient  
\( SEM \) - standard error about the mean  
\( Sh \) - Sherwood number  
\( t \) - time, s  
\( T \) - absolute temperature, K  
\( u \) - velocity in the x direction, m s\(^{-1}\)  
\( u(x,y,z) \) - velocity in the x direction at position \((x,y,z)\), m s\(^{-1}\)  
\( U \) - bulk free-stream velocity in x direction, m s\(^{-1}\)  
\( v \) - velocity in the y direction, m s\(^{-1}\)  
\( \vec{v} \) - velocity vector  
\( \vec{v}_0 \) - superficial or Darcy velocity, volume of flow through a unit cross-sectional area of the solid plus fluid, m\(^3\) m\(^{-2}\) s\(^{-1}\)  
\( V_w \) - volumetric water flux rate, m\(^3\) m\(^{-2}\) s\(^{-1}\)  
\( w \) - width between two adjacent membrane support bars, width of OA flow channel, m  
\( \omega \) - velocity in the z direction, m s\(^{-1}\)  
\( x \) - horizontal distance parallel to the membrane, m  
\( x \) - coordinate  
\( x_i \) - mole fraction of component A  
\( y \) - distance perpendicular to the membrane (across membrane or away from the membrane), m  
\( y \) - coordinate  
\( Y \) - solute mass fraction, solute mass fraction in OA circuit, g (g solution\(^{-1}\))  
\( Y(x,y,z) \) - solute mass fraction at position \((x,y,z)\), g (g solution\(^{-1}\))  
\( z \) - coordinate  
\( \nabla \) - vector differentiation operator

**Greek symbols**  
\( \alpha \) - power term for viscosity in relationship with diffusion coefficient  
\( \alpha \) - power term for velocity profile equation  
\( \gamma \) - activity coefficient  
\( \delta \) - velocity boundary layer thickness, m  
\( \delta(x) \) - velocity boundary layer thickness as a function of x, m  
\( \delta_c \) - concentration boundary layer thickness, m  
\( \Delta \) - membrane deflection between two membrane support bars, m  
\( \varepsilon \) - porosity
<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\kappa$</td>
<td>proportionality constant</td>
</tr>
<tr>
<td>$\lambda$</td>
<td>membrane thickness, m</td>
</tr>
<tr>
<td>$\mu_i$</td>
<td>chemical potential of component $i$, J mol$^{-1}$</td>
</tr>
<tr>
<td>$\mu$</td>
<td>fluid or solution viscosity, kg m$^{-1}$ s$^{-1}$</td>
</tr>
<tr>
<td>$\mu_0$</td>
<td>absolute viscosity of solvent at T °C, kg m$^{-1}$ s$^{-1}$</td>
</tr>
<tr>
<td>$\mu(Y)$</td>
<td>viscosity of solution at concentration $Y$, kg m$^{-1}$ s$^{-1}$</td>
</tr>
<tr>
<td>$\nu$</td>
<td>kinematic viscosity, m$^2$ s$^{-1}$</td>
</tr>
<tr>
<td>$\pi$</td>
<td>osmotic pressure, MPa</td>
</tr>
<tr>
<td>$\pi_A$</td>
<td>osmotic pressure of solution $A$, MPa</td>
</tr>
<tr>
<td>$\pi(Y)$</td>
<td>osmotic pressure of solution at concentration $Y$, MPa</td>
</tr>
<tr>
<td>$\Delta\pi$</td>
<td>osmotic pressure difference, MPa</td>
</tr>
<tr>
<td>$\rho$</td>
<td>fluid or solution density, kg m$^{-3}$</td>
</tr>
<tr>
<td>$\rho(Y)$</td>
<td>density of solution at concentration $Y$, kg m$^{-3}$</td>
</tr>
<tr>
<td>$\tau$</td>
<td>tortuosity</td>
</tr>
<tr>
<td>$\upsilon_i$</td>
<td>partial molar volume of solvent $i$, m$^3$ mol$^{-1}$</td>
</tr>
<tr>
<td>$\phi$</td>
<td>osmotic coefficient</td>
</tr>
<tr>
<td>$\psi$</td>
<td>water potential, J kg$^{-1}$</td>
</tr>
</tbody>
</table>

**Subscripts**

0 - position at the interface between the support layer and the velocity boundary layer

1 - position at the surface of the active layer on OA side

$a$ - active layer

$C$ - bulk OA free-stream solution and bulk OA free-stream solution at flow channel entry

$f$ - fructose

$i$ - component in solution, normally the solvent

$j$ - juice circuit solution

$m$ - mass flow

$m$ - membrane

$m, n$ - number of periods in Fourier series [Equation (8.24)]

OA - osmotic agent solution

$s$ - support layer

$w$ - water