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Analogy of Mass and Heat Transfer in Osmotic Membrane Distillation Process

A Thesis

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By

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دِسم الله الرحمن الرحيم متالوا شيعانك لا عِلمَ لنا إلاَّ ما عَلَّمة ذَا إِذَا مَ أَذَبَّ مَا عَلَّمة ذَا إِذَا مَ الْحَلِيم الحكيم حدق الله العظيم سورة البقرة (٢٢

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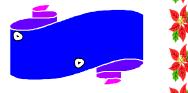
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Dedication

Especially Dedicated To....

My lovely father and mother My brothers and sisters My husband and Children Ibrahim and Masarra Your care and love always surround and

Encourage me



Acknowledgment

First of all, I thank God who offered me patience, power and faith in a way that words cannot express.

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Abstract

Osmotic membrane distillation is a novel membrane process for the removal of water from dilute aqueous solutions, such as liquid foods or natural colors, concentrating them, while retaining the organoleptic and nutritional properties. Experiments were performed with real system (pure water) in a flat sheet membrane module type Polytetrafluoroethylene (PTFE) macroporous layer supported by a polypropylene (PP) net (TF200 from pall-Gelman). The effect of concentration of osmotic agent solution on the transmembrane flux was evaluated in case of calcium chloride (1-5) M and sodium chloride (2-5) M. For both the osmotic agents, higher transmembrane flux was observed at maximum osmotic agent concentration. In comparison with sodium chloride, higher transmembrane flux was observed in case of calcium chloride. The feed and osmotic agent side mass transfer resistances were estimated based on classical empirical correlation of dimensionless numbers. Molecular and Knudsen diffusion mechanisms were tested to model the vapour transport across the membrane. When using the global structural characteristics specified by the membrane manufacturer, the mass transfer mechanism was found to be in the molecular diffusion region when Knudsen number < 0.01 when it is estimated. The heat transfer associated with water transport is integrated into the mass transfer equations. The flux across the membrane during the process was predicted using resistances-in-series model. The experimental values were found to correlate well with the predicted values.



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<u>Nomenclature</u>

Symbol	Definition	Unit
a	Activity	
С	Solute molar concentration	$mol l^{-1}$
C _p	Heat capacity	$J kg^{-1} K^{-1}$
d	Diameter	m
D	Diffusion coefficient	$\frac{\text{m}}{\text{m}^2 \text{ s}^{-1}}$
h	Heat transfer coefficient	W/m ² .K
J	Flux	$1 \text{m}^{-2} \text{h}^{-1}$
k	Mass transfer coefficient	$\frac{\text{m s}^{-1}}{\text{kgm}^{-2} \text{ h}^{-1} \text{ Pa}^{-1}}$
K	Mass transfer coefficient	$kgm^{-2}h^{-1}Pa^{-1}$
K _B	Boltzmann constant	$1.3807 * 10^{-23}$
		$\mathbf{J}\mathbf{K}^{-1}$
k ^T	Thermal conductivity	$Wm^{-1} K^{-1}$
L	Length of the fluid circulation channel	m
М	Molecular weight	kg mol ⁻¹
N	Vapour flux, mass	$\frac{\text{kgm}^{-2} \text{h}^{-1}}{\text{molm}^{-2} \text{s}^{-1}}$
	Molar	$molm^{-2} s^{-1}$
	or volume	$m^3 m^{-2} s^{-1}$
Р	Pressure	Pa
P*	Saturation vapour pressure	Pa
Q	Heat flux	Wm ⁻²
r	Pore radius	m
R	Universal gas constant	8.314 JK ⁻¹
		mol^{-1}
Т	Temperature	°С. К
u	Mean brine circulation velocity	$m s^{-1}$
Х	mass fraction (w/w%) or molar fraction	
	(Mol/mol %)	
Y _{ln}	Mole fraction of air (log-mean)	



Greek symbols

3	volume porosity factor	
δ	thickness	m
Δ	difference	
γ	activity coefficient	
λ	mean molecular free path	m
μ	liquid dynamic viscosity	Pa s
χ	tortuosity factor	
ρ	liquid density	kgm ⁻³
σ	mean collision diameter	m
Θ	thermal effect factor	
Ψ	association factor, 2.26 for water	
Ω	collision integral	A°

<u>Groups</u>

Kn	Knudsen number
Nu	Nusselt number
Pr	Prandtl number
Re	Reynolds number
Sc	Schmidt number
Sh	Sherwood number



<u>Subscripts</u>

f	feed
h	hydraulic
L	liquid
m	membrane
р	permeate (brine)
S	solute
W	water or vapour

<u>Superscripts</u>

b	bulk location or exponent
K	Knudsen diffusion
m	membrane location
М	molecular diffusion
t	total

Chapter One Introduction

1.1 Introduction

Filtration is defined as the separation of two or more components from a fluid stream based primarily on size differences. In conventional usage, it usually refers to the separation of solid immiscible particles from liquid or gaseous streams. Membrane filtration extends this application further to include the separation of dissolved solutes in liquid streams and for separation of gas mixtures [1].

Separation of the mixture associated with membrane is known as membrane separation where the membrane acts as a selector that permits some components in the mixture to pass through, while other components are retained. The membrane in most cases is a thin, porous or nonporous polymeric film, or may be ceramic or mental materials, or even a liquid or gas. The selectivity of the membrane mainly depends on its structure and properties of the membrane material and the components in the mixture. Unlike conventional filtration process applied only to solid-liquid mixture, membrane separation is capable of the separation of homogeneous mixtures that are traditionally treated by distillation, absorption or extraction operations. The replacement of traditional separation processes with membrane separation has the potential to save large amounts of energy, since membrane process is mostly driven by pressure gradient or concentration gradient through the membrane. Although this replacement requires the production of high mass-transfer flux, defect-free, long-life membranes on a large scale and the fabrication of the membrane into compact, economical modules of high surface area per unit volume[2].

The membrane separation involves the process in which some components penetrate through the membrane and thus mass transfer occurs. Based on the



difference in driving forces of mass transfer and effective range of separation scale (from 0.1 nanometer to 10 microns, 5 orders span), the membrane 'family' includes more than 10 members, and most of them, such as reverse osmosis (RO), gas permeation (GP), microfiltration (MF), pervaporation (PV), have been accepted as the alternatives to some conventional separation techniques in industry. The related fields of membrane separation varies from the desalination of sea water or bitter water, concentration of solutions, waste water treatment to the recovery of valuable substance from solutions, the separation of gas mixture, etc[2].

Membrane distillation (MD) is a new comer of the membrane family. Although the discovery of MD phenomenon can be traced back to the 1960s, it hasn't received more attention until 1980s when membrane fabrication technique gained remarkable development. Today, MD is considered as a potential alternative to some traditional separation techniques, and is believed to be effective in the fields of desalination, concentration of aqueous solution, etc. That difference between MD and other membrane separation techniques is the driving force of mass transfer through the membrane. Unlike other members, MD is a thermally driven process. That's why it is denominated as a distillation process [2].

1.2 Osmotic Process

Osmotic is the transport of water across a selectively permeable membrane from a region of higher water chemical potential to a region of lower water chemical potential. It is driven by a difference in solute concentrations across the membrane that allows passage of water, but rejects most solute molecules or ions. Osmotic pressure (π) is the pressure which, if applied to the more concentrated solution, would prevent transport of water across the membrane. Forward osmosis uses the osmotic pressure differential $\Delta \pi$) across the membrane, rather than hydraulic pressure differential, as the driving force for transport of water through



the membrane. The forward osmosis process results in concentration of a feed stream and dilution of a highly concentrated stream [3].

1.3 Osmotic Distillation

Osmotic distillation is a separation process in which a liquid mixture containing a volatile component is contacted with a microporous, non liquid wettable membrane whose opposite surface is exposed to a second liquid phase capable of absorbing that component is nearing commercialization for the concentration of beverages and other liquid foodstuffs, and is under evaluation for the concentration of aqueous solutions of thermally labile pharmaceutical products and biological. Its primary advantage lies in its ability to concentrate solutes to very high levels at low temperature and pressure, with minimal thermal or mechanical damage to or loss of those solutes. The process also can enable the selective removal of a single volatile solute from aqueous solution using water as the extracting solvent [4].

Osmotic distillation (OD) promises to become an attractive complement or alternative to other thermal or low temperature separations techniques such as ultrafiltration (UF), reverse osmosis (RO), pervaporation, and vacuum freeze drying [4].

1.3.1 Process Fundamentals

Osmotic distillation OD, which is also called "isothermal membrane distillation," is a membrane transport process in which a liquid phase (most commonly an aqueous solution) containing one or more volatile components is allowed to contact one surface of a micro porous membrane whose pores are not wetted by the liquid, while the opposing surface is in contact with a second no wetting liquid phase (also usually an aqueous solution) in which the volatile



components are soluble or miscible. The membrane thereby functions as a vapor gap between the two liquid phases, across which any volatile component is free to migrate by either convection or diffusion. The driving potential for such transport is the difference in vapor pressure of each component over each of the contacting liquid phases. The mechanism is illustrated schematically in Figures (1.1) and (1.2). If the sole or primary volatile component in solution is the solvent, then evaporation of solvent from the solution of higher vapor pressure into that of lower vapor pressure will result in concentration of the former and dilution of the latter. Thus, the rate of transport of solvent from one liquid phase to the other will increase as the solvent vapor pressure over the receiving phase is reduced. If the solvent vapor pressure over the liquid being concentrated drops to a value equal to that over the receiving phase, no further transport will occur.

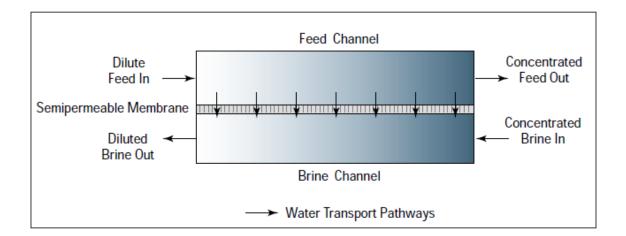


Figure (1.1) In osmotic distillation, a semi permeable membrane acts as a vapor gap that allows migration of volatiles in a single direction [4].



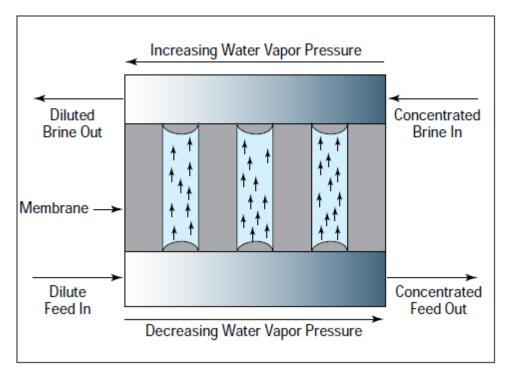


Figure (1.2) Mechanism of osmotic distillation through a micro porous hydrophobic membrane [4].

In most applications of practical interest, the solutions to be concentrated contain relatively low concentrations of nonvolatile solutes of moderate to high molecular weight (sugars, polysaccharides, carboxylic acid salts, proteins, and so on) which have limited stability to elevated temperatures and shear stresses. Because of the low osmotic activity of such solutes, the vapor pressure of water over such solutions is very nearly that of pure water, and decreases quite slowly with increasing solute concentration. Hence, if the receiving or "strip" solution on the opposite membrane face contains a high concentration of nonvolatile solute of high osmotic activity (meaning a solute of low equivalent weight and high water solubility), its water vapor pressure will be low and will increase slowly on dilution. This makes it an attractive candidate for favoring rapid transfer of water vapor through the membrane.



The basic transport process is illustrated schematically on a macro scale in Figure (1.3) OD is unique among membrane-separation processes in that it involves the transport of volatile components between two inherently miscible liquid streams, driven by differences in component activity between those streams. Its closest analogs are probably dialysis and membrane solvent extraction, although the former involves transport of solutes (whether volatile or nonvolatile) between two miscible liquid phases, and the latter transport of solutes between two immiscible liquids. In as much as the strip solution, following its dilution by water transferred from the feed stream, must be reconcentrated by evaporation so that it can be recycled and reused in the OD operation, it is important that the strip solute itself be thermally stable to quite high temperatures and also preferably nontoxic, noncorrosive, and of low cost. Water-soluble salts are the most attractive prospects for this purpose; those that have been most frequently employed are the alkali and alkaline earth metal halides (particularly sodium and calcium chloride). Sodium chloride, however, has relatively low water solubility and a rather low temperature coefficient of solubility, while calcium chloride is sensitive to precipitation in the presence of carbon dioxide; both are quite corrosive to ferrous alloys at elevated temperature. Salts that display large increases in solubility with temperature are desirable, because they can be evaporative concentrated to very high levels without danger of crystallization in the evaporator or during storage prior to recycle. It has found that, for osmotic concentration of foodstuffs and pharmaceutical products, the most attractive strip solutes are the potassium salts of ortho- and pyrophosphoric acid. These have quite low equivalent weights, very high water solubility, and very steep positive temperature coefficients of solubility. They also have the advantage of being normally present in biological fluids and, thus, safe for food when present in low concentrations. The or drug use vapor pressure/concentration relationships for a representative feed to be concentrated

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(for example, an aqueous sucrose solution) and several candidate brines as strip solutions are shown in Figure (1.4). The equivalent weights of the salts increase in the order NaCl > CaCl₂ > K₂HPO₄, as do their water solubilities. Because the "osmotic activity" of a salt is determined by the ratio of its water solubility to its equivalent weight, this in part accounts for the attractiveness of concentrated dipotassium orthophosphate brine for this application [4].

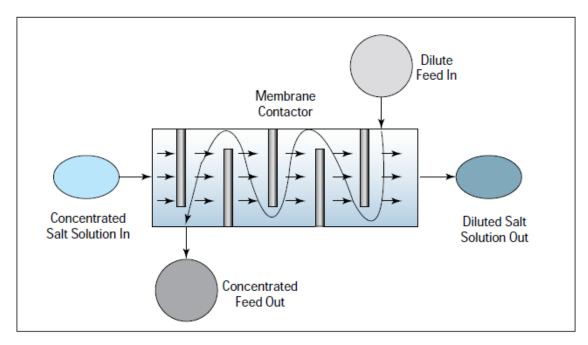


Figure (1.3) The basic transport process in osmotic distillation [4].



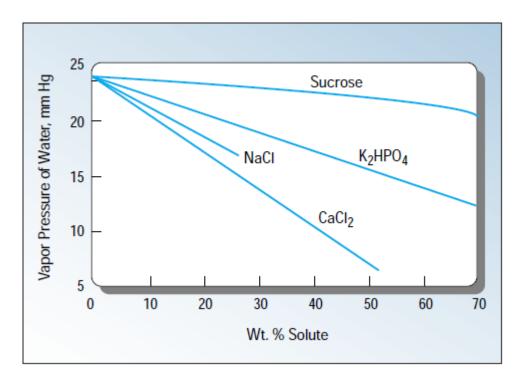


Figure (1.4) Generalized vapor-pressure relationships for sugar and salt solutions at $25^{\circ}C$ [4].

1.4 Process thermodynamics

The water transport process across the membrane takes place in three consecutive steps: (1) evaporation of water at the liquid meniscus at a pore entry; (2) diffusion or convective transport of water molecules as vapor through the membrane pore; and (3) condensation of water vapor on the brine-side liquid meniscus at the pore exit. The evaporative process requires the supply of the latent heat of vaporization at the upstream meniscus; this only can be provided as sensible heat via conduction or convection from the bulk upstream liquid, or via conduction across the solid phase comprising the membrane. Conversely, at the downstream face of the membrane, condensation of water vapor into the strip requires removal of the heat of condensation by the same mechanisms. Supplying or removing this energy by conduction/convection from the bulk liquid phases would, of course, cool the feed and heat the strip, thereby reducing the driving force for water transport. Fortunately, however, the thermal conductance of the membrane is

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sufficiently high that virtually all the energy of vaporization can be supplied by conduction across the membrane at a quite low temperature gradient [4].

As a consequence, under normal operating conditions, the temperature difference between the liquids on opposite sides of the membrane ("temperature polarization") is quite small seldom greater than 2°C. Hence, the process is essentially isothermal with respect to both liquid streams. For this reason, membranes prepared from solids of high thermal conductivity and of minimum practical thickness are desirable. It is interesting that the situation is exactly the opposite for the process of "membrane distillation". Many liquid feeds whose concentration is desired (such as fruit and vegetable juices, and vegetable extracts such as tea or coffee) also contain small concentrations of essential volatile, lipophilic organic solutes (flavor and fragrance components), the loss of which would make the product unpalatable and unmarketable. While such products can be concentrated by evaporation, losses of these essential volatiles with the water vapor are severe. Condensation of the vapor mixture, followed by rectification to recover these volatiles for reblending with the concentrate, can offset this somewhat but worsens thermal deterioration of these components and results in a significant incremental processing cost.

With OD, several factors make it possible to achieve concentration by selective removal of water without significant depletion of these important flavor/fragrance components. First, if the concentration is carried out at low temperature, the vapor pressure of these components (relative to that of water) is substantially depressed, reducing the driving force for transmembrane transport of these solutes. Second and perhaps more important the solubilities of these lipophilic solutes are substantially lower in concentrated saline solutions than in pure water; as a consequence, the vapor pressures of these solutes when present in any given concentration in such a solution are much higher than they are over water



at the same concentration. Thus, the vapor pressure driving force for vapor phase transfer of these solutes from the feed to the strip is far lower than that encountered in simple evaporation. Additionally, because the molecular weights of these solutes are far higher than those of water, their diffusive permeability through the membrane is much lower. The end result of these factors is that volatile flavor and fragrance losses from such feeds during OD often are too low to be significant. Indeed, concentrate, when rediluted with distilled water to its original volume, is organoleptically very similar to that of the original feed; this, of course, makes OD particularly attractive for food and beverage processing [4].

1.5 The Aim of the present work

The objective of the present work is to study effect of osmotic agent concentration and its type on osmotic distillation performance. Such performance could be achieved by combining the use of highly permeable membranes, calcium chloride solutions close to saturation with low water activity and a membrane module with favorable hydrodynamic conditions. Knudsen and molecular diffusion mechanisms are used for modeling the vapour transport through the porous asymmetric membrane. Classical correlation of dimensionless numbers is tested to predict the boundary layer mass transfer coefficient within the brine side of the membrane module. The fitting quality of these models is exposed and the limit of their application is discussed. The heat transfer is also investigated and the thermal effects associated with mass transfer in OD are estimated.

Chapter Two

Theoretical Concepts and Literature Survey

2.1 Introduction

Osmotic membrane distillation (OMD) is one of the membrane distillation (MD) variants, operated at low temperature. The MD comprises a relatively novel membrane process, which can be applied for the separation of various aqueous solutions. The hydrophobic membranes, with the pores filled by the gas phase, are used in this process [5, 6]. The hydrophobic nature of the membrane prevents penetration of an aqueous solution into the pores. Therefore, only volatile components of the feed may be transported through the membrane in the MD process. The different content of the particular components in the gas phase at both ends of the membrane pores (concentration gradient) causes their transport across the membrane. The composition of the gas phase above the liquid surface is often expressed by partial pressure, and the partial pressure difference was therefore accepted as a driving force of MD process. The value of this driving force depends on the solution temperature and composition in the layers adjacent to the membrane surface [5, 7, and 8].

The definitions of MD process do not consider the reasons for formation of driving force [5, 6]. These reasons may only affect the value of driving force and installation design, but they do not alter the MD process principles. The gradient of partial pressure across the membrane may be formed not only by temperature difference but also by the concentration difference and by the properties of solutions separated by the membrane [5, 7, 10 and 11]. The application of vacuum on the distillate side or the flow of dry gas also allows obtaining the desired effect [5, 12].

Various types of MD are known for several years (Figure (2.1)): directcontact MD (DCMD), air gap MD (AGMD), sweeping gas MD (SGMD) and vacuum MD (VMD). The OMD process, which has been developed dynamically in recent years also, can be included in this group. The addition of word



"osmotic" to "MD" is consistent with historical development of MD process nomenclature. This word indicates that OMD is a variant of the membrane distillation the course of which is significantly influenced by the solution concentration. Additionally, from the OMD term ensue (via analogy to the creation of terms VMD and SGMD) that a reason for the driving force formation is associated with the osmotic pressure (water activity).

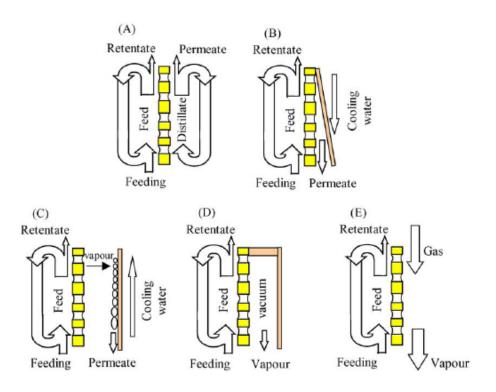


Figure (2.1) Types of MD process: (a) DCMD; (b) DCMD with liquid gap (Gore's design); (c) AGMD; (d) VMD; (e) SGMD.

2.2 Fundamentals of osmotic membrane distillation (OMD) process

Several authors consider the osmotic membrane distillation (OMD) process as distinctive from membrane distillation. Therefore, in the literature besides the term OMD [23, 25] and isothermal membrane distillation (IMD) [26, 27] the following terms are used: osmotic distillation (OD) [22, 28, 30], osmotic evaporation (OE) [13, 31, 35], gas membrane extraction [36] and membrane osmotic distillation (MOD) [29].



Osmotic evaporation is a concentration technique based on the use of mesoporous or macroporous and hydrophobic membranes [31]. This relatively new membrane process is performed at or below ambient temperature and under atmospheric pressure. The porous hydrophobic membrane separates two aqueous solutions (feed and osmotic solution) having different water activities. The membrane is not wet and the pores remain full of air. The driving force of the process is the water vapour pressure gradient between both sides of the membrane. The feed which is the solution to be concentrated and the stripping solution (usually concentrated brine) are generally isothermally pumped over opposite sides of the membrane. Water evaporates from the solution of higher vapour pressure (feed) then; the vapour diffuses through the pores and condenses into the solution of lower vapour pressure (osmotic solution).

Osmotic evaporation is interesting in the processing of heat-sensitive liquids because it takes place under mild operating conditions and then, thermal degradation of these solutions is avoided. It can be applied in pharmaceutical industry [38] or in food industry such as for concentration of fruit juices [39] which nutritional and organoleptic properties are thus preserved [40].

The second membrane contactor, membrane distillation, is also a concentration technique in which a porous hydrophobic membrane is used as barrier between the feed and the distillate [5]. A temperature difference between both sides of the porous hydrophobic membrane causes a vapour pressure difference which is the driving force of the process. This vapour pressure gradient causes a mass-transfer across the membrane. Thus, this process can be considered in the same category as distillation at temperature below the boiling point of the components to be separated. Indeed, the gaseous phase is only present within the membrane pores [41, 25].

The difference between OD and membrane distillation (MD) is that in MD the physical origin of the vapour pressure difference is temperature gradient, whereas in OD the physical origin is composition difference. OD is a



process in which two aqueous solutions (feed and brine) with different vapour pressures are separated by a microporous hydrophobic membrane; the pores of the membrane are not wetted and allow vapour transport. The process is depicted in Figure (2.2). Transport by OD involves three stages: [43]

- 1. Evaporation at the feed side of the membrane;
- 2. Transport of the vapour through the pores of the hydrophobic membrane;
- 3. Condensation of the vapour at the permeate side of the membrane.

Osmotic distillation (OD) is a relatively new process that is being investigated as an alternative to conventional separation processes for product concentration in the food industry. The conventional processes are thermal evaporation and reverse osmosis. The former results in heat degradation of the product while the latter is limited at high concentrations which are difficult to achieve because of the exponential increase in osmotic pressure with concentration. While these problems are largely avoided in membrane distillation, some loss of volatile components and heat degradation may still occur. OD, on the other hand, does not suffer from any of the problems mentioned above when operated at room temperature. It is therefore a convenient method of concentration for the food and pharmaceutical industries [31, 44].

The advantages of OD compared to other separation processes can be summarized as follows:

- 1. Ambient operating temperature and pressure;
- 2. Less demanding mechanical property requirements;
- 3. No or less degradation of heat-sensitive components;
- 4. Higher concentrated feed can be achieved.

Osmotic distillation (OD) has two limitations. Firstly, fluxes tend to be low due to the low driving force. This restricts OD to processing high value materials. The other limitation of the process is the possibility of wetting of the



hydrophobic microporous membrane and consequent loss of flux and separation performance [43].

The main advantage of osmotic evaporation and membrane distillation, compared to other membrane processes is the high selectivity for non-volatile compounds (100% retention of ions, macromolecule, colloids, and cells). In addition these processes are less energy consuming compared to traditional water distillation in a single step. The selectivity of both processes is controlled by the partial pressure differences of the feed components [42].

The osmotic membrane distillation (OMD) process was patented at the end of the last century [13, 14, 15 and 16], however, its beginning can be found in the earlier works [14, 18, 19 and 20]. In this variant, the feeding solution temperature is low and close to the temperature of the solution flowing on the other side of the membrane. The vapour pressure difference across the membrane was obtained by using a solution with a low water vapour pressure (extraction solution) on the distillate side. In order to increase the partial pressure difference, some authors proposed to maintain the feed temperature a few degrees higher than the temperature of extraction solution [24].

The extraction solutions comprise the concentrated solutions of salt (NaCl, CaCl₂, MgCl₂, MgSO₄) [32] and some organic liquids (glycerol, polyglycols) [33]. A choice of extraction solution plays a significant role, but information concerning this subject is limited. The different permeate fluxes were obtained for different extracting solutions under similar operational conditions (equal flow rates, temperature and concentration) [34]. However, if the dependence of flux on the Reynold number (Re) and on the difference of the water activity will be investigated, then it appeared that no difference between the used solutions was found.

The osmotic membrane distillation (OMD) process is most often used to remove water from liquid foods such as fruit and vegetable juices, milk, instant coffee and tea, and various nonfood aqueous solutions being thermally non-



resistant [37]. Low operation temperatures eliminate many problems associated with the conventional evaporation processes (reaction of non-enzymatic browning—Maillard's reaction), inhibit the degradation of flavour and colour, and a loss of volatile aromas (volatility of which rapidly increases with feed temperature).

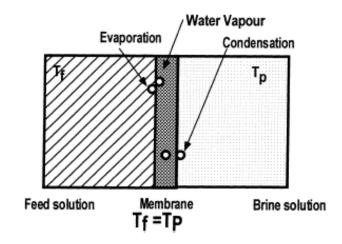


Figure (2.2) Schematic presentation of OD [43].

2.2.1 Main characteristics of osmotic evaporation

Osmotic evaporation (OE), also called osmotic distillation or direct osmotic distillation which is a competitor of reverse osmosis. In the OE technique a macroporous hydrophobic membrane separates two aqueous solutions having different osmotic pressure. Water evaporates in the solution of higher chemical potential and the vapour crosses the membrane before being condensed in the solution of the lower water potential. The driving force of OE is the difference in the vapour pressure of the solvent, usually water, in the solutions at both sides of the membrane. This difference is mainly due to two phenomena [31]:

(a) The difference in the chemical potential or of the osmotic pressure between the two liquids, due to the different nature and concentration of the solute components. As a rule of thumb, the osmotic pressure in an electrolyte solution is about ten times higher than in an equimolar solution of electrically uncharged



particles such as sugar molecules. Therefore salts solutions are efficient and relatively cheap systems to create high pressure differences. However, in the literature concentrated sugar solutions instead of salt solutions are also mentioned as "osmotic agent solutions" [79].

(b) The evaporation process cools down the solutions to be concentrated and the simultaneous condensation process at the other side of the membrane heats up the osmotic agent solution. This temperature difference causes a reduction of the vapour pressure difference and hence a decrease of the driving force. In practice, this undesirable effect can be controlled by a thermostat and intensive stirring of the two solutions. Furthermore, the membranes should be as thin and as heat conductive as possible in order to achieve rapid temperature equilibrium near the membrane. This is a clear difference with membrane distillation where a temperature gradient is the driving force and consequently, the membranes must have low thermal conductivity. Even small temperature differences can counterbalance considerable osmotic pressure differences and must hence be minimized in OE [31].

2.3 Theoretical Concepts

The process of osmotic distillation (OD) is a membrane contactor technique using a macroporous hydrophobic membrane. The faces of the porous element are put in contact with two circulating aqueous solutions of different solute concentration; a dilute solution on one side and a hypertonic salt solution on the other side (Figure (2.3)). Thanks to the hydrophobicity of the polymer, the membrane cannot be wetted by the liquids, thus creating vapour–liquid interfaces at each entrance of the pores. The water activity difference between the two liquid compartments translates into a vapour pressure difference at the vapour–liquid interfaces, which constitutes the driving force of the water transport. In principle, it is commonly admitted that the mass transport process can be split into three steps: the initial and final steps correspond to the water



transport in condensed phase, from the bulk of the dilute solution towards the evaporation interface and vice versa from the condensation surface to the bulk of the brine. The intermediate step corresponds to the water transfer in vapour phase across the porous material [22].

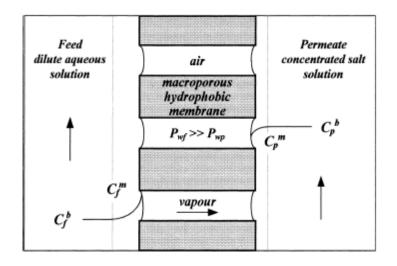


Figure (2.3) Principle of the osmotic distillation (OD) process [22]

Osmotic membrane distillation is a thermal membrane process, which employs hydrophobic membrane to separate two aqueous solutions having different osmotic pressures. The driving force for the mass (water) transfer is the difference in vapor pressure of the solvent (water) across the membrane. Water evaporates from the surface of the solution having higher vapor pressure, diffuses in the form of vapor through the membrane, and condenses on the surface of the solution [44]. The evaporation process requires the supply of the latent heat of vaporization at the upstream meniscus. This is provided as sensible heat via conduction or convection from the bulk upstream liquid, or via conduction across the solid phase comprising the membrane. Conversely, at the downstream face of the membrane, condensation of water vapor into the osmotic agent solution occur releasing heat of condensation. The thermal conductance of membrane should be sufficiently high, so that all the energy of vaporization can be supplied by conduction across the membrane at a low temperature gradient.



As a consequence, under normal operating conditions, the temperature difference between the liquids on either sides of the membrane is quite small (≈ 2 °C) [22], and the process could be considered as isothermal in certain cases.

2.3.1 Mass Transfer

The mass transfer mechanism in OD as well as in MD has been widely discussed: at both pore entrances vapour–liquid equilibrium are established giving rise to a partial pressure difference of water vapour which diffuses through the gas immobilised within the pores. The water vapour pressures at the pore mouths are related to the temperature and activities prevailing in the liquids facing the membrane by:

$$P_{W_1} = P_{W_1}^* a_{W_1} \tag{2.1}$$

$$P_{W_2} = P_{W_2}^* a_{W_2} \tag{2.2}$$

In which P_W^* represents the vapour pressure of pure water and a_W the water activity in the solutions. In osmotic distillation (OD) the driving force $(P_{W_1} - P_{W_2})$ for water transport is sustained by an activity difference i.e. $a_{W_2} < a_{W_1}$. In membrane distillation, on the contrary, the activity difference represents a counter driving force to be overcome by a temperature difference across the membrane, i.e. in that case $a_{W_1} < a_{W_2}$, $P_{W_1}^* > P_{W_2}^*$. Of course situations are possible in which both temperature and activity differences act synergistically [45].

The basic equation, which relates the transmembrane flux (J) to the driving force represented by the difference in vapor pressures of the bulk liquids (feed and OA) and is given by

$$J = K\Delta P \tag{2.3}$$

Where 'K' is the overall mass transfer coefficient which accounts for all the three resistances for water transport and is given by



$$K = \left(\frac{1}{K_f} + \frac{1}{K_m} + \frac{1}{K_{OA}}\right)^{-1}$$
(2.4)

Where K_f , K_m and K_{OA} are the mass transfer resistances in feed layer, membrane and osmotic agent layer, respectively [48].

2.3.1. A. Mass transfer through the membrane

The resistance for the diffusive transport of water vapor across the microporous hydrophobic membrane is offered by the membrane pore structure as well as air present in the pores. The diffusion of water vapor through this stagnant gas phase (air) of the membrane pore can be described either by Knudsen diffusion or molecular diffusion depending on the pore size [47].

When the mean free path is significant relative to the pore size, the diffusing molecules collide more frequently with the pore wall and the diameter of the pore is important. Such mass transfer is termed as Knudsen diffusion and the membrane diffusion coefficient (K_{mK}) is given by [48].

$$K_{mK} = 1.064 \frac{r\varepsilon}{X^{\delta}} \left(\frac{M}{RT}\right)^{0.5} \tag{2.5}$$

When the membrane pore size is relatively large, the collisions between the diffusing molecules themselves are more frequent. Such mass transfer is termed as molecular diffusion and the membrane diffusion coefficient is expressed as [49].

$$K_{mm} = \frac{1}{\gamma_{ln}} \frac{D\varepsilon}{X^{\delta}} \frac{M}{RT}$$
(2.6)

Where 'D' is the Fick's diffusion coefficient and can be predicted by

$$D = \frac{0.001858T^{3/2} (1/M_A + 1/M_B)^{1/2}}{P\sigma_{AB}^2 \Omega_D}$$
(2.7)

Both these approaches are useful for predicting the mass transfer through the membrane mainly depending on the membrane pore size. However, each of them has its own limitations. The Knudsen equation requires details of membrane pore geometry (such as pore radius, membrane thickness, tortuosity). Molecular diffusion does not hold at low partial pressure of the air, as ' Y_{ln} ' tends to 0 and hence, molecular diffusion is clearly undefined; thereby diffusion mechanism approaches Knudsen diffusion [48].

2.3.1. B. Mass transfer across the boundary layers

The boundary layers are present in the feed and the OA on either side of the membrane. These layers may offer significant resistance to mass transfer, depending on the physical properties of the solution (feed and OA) as well as the hydrodynamic conditions of the systems. The liquid mass transfer coefficients in the boundary layers of feed and OA (K_f and K_{OA}) can be estimated by using empirical equations given below, involving only physical properties and hydrodynamic conditions of the solutions.

$$Sh = b_1 R e^{b_2} S c^{b_3}$$
 (2.8)

Where b_1 , b_2 and b_3 are the constants and are to be selected appropriately for the given hydrodynamic conditions, and

$$Sh = \frac{L k_i}{D_W}$$
, $Re = \frac{\rho u L}{\mu}$ and $Sc = \frac{\mu}{\rho D_W}$ (2.9)

Where D_W is the water diffusion coefficient and can be estimated by using following empirical equation [50, 51].

$$D_W = \frac{(117.3 \times 10^{-18})(\varphi M_W)^{0.5}T}{\mu \nu_A^{0.6}}$$
(2.10)



In order to obtain K_f and K_{OA} in the same units of K_m the following equation was used [22].

$$K_i = \frac{k_i C^t M_W}{(x_s)_{lm} \gamma P^*} \tag{2.11}$$

Where C^t is the molar concentration of the solution, γ activity coefficient and P^* the saturation vapor pressure.

2.3.2 Heat transfer

The water transport in osmotic membrane distillation is a simultaneous heat and mass transfer process. Even though osmotic membrane distillation is carried out with the same bulk solution temperature, a temperature difference is created at membrane interfaces due to the water evaporation in one side and further condensation in the other side. Evaporation cools the feed side and condensation warms up the osmotic agent side. The resultant temperature gradient across the membrane, translates into a lower vapor pressure gradient, which in turn, decreases water transport across the membrane. Heat transfer also occurs between bulk solutions and membrane surface. At steady state conditions, the heat transfer equations are as follows [45, 22].

$$Q = h_1(T_1 - T_{m1}) = h_2(T_{m2} - T_2)$$

$$Q = N_W \Delta H_v - h_m(T_{m2} - T_{m1})$$
(2.12)
(2.13)

Where Q is the heat flux, h_1 , h_2 and h_m the heat transfer coefficients of the feed side, osmotic agent side and membrane heat transfer coefficients, respectively, T_1 and T_2 the bulk temperatures of feed and osmotic agent, respectively, T_{m1} and T_{m2} the temperatures at the membrane interfaces of feed and osmotic agent sides, respectively, ΔH_v the water latent heat of vaporization



and N_W is the mass flux. From above equations, and considering that the bulk temperatures are equal and maintained constant ($T_1 = T_2$, near-isothermal condition for all practical purposes), the transmembrane temperature difference ΔT_m is given by [22].

From Equations (2.13) and (2.14), an equation that relates the heat flux to the water flux and to individual heat transfer coefficients is given by

$$Q = \frac{N_w \Delta H_v}{1 + (\frac{h_m}{h_1} + \frac{h_m}{h_2})}$$
(2.15)

The membrane heat transfer coefficient is given by the ratio of conductivity of the solid barrier to its thickness as given by the following equation [52].

Where k_{gas}^{T} and $k_{polymer}^{T}$ are the thermal conductivities of the air and of the membrane material, respectively.

Simulations of this thermal effect for the OD of fruit juices were carried out by Sheng et al. who showed that the vapour flux could be significantly improved by reducing the transmembrane temperature gradient [53]. Several theoretical approaches have emerged in the literature to try and couple mass and heat transfer in the OD system [25, 45]. Basically, it consists of using Clapeyron's law (2.17) to express the vapour pressure of a non-ideal aqueous



solution as a combination of an activity variable and a temperature variable (2.18), P^* being the saturation vapour pressure:

$$\frac{dP^*}{dT} = \frac{P^*(T)M_W\Delta H_v}{RT^2} \qquad (2.17)$$

$$dP_{w} = P^{*}(T)da_{w} + a_{w}\frac{P^{*}(T)M_{w}\Delta H_{v}}{RT^{2}}dT \qquad (2.18)$$

With
$$P_W = a_W P^*(T)$$
 (2.19)

In that way, the real driving force ΔP_w^m corresponding to activity and temperature at membrane conditions is expressed as a function of ΔP_w^b corresponding to the same variables as those at bulk conditions (2.20). The vapour flux can then be related to bulk concentration and temperature conditions that are experimentally accessible:

$$\Delta P_w^m = \Delta P_w^b + a_w \frac{P^*(T)M_w\Delta H_v}{RT^2} \Delta T^m \qquad (2.20)$$

Where a_w is the mean water activity of the liquid compartments and *T* the mean temperature. Gostoli proposed a synthetic way of integrating the thermal effect in the basic OD mass transfer equation, by introducing a sort of efficiency coefficient Θ (2.21) [45]. Such a coefficient is an analogue to an MD temperature polarization coefficient and represents the fraction of the driving force really effective for mass transfer through the membrane:

2.3.2. A. Heat transfer in the liquids

The liquid heat transfer coefficients depend on the physical properties of the solutions as well as on the hydrodynamic conditions prevailing in the module. By analogy with what was presented for mass transfer in the liquid compartments, h_f and h_p can be estimated either from experiments or with the help of empirical correlations of dimensionless numbers, like Nusselt (*Nu*), Reynolds (*Re*) and Prandtl (*Pr*) numbers. The example given by correlation (2.22) corresponds to the heat transfer coefficient h_L for a liquid flow in a tube, with a correction factor μ^b/μ^m used to express the thermal dependence of the fluid viscosity [54]:

$$Nu = b_1 Re^{b_2} Pr^{b_3} (\frac{\mu^b}{\mu^m})^{b_4}$$
 With $Nu = \frac{h_L d_h}{k^T}$ and $Pr = \frac{\mu c_p}{k^T}$ (2.22)

Where b_i is the correlation coefficient, k^T is the thermal conductivity, c_p the heat capacity and μ the viscosity of the fluid. Temperature polarization has been of major interest in the field of MD studies, since heat transfer across the liquid boundary layers is often the rate limiting step of the MD process. For OD, no study has been published until now about the magnitude of the heat boundary layer resistance. However, the global temperature polarization phenomenon was estimated to be responsible for 20% flux decay in the OD of orange juice with glycerol [56].

2.3.3 Polarization phenomena

2.3.3. A. Temperature polarization

The temperature polarization causes that the temperatures in the layers adjacent to the membrane (T_1 and T_2) differ from those measured in the bulk of the feed (T_F) and distillate/brine (T_D) [55]. However, the temperature profiles formed in the DCMD and OMD are different Figure (2.4). In DCMD, we have



 $T_1 > T_2$, whereas in OMD is inversely, $T_1 < T_2$. The temperature gradient is obtained due to the evaporation at the feed side and the condensation at the distillate side, even if the bulk temperatures of the two liquids are equal, as in OMD [59, 45]. Conduction of heat from the brine to feed induces a decrease of the polarization effect in OMD. In case of DCMD, the situation is reversed, heat is associated with the mass transfer as well as conducted through the membrane flows in the same direction, i.e. from the feed to distillate side, therefore, the temperature polarization effect increases.

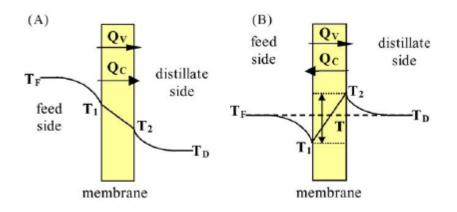


Figure (2.4) Temperature profiles during DCMD (A) and OMD (B) processes

In the case where the bulk temperature is controlled and maintained at the same level in both liquids ("well stirred" cell), the transmembrane temperature difference is given by [59, 45]:

$$\Delta T = \frac{N \,\Delta H}{h_m + (\frac{1}{h_f} + \frac{1}{h_p})^{-1}} \tag{2.23}$$

The temperature difference $(\Delta T = T_2 - T_1)$ in OMD process should be minimized because the vapour pressure gradient induced by this temperature difference is opposite to the pressure gradient created by the concentration gradient [57, 58, 59, 60]. Therefore, the occurring ΔT causes that the driving force for vapour transport decreases. This dependence in DCMD is reversing,



because the permeate flux increases for higher ΔT . Mengual et al. [58] estimated the temperature difference in OMD system on the basis of the equation:

$$N\Delta H = h A \Delta T \tag{2.24}$$

Where A is membrane area. They obtained the value of ΔT within the range from 0.5 to 0.8K (on each membrane side) for $h = 1900 \text{ W/m}^2 \text{ K} (h_D = h_F)$ and $N = 5-8 \times 10^{-8} \text{ kg/m}^2$ s. Therefore, they concluded that such values would lead to negligible decrease of the vapour flux. On the contrary, Vahdati and Priestman [61] have found that a small temperature difference can significantly affects the driving force induced by even very high concentration gradients. The thermal effect due to evaporation and condensation at both liquid-membrane interface increases with increasing permeate flux.

Gostoli proposed to use the coefficient Θ which represents the fraction of the driving force which is effective for the mass transport through the membrane [45]:

$$N = L_m \Delta P = \Theta L_m \Delta P^b \tag{2.25}$$

The value $\Theta = 0.85$ was obtained for TF200 membrane used to evaporate pure water at 25°C with NaCl solution in a stirred cell module. In the case of co-current flow system, with the same liquids, a 31% decrease of flux was obtained as a result of the temperature difference formed across the membrane [59].

The temperature profiles presented in Figure (2.4B) may be obtained only in a stirred cell of small laboratory apparatus. However, in the OMD pilot plant modules the existing temperature gradients causes the heat transfer between the



membrane surfaces and the bulk of solutions. As a consequence, the feed temperature decreases and the brine temperature increases, what is presented in Figure (2.5A). The formation of the temperature difference ($\Delta T_b = T_D - T_F$) causes a further decrease of the driving force formed by the concentration difference. In the extreme case, the mass transfer can be even stopped. For this case, considering that ΔT is proportional to ΔT_b , it is possible to find the relationship between the activity difference corresponding to an arbitrarily chosen concentration difference and its "equivalent" bulk temperature difference, given by the following expression [62]:

$$\Delta T_b \propto \frac{RT_m}{L} \frac{\Delta a}{a_m}$$
 (2.27)

The temperature profile also changes along the OMD module [60, 45]. Near the entry section, the profile is similar to that represented in Figure (2.4B), obtained for symmetric case (equal inlet temperatures and heat capacities). As a result, the heat transfer between the membrane surfaces and the bulk of solutions the temperature gradient presented in Figure (2.5A) is created. Moreover, in this system the temperature difference ΔT across the membrane increases. An asymptotic value ΔT_{∞} is finally reached for which the convective heat flux (Q_V) through the membrane (mass transfer) is exactly balanced by the conductive heat back-flux (Q_C) [59, 60, and 45]:

$$\Delta T_{\infty} = \frac{N \,\Delta H}{h_m} \tag{2.28}$$

The temperature profile presented in Figure (2.5B) will be obtained if the OMD system works without the heat losses.

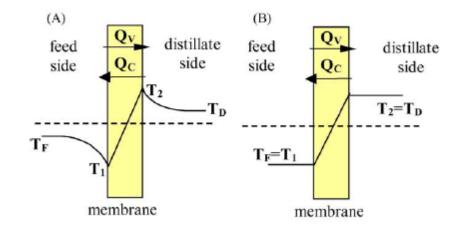


Figure (2.5) Temperature profiles during OMD process carried out in a largearea module: (A) initial temperature profile; (B) final temperature profile.

2.3.3. B. Concentration polarization

In OMD process (similar as in other MD variants), only water passes through the membrane, therefore the solute concentration near the membrane on the feed side is larger than that in the bulk, whereas the solute concentration on the extract side is lower than that in the bulk. This phenomenon is termed as the concentration polarization and is resulting in a driving force reduction across the membrane [5, 22, and 61].

The concentration polarization in DCMD process is usually considered in the context of a solute build-up on the feed side, with the distillate being pure liquid, as in typical desalination applications [18]. This effect has a smaller influence on the performance of DCMD. Therefore, several authors assume that the temperature polarization is essential in the case of DCMD, whereas the concentration polarization is essential in the case of OMD [22]. However, the polarization effect in DCMD variant becomes important in the case of concentration of solutions with solute content close to the saturated state [63].

The concentration polarization in the OMD process is important, particularly on the side of extracting solution. The presence of the layers of concentrated feed and diluted brine along the surface of the membrane can significantly decrease the difference of water activity (Figure (2.6)).

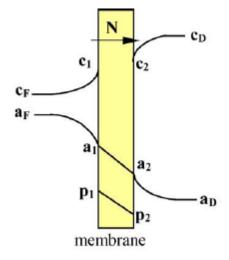


Figure (2.6) Concentration polarization profile in OMD process

A significant concentration polarization effect was observed during OMD with glycol [60]. As a result, the permeate flux varies in the non-linear way with the driving force evaluated under the conditions prevailing in the bulk phase. The influence of concentration polarization increases with the increase of the permeate flux.

Some authors [61] have assumed that the brine concentration in the layer adjacent to the membrane (C_2) can be reduced to zero when temperature differences are applied across the membrane ($T_F > T_D$ such as in DCMD) in order to increase the OMD flux [29]. Along with the increase of permeate flux, the diffusion resistance in the salt solution was much higher than the heat resistance. Therefore, a high flux established due to the temperature difference results in the complete concentration polarization, i.e. the vapour pressure difference due to the concentration gradient was equal to zero, with almost pure water on both membrane surfaces. This result was confirmed by repeating the experiment with pure water on both sides, i.e. the same flux was achieved as with the salt system [61].

In accordance with the film theory, the salt concentrations in the bulk phase (C_D) , and at the solution–membrane interface (C_2) are related to the volume flux by equation [22, 58]:

$$C_D = C_2 \exp(\frac{J_V}{k_M}) \tag{2.29}$$

However, when the flux in OMD process is much lower than the mass transfer coefficient (k_M) we can assume that C_D and C_2 have similar values. In this case, the flux is given by [58, 31]:

$$N = \frac{L_m k_M}{k_M + \left[\frac{d\Delta P}{dc}\right]_b L_m c_D} \Delta P^b \tag{2.30}$$

Where k_M is the mass transfer coefficient for solute in the layer adjacent to the membrane.

2.4 OMD modules and process applications

The major application of OMD process includes the concentration of fruit juices and other temperature nonresistance liquids. Process allows the production of concentrated fruit juices with quality and composition very close to fresh one, without added flavours and with high vitamin content at a cost comparable with the conventional product obtained by evaporation [59]. The limiting concentration of juices in OMD process is associated with very high viscosity of juices (>0.2 N/m) at concentrations of sucrose exceeding 68°Brix [26, 27, 13, 34]. For the feed concentration below 40 wt% of sugar, the permeate flux varies in accordance with the membrane permeability, which indicates that the process is controlled by the membrane resistance. At higher concentration



(50–68 wt %), the mass transfer resistance on the feed side is dominant, and there is no advantage in using a more permeable membrane [27].

Different configurations of modules were tested for OMD applications. The membranes may be flat [22, 27, 14] or in a form of hollow fibers [18, 37]. Considering a high viscosity of the liquids flowing through a OMD module the application of plate-and-frame [14] or spiral-wound [18, 31] configurations is recommended. Moreover, the plate and frame module (also spiral-wound) have membrane with lower thickness, thus the obtained flux is larger in comparison to capillary modules [36].

The performance of OMD process was also evaluated on a commercial scale with modules having the area above $1m^2$ [64, 31, 36, and 65]. In the pilot plant or industrial installation, the two solutions are pumped along the opposite sides of the membrane, e.g. in a counter-current flow. However, the hydrodynamic conditions are slightly different from those created by simple agitation often used at laboratory scale [31, 66]. Under unfavourably conditions of fluid mixing, the OMD process can be strongly limited by the concentration polarization [36, 22]. A relatively high flux (8–12 dm³/m² h) was obtained with the new plate-and frame lab-scale OMD module [22]. In this design, the fluids were circulated into square channels evenly distributed so that the velocity of the fluids is identical throughout entire membrane surface. The most advantageous hydrodynamic conditions were obtained for *Re* > 1000.

Helically-wound hollow fibre modules offer a significant improvement in the hydrodynamic conditions on the shell side in comparison with the axial flow modules. In the case of processing of viscous feeds in OMD process with the use of helical module, the higher concentrations of solutes and two times larger flux were obtained than those obtained for module design with axial flow [67].

2.5 Previous Studies for OMD

OMD is a membrane separation process in which water evaporates from the feed stream and is then transferred in the vapor phase through the pores of a hydrophobic membrane to the strip stream where it re-condenses. The only driving force for the water vapor mass-transfer in the pores of hydrophobic membranes is the water vapor pressure gradient. To date, OMD has been hardly used in industry, and almost exclusively for the concentration of aqueous, food process streams, such as juices, fermentation broths or pharmaceutical intermediates. In general, it is not possible to concentrate these products by conventional thermal evaporation without negatively affecting their organoleptic or therapeutic properties. In such applications users want to avoid thermal degradation of the feed products, such as flavour compounds. This requirement limits the OMD process operating temperature to an ambient level [77].

Carlo, 1999, investigated both theoretically and experimentally the thermal effects associated to the mass transfer in OD. Due to the evaporation at the feed side and condensation at the brine side, a temperature difference is created through the membrane even if the bulk temperatures of the two liquids are equal. Of course this thermal effect reduces the driving force for water transport. It is expected that the temperature difference will be quite small, owing to the low membrane thickness. However, it is known from membrane distillation studies that a small temperature difference cancels out a large concentration difference [45].

Mansouri and Fane, 1999, described the development of modified hydrophobic membranes for osmotic distillation (OD) which are tolerant to oily feeds. Three commercial membranes were chosen as substrates including the Celgard 2500, Millipore GVSP and the UPVP. The focus has been on using PVA coatings which were found to have an insignificant effect on flux. For concentrated sucrose solutions, flux was reduced significantly by viscosityrelated concentration polarization effects. The presence of the coating did not



increase this effect, suggesting that the flux limitation mostly occurred adjacent to the feed-side of the laminate and not within the top-layer or inside the pores. The laminate membrane is a necessity for oily feeds since the uncoated membranes were promptly wetted out even for low concentration of oil (limonene) dispersion in water. The laminate membranes were all stable in oil emulsions when tested for periods up to 24 h; i.e. the membranes did not wet out during flux measurement and no visual damage nor was coating detachment observed for laminates. These observations confirm the efficacy of having coated membranes for OD of oily feeds [43].

Mathilde et al ,2000, applied several models to represent the mass transfer in osmotic distillation (OD) systems are applied to the results of pure water OD experiments carried out with two commercial asymmetric porous membranes. Molecular and Knudsen diffusion mechanisms were tested to model the vapour transport across the membrane. When using the global structural characteristics specified by the membrane manufacturer, both diffusion models underestimate the membrane permeability to water vapour. The exceptionally high experimental permeability can be predicted by a Knudsen mechanism when considering the Teflon top layer alone. The membrane support was envisaged as an additional resistance to water transfer in the liquid form, leading to splitting of the asymmetric membrane into a series of two resistances: one resistance to gas transfer in the top layer and another to liquid transfer in the support layer. In this model, the gas membrane contribution was estimated to cover 40-70% of the total mass transfer resistance; the film of diluted brine entrapped in the membrane support can cover up to 30% of the total mass transfer resistance and the diluted brine boundary layer up to 60%, indicating the sensitivity of the OD system to concentration polarization. Classical empirical correlations of dimensionless numbers were fitted to the experimental flux results to try and predict the mass transfer coefficients of the brine boundary layer in the OD system. The poor quality of the model was attributed to the special



hydrodynamics of the membrane module whose geometry did not fit in the reference of the correlations, i.e. straight circular ducts. The heat transfer associated with water transport is integrated into the mass transfer equations. The thermal effect due to evaporation and condensation at both liquid–membrane interfaces appears to be significant: a high vapour flux of 12 kgm⁻² h^{-1} generates a transmembrane temperature difference of approximately 2°C inducing a 30% driving force reduction. Finally, the temperature polarization in the liquid compartments was shown to be of minor importance in comparison with the heat transfer resistance constituted by the membrane itself [22].

Mathild et al, 2000, studied a recent membrane technique osmotic distillation (OD), which was used to concentrate binary water-sucrose solutions at ambient temperature under atmospheric pressure. The principle was based on the extraction of water vapour from a dilute aqueous solution, which was put in contact with a hypertonic salt solution by means of a macroporous hydrophobic membrane. The concentration difference between both solutions translated into a transmembrane vapour pressure drop that constituted the driving force for mass transfer. An experimental device was designed at laboratory scale for this study, allowing achievement of vapour fluxes of 10 kgm⁻² h⁻¹ under standard conditions. The effect of various operating parameters on vapour flux was studied. The solute content resulted in being the most influencing variable via water activity in brine and via viscosity in sugar solutions. The effect of concentration polarization on the brine side was not negligible and had to be taken into account for process optimization. This phenomenon could not be quantified on the sugar solution side due to pressure drop limits of the pilot rig. Eventually, the vapour flux can be significantly increased by adding a temperature difference to the transmembrane concentration difference, when pure water was evaporated [38].

Bailey et al, 2000, investigated the potential for osmotic distillation flux enhancement in grape juice concentration by ultrafiltration pretreatment using



ultrafiltration membranes with a range of pore diameter/MWCO values. Juice viscosities were determined over the concentration range to assist with interpretation of the flux data. Ultrafiltration using membranes with pore diameters of 0.1 mm or less resulted in appreciable osmotic distillation flux increases over that observed for juice not subjected to ultrafiltration. HPLC measurements showed that the normal concentration of fermentable sugars in standard 68°Brix concentrate can be achieved at a lower Brix values with UF permeate, thereby providing a possible means of reducing the handling of highly viscous streams. Ultrafiltration also resulted in an increase in juice surface tension with a consequent reduction in the tendency for membrane wet-out to occur [37].

Celere and Gostoli, 2005, studied OD experiments with various extractants in a plate and frame module expressly designed to investigate separately the simultaneous effects of the heat and mass transfer. In co-current operation, the temperature difference between the two streams approaches an asymptotic value for large residence time, i.e., for long modules or low flow rates. In asymptotic conditions, which correspond to no heat flux through the system, the problem looks like a pseudo isothermal case; this allows to obtain the relevant membrane transport properties and to evaluate the role played by the concentration polarization. The temperature difference created by the thermal effect, even of few degrees, greatly reduces the driving force for mass transfer through the membrane; the resulting loss of flux was relevant and increases with the temperature. The concentration polarization plays a comparable role in the OD with calcium chloride or glycerol at room temperature, while was less important in OD with glycerol-NaCl mixtures, and negligible in the case of sodium chloride. Among the extractant used, CaCl₂ was the most effective; however, the advantage over the Glycerol-NaCl mixture was not so high as would appear comparing the respective theoretical driving forces for mass transfer. The length of module needed to approach asymptotic

conditions (thermal entry length) can be estimated to be lower than the typical length of industrial apparatuses. The flux achievable in OD applications was, thus, close to the asymptotic value, larger fluxes can only be obtained in lab devices [69].

Tzahi et al, 2005, evaluated an innovative system that combines three different membrane processes for reclamation and reuse of waste water in future space missions. The direct osmotic concentration (DOC) system consists of an array of five reverse osmosis (RO) elements, a direct osmosis (DO) pretreatment process, and a combined direct osmosis/osmotic distillation (DO/OD) pretreatment process. Optimized operating conditions, including RO pressures, salt load in the brine loop, and flow velocities were determined for the three subsystems. Mass and heat transfer in the pretreatment processes were measured. Water flux in the DO process was found to be strongly dependent on the type of membrane used; it ranged from 10 to 25 $l/(m^2 h)$ for a cellulose triacetate membrane specifically designed for this application and from 0.5 to 2 $1/(m^2 h)$ for commercially available RO membrane. Water flux through the dual DO/OD process was also found to be highly dependent on temperature gradient across the membranes-increasing with increasing temperature gradient. The conditions for minimum energy consumption of the system were determined and used in estimating the specific energy cost of treating the wastewater generated in space. The weight of salt resupply for continuous operation was also estimated. When compared to alternative technologies, the DOC system provides high wastewater recovery (>95%), at low energy cost ($<90\times103$ J/l (25 Wh/l)), with minimal resupply (<20 kg/year) [70].

Tzahi et al, 2005, investigated the improvement of an innovative dual membrane contactor process for treatment of combined hygiene and metabolic waste water. Flux and solute rejection in the combined direct osmosis/osmotic distillation (DO/OD) process were enhanced by incorporating membrane distillation (MD) concepts into the process. Two new configurations were



investigated: DO/MD, in which the driving force was temperature gradient only, and DO/membrane osmotic distillation (DO/MOD) in which the driving forces were temperature gradient and concentration gradient. Development of a temperature gradient across the membranes substantially enhances the flux of the dual membrane process. It was demonstrated that water flux could be increased by up to 25 times with only a 3-5 °C temperature difference across the membranes. Solutes in the feed wastewater, including urea, were completely rejected. It was demonstrated that complex wastewaters that cannot be treated by one process only could be well treated using a dual membrane process [71].

Naveen et al,2006, studied the effect of various process parameters such as type, concentration and flow rate of the osmotic agent; type (polypropylene membranes) and pore size (0.05 and 0.2 μ m) of the membrane; temperature with respect to trans membrane flux. Experiments were performed with real systems (pineapple/sweet lime juice) in a flat membrane module. Osmotic agents namely sodium chloride and calcium chloride at varying concentrations are employed. For both the osmotic agents, higher transmembrane flux was observed at maximum osmotic agent concentration. In comparison with sodium chloride, higher transmembrane flux was observed in case of calcium chloride. A mass transfer-in-series resistance model has been employed, considering the resistance offered by the membrane as well the boundary layers (feed and brine sides) in case of real systems for the first time. The model could predict the variation of transmembrane flux with respect to different process parameters [45].

Ravindra et al, 2006, evaluated the effect of various process parameters, such as concentration (2-10 M) and flow rate $(25-100 \text{ ml min}^{-1})$ of feed and osmotic agent on the transmembrane flux in case of phycocyanin and sweet-lime juice. The increase in the osmotic agent concentration and flow rate resulted in an increase in transmembrane flux. A three-fold concentration in case of phycocyanin and 10-fold in case of sweet-lime juice could be achieved. The

magnitude of temperature polarization is small and the maximum temperature difference across the membrane was only 0.8K that too for 10M calcium chloride solution. The feed and osmotic agent side mass transfer resistances were estimated based on classical empirical correlation of dimensionless numbers, whereas membrane resistance was estimated using Dusty-gas model. The mass transfer mechanism was found to be in the transition region that is between Knudsen and molecular diffusion. The type of mass transfer mechanism was dominating depending upon the pore size of the membrane. In case of membrane with pore size of 0.05 μ m, the contribution of Knudsen diffusion is higher (74% of the membrane resistance), whereas for 0.20 μ m the contribution of molecular diffusion is higher (59% of the membrane resistance). The flux across the membrane during the process was predicted using resistances-in-series model. The experimental values were found to correlate well with the predicted values [72].

Alves and Coelhoso, 2006, compared between the osmotic evaporation (OE) and membrane distillation (MD) processes in terms of water flux and aroma retention. The concentration of a sucrose solution, used as a model fruit juice, was carried out in a hollow fibre membrane contactor. Although a similar overall driving force was used, the flux obtained using the MD process was less than half of that observed using the OE process, due to temperature polarization effects. The transport of citral and ethyl butyrate, two aroma compounds relevant in the orange juice aroma, was also compared for both processes. A higher retention of these compounds was observed with the OE process. From these studies, it could be concluded that, the OE process has advantages over the MD process, not only in terms of water flux, but also regarding the retention of aroma compounds. Finally, the concentration of a commercial orange juice by osmotic evaporation was carried out. Due to the presence of suspended solids and macromolecules, a higher mass transfer resistance was observed [73].

Thanedgunbaworn et al, 2007, studied the effect of packing density of hollow fibre modules on mass transfer in the shell side of osmotic distillation process. The osmotic distillation experiments were carried out with several modules of the packing densities ranging from 30.6 to 61.2%. It was found that the Reynolds number was a function of packing density and packing density affected mass transfer performance. The shell-side mass transfer coefficient increased with the brine velocity. The membrane permeability can be predicted from the experimental flux at the maximum brine velocity. The mass transfer coefficient in the randomly packed modules for osmotic distillation process. The empirical correlation proposed was fitted to the experimental results and it was found that the mass transfer coefficients calculated from the proposed correlation were in good agreement with those from the experimental data [74].

Hengl et al, 2007, developed a new technique of membrane evaporation (ME) which is a new membrane contactor process combining the advantages of osmotic evaporation and membrane distillation. It is based on the concentration of an aqueous solution by continuous evaporation through a metallic hydrophobic membrane able to be heated. The driving force of the process is the difference of water vapour pressure between a feed of an aqueous solution and a downstream flow of dry gas at low pressure. The objective of these introductory attempts is to validate this new concept; hence preliminary experiments have been realized with pure water. Then, a first approach to model this system and a comparison with experimental results has been carried out [75].

Thanedgunbaworn et al, 2007, carried out the Osmotic distillation process on a hollow fibre membrane module using fructose solutions and clarified grape juice as feeds. The influence of operating parameters such as feed and brine flow velocities, feed concentration, and temperature, on the osmotic distillation flux was studied. For the experimental conditions employed, the water flux varied from 0.58 kg m⁻² h⁻¹ to 2.02 kg m⁻² h⁻¹. Temperature and feed concentration had

significant effect on flux. On the contrary, feed and brine velocities had little effect on flux. The increase of feed and brine velocities or change of the hydrodynamic conditions also enhanced flux. The concentration polarization at high flow velocities can be neglected. In this work, the value of temperature polarization was small and the polarization coefficient H was estimated. It was found that temperature polarization reduced the driving force for mass transfer approximately 5–6%. However, this phenomenon affected the transport resistance of the system. The concentration and temperature polarization can be reduced by operating the process at high Reynolds number and low temperature. The transport resistance was affected by changing of operating condition such as feed velocity and temperature. The major transport resistance of the process was in the membrane [76].

Chapter Three Experimental Work

This chapter explains and views in details the experimental part of this work. It includes the description of experimental rig to study the osmotic membrane distillation process and measuring the experimental data.

This work was carried out in Spain – Madrid – University of Complutense faculty of physics – Department of applied physics.

3.1 The Experimental Rig

The layout of experimental rig used in the present work is shown schematically in Figure (3.1). The general view of the rig is shown in Figure (3.2). The experimental rig consists of the following items:-

1. Feed Tank

The feed tank is a cylindrical vessel of stainless steel it is filled with distilled water and surrounded by jacket to control the feed tank temperature at 30 °C.

2. Permeate Tank

It is a cylindrical vessel of stainless steel, filled with salt solution which consists of NaCl solution and $CaCl_2$ solution which is used as osmotic solution, also it is surrounded by Jacket to maintain the temperature of osmotic solution as same as feed temperature. The salt solution was prepared by different concentrations from (1M to 5 M).



3. Membrane module

The flat sheet commercial membrane, TF200 From Pall – Gelman It is made of a thin polytetrafluoro ethylene (PTFE) macroporous layer supported by a polypropylene (pp) net and it is selected because of high vapour permeability in the process. A flat membrane module having a membrane area of 2.75×10^{-3} m² was used, its characteristics are shown in table (3.1).

membrane area	2.75 *10 ⁻³ m ²
membrane thickness δ	$(55\pm6)\mu m$
liquid entry pressure of water LEP_w	(2.76 ± 0.09) bar
void volume ε	(69 ± 5) %
mean pore size μ_P	198.96 nm
effective porosity ϵ / L_P	7878.1 m ⁻¹
measured total thickness	$165\pm8\mu m$

 Table (3.1) Membrane characteristics

4. Thermostat

Two types of thermostat were used one of them was temptle TE - 8DTechne for feed side to maintain the temperature of feed at required temperature which was 30°C.The second type is a refrigerated bath RB – 12 Techne for permeate side to maintain the temperature of permeate at required temperature which was equal to temperature of feed side.

5. Temperature measurement

To measure the temperature of the feed solution and permeate solution during the time of experiment run. Sensible device by instrument type (Temp – Mebgreat pt 100, Plty we 11759 and Nr 000719) was employed.



6. Conductivity meter

Metrohm Ω 712 Digital conduct meter types 1.712.0010 and Nr. 10191 was used to measure the conductivity of permeate solution.

3.2 The Experimental procedure

The experimental device used in this study was designed as a laboratory scale and allows control of the temperature within range of \pm 0.2°C. The feed tank was filled with a distilled water, the permeate tank was filled with a salt solution. Two types of salt solution were used, CaCl₂ solution (from 1M to 5M) and NaCl solution (from 2M to 5M).

Flat-sheet commercial membrane, TF200 from pall–Gelman was used in this study. It was made of a thin polytetrafluoroethylene (PTFE) macroporous layer supported by a polypropylene (PP) net. The total surface area of the membrane was $2.75 * 10^{-3}$ m² and all membrane characteristics are listed in Table (3.1). The transmembrane flux was calculated by measuring the increase in volume of osmotic agent every 30 minutes. All the experiments were performed for a period of 3 h and the average values of the flux with the standard deviation were reported.



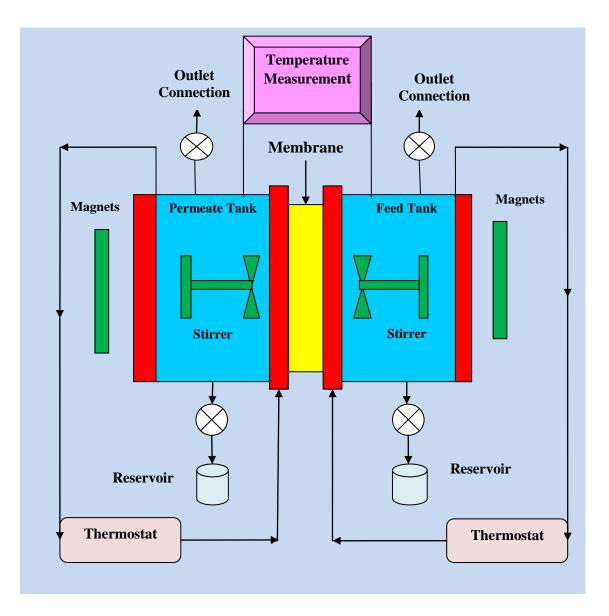


Fig (3.1) Schematic diagram of the experimental rig



Figure (3.2) The general view of the experimental rig

Chapter Four

Mathematical Model

4.1 Introduction

Osmotic distillation (OD) is an isothermal membrane process, which can be operated at ambient temperature and atmospheric pressure. The microporous hydrophobic membrane, which the pores are filled with the gas phase, is used to separate two aqueous solutions. The membrane is contacted with the diluted feed solution on one side and the other side of the membrane is contacted with the extraction solution with a low water activity such as concentrated salt solutions. Hence, the driving force of this process is water activity difference between the two sides of the membrane which induces the water vapour pressure difference and, consequently, the water vapour is transferred through the membrane. The transport process of the water vapour is typically divided into three steps: the evaporation of water at the membrane surface of the diluted solution side, the diffusion of water vapour across the porous media, and the condensation of water vapour in the extraction solution (osmotic solution)[76].

This chapter includes the review of the mathematical model which represented by the mass and heat transfer in the membrane and in the liquids (feed and osmotic solution).

4.2 Mass Transfer

The basic model used to describe water transport in the system is given by Equation (4.1) that relates the mass flux (*N*) to the driving force represented by the vapour pressure difference at both liquid–vapour interfaces of the membrane (ΔP_w^m) , via a proportionality coefficient(K_m) which is considered as membrane permeability:



$$N = K_m \Delta P_w^m \tag{4.1}$$

The driving force depends on the solute concentration as well as on the temperature conditions prevailing at the vapour–liquid interfaces. As such interface conditions are not always accessible; a more complete representation is given by Equation (4.2) which refers to the bulk conditions of the liquid compartments by integrating the various mass transfer resistances:

The global coefficient (*K*) of the system is given by the series of resistances opposed by the feed solution $(1/K_f)$, the membrane $(1/K_m)$ and the permeate solution $(1/K_p)$ (see Figure (4.1a)). A comparable mass transport mechanism is involved in the three different compartments, i.e. mass transfer across a stagnant film: on one hand, either the diluted brine layer contiguous to the condensation side of the membrane or the concentrated layer along the evaporation side; or on the other hand, the film of air supported by pores of the polymer [22].

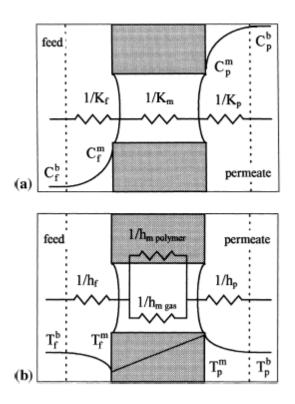


Figure (4.1) (a) Concentration profile and mass transfer resistances in OD, (b) Temperature profile and heat transfer resistances in OD [22].

4.2.1. Mass transfer in the membrane

The water transfer mechanism in gas phase depends on various parameters like operating pressure and membrane structure. As far as OD is concerned, i.e. pores filled with air and atmospheric operating pressure, only two mechanisms can be theoretically involved in the vapour transfer: molecular diffusion and Knudsen diffusion, for which the corresponding membrane permeability is expressed by Equations (4.3) and (4.4), respectively [80]:

$$K_m^M = \frac{M_W}{RT} \frac{\varepsilon D_{Wa}}{X^\delta} \frac{P}{(P_a)_{lm}}$$
(4.3)

$$K_m^K = \frac{M_W}{RT} \frac{2\varepsilon r}{3X^\delta} \sqrt{\frac{8RT}{\pi M_W}}$$
(4.4)

The Knudsen number (*Kn*) defined by Equation (4.5) can be used as a first criterion to determine which of the two diffusion mechanisms will be predominant, by comparing the mean molecular free path (λ) of the diffusing molecule with the mean pore diameter of the membrane:

$$Kn = \frac{\lambda}{2r} \quad with \quad \lambda = \frac{k_B T}{P\sqrt{2}\pi\sigma^2} \qquad \dots \qquad (4.5)$$

According to the gas kinetic theory, λ is expressed as a function of temperature (*T*), pressure (*P*) and mean collision diameter of the molecule (σ). For a relatively small pore size, $Kn \ge 10$, the diffusing molecules tend to collide frequently with the pore walls and Knudsen diffusion is the prevailing mechanism. When the pore size is relatively large, $Kn \le 0.01$, the collisions between the gas molecules themselves are more frequent and molecular diffusion is considered predominant. Between these two limits, both mechanisms will coexist. The combination of different gas transfer mechanisms can be described by the dusty gas model which is a general approach accounting for mass transport in porous media [81].

Where D_W is the water diffusion coefficient and can be estimated by using the following empirical equation [82, 83].



$$D_W = \frac{(117.3 \times 10^{-18})(\varphi M_W)^{0.5}T}{\mu v_A^{0.6}}$$
(4.6)

4.2.2. Mass transfer in liquids

The boundary layers are present in the feed and the osmotic agent on either side of the membrane. These layers may offer significant resistance to mass transfer, depending on the physical properties of the solution (feed and permeate) as well as the hydrodynamic conditions of the systems. The liquid mass transfer coefficients in the boundary layers of feed and osmotic agent (OA) (K_f and K_p) can be estimated by using empirical equations given below, involving only physical properties and hydrodynamic conditions of the solutions [48].

$$Sh = b_1 Re^{b_2} Sc^{b_3}$$
 (4.7)

Where b_1 , b_2 and b_3 are the constants and are to be selected appropriately for the given hydrodynamic conditions, and

$$Sh = \frac{L k_i}{D_W}$$
, $Re = \frac{\rho u L}{\mu}$ and $Sc = \frac{\mu}{\rho D_W}$ (4.8)

Where D_W is the water diffusion coefficient and can be estimated by using equation (4.6), d_h is the hydraulic diameter, u the velocity, ρ the density and μ the dynamic viscosity of the fluid. For the case of a non-circular flow channel, like for instance a square channel, an equivalent diameter is given by d_h=4S/P where S is the cross-sectional area and P the wetted perimeter of the flow channel. In order to obtain K_f and K_p in the same units of K_m the following equation is used

[22],



$$K_i = \frac{k_i C^t M_W}{(x_s)_{lm} \gamma P^*} \tag{4.9}$$

Where C^t is the molar concentration of the solution, γ activity coefficient (Table 4.1 shows, the activity coefficients of osmotic agent solutions at various concentrations) and P^* the saturation vapor pressure, the values of which were obtained from literature [84, 85].

Concentration Activity Activity coefficient coefficient (M) $(CaCl2 \cdot 2H2O)$ (NaCl) 2 0.879 0.679 0.879 3 0.718 0.788 4 0.681 5 0.681 0.878

Table (4.1) Water activity coefficients for the osmotic agents at variousconcentrations [86]

4.3 Heat transfer

The process of OD is considered isothermal as long as no external temperature difference is imposed across the membrane. However, due to the phase changes at the membrane walls, the mass transfer is associated with heat transfer. Evaporation tends to cool down the feed side of the membrane, whereas condensation warms up the brine side. This resulting temperature difference translates into a lower vapour pressure gradient resulting in driving force decay. The latent heat for phase changes has to be transported between the bulk of the solution and the vaporization or condensation interfaces. Figure (4.2b) depicts the heat transfer mechanism in OD as a set of resistances with the temperature profile for the particular case of imposed mean bulk temperature. As was established for

MD [5], the balance of heat transfer in the various compartments of the system is given by relation (4.10) and the overall heat transfer coefficient of the OD process is given by (4.11):

$$Q = H\Delta T^{b} = h_{f} \left(T_{f}^{b} - T_{f}^{m} \right) = N\Delta H_{v} + h_{m} (T_{f}^{m} - T_{p}^{m}) = h_{p} (T_{p}^{m} - T_{p}^{b}) \dots (4.10)$$

$$H = \left[\frac{1}{h_f} + \frac{1}{h_m + \frac{N\Delta H_v}{\Delta T_m}} + \frac{1}{h_p}\right]^{-1}$$
(4.11)

Where Q is the total heat transferred across the membrane, N the molar vapour flux and ΔH_v the mass latent heat of vaporization; h_f and h_p represent the feed and permeate thermal boundary layers and h_m is the heat transfer coefficient of the membrane. In the situation where the bulk temperature is controlled and maintained equal in both liquids, the transmembrane temperature difference ΔT_m is given by (4.12):

The thermal balance of the OD system helps highlighting the fundamental difference with the MD process. In the case of OD, the latent heat of vaporization N ΔH_V is integrally compensated by conduction across the membrane, $h_m \Delta T_m$. Therefore, OD membranes will have to be as heat conductive as possible. In an MD process, the conduction of heat across the membrane is a loss mechanism since it has no corresponding transfer of mass and should thus be minimized [78]. The conduction heat transfer coefficient of the membrane is given by relation (4.13) where the total thermal conductivity k^T is a combination of the

thermal conductivity of the gases — a mixture of air and water vapour — and of the membrane polymer (4.14) [87]:

$$h_m = \frac{k^T}{\delta} \tag{4.13}$$

$$K^{T} = \varepsilon k_{gas}^{T} + (1 - \varepsilon) k_{polymer}^{T} \qquad (4.14)$$

The water vapour pressures within the membrane are not directly measurable, and then it is convenient to express equation (4.15) in terms of temperature. For low values of the transmembrane bulk temperature difference ($T_{b,f}$ - $T_{b,p} \le 10$ K), the following expression may be used as indicated in [50].

$$N = K_m \left(\frac{\mathrm{d}p}{\mathrm{d}t}\right) \left(T_{\mathrm{m,f}} - T_{\mathrm{m,p}}\right) \qquad (4.15)$$

Where (dp / dt) can be evaluated from the Clausius Clapeyron equation, using Antoine equation to calculate the vapour pressure [5, 89].

$$\left(\frac{dp}{dT}\right)_{Tm} = \frac{\Delta H_v}{RT_m^2} \exp(23.238 - \frac{3842}{T_m - 45}\right) \quad \dots \qquad (4.16)$$

4.3.1 Heat transfer in the liquids

The liquid heat transfer coefficients depend on the physical properties of the solutions as well as on the hydrodynamic conditions prevailing in the module. By analogy with what was presented for mass transfer in the liquid compartments, h_f and h_p can be estimated either from experiments or with the help of empirical correlations of dimensionless numbers, like Nusselt (*Nu*), Reynolds (*Re*) and



Prandtl (Pr) numbers. The example given by correlation (4.17) corresponds to the heat transfer coefficient hl for a liquid flow in a tube.

The physical properties such as viscosity and density of the feed and osmotic agent OA solutions were evaluated from literature [48].

Chapter Five Results and Discussion

This chapter illustrates the experimental results of the effect of operating variables on transmembrane flux and analysis the mass and heat transfer phenomena on osmotic membrane distillation. Mass transfer in the boundary layer is estimated as a function of dimensionless numbers, whereas, the mass transfer through the membrane is accounted for based on either by Knudsen or molecular diffusion. The use of dimensionless numbers enables the quantification of the hydrodynamics on the feed as well as OA boundary layers, based on the overall resistance estimated; the model could predict reasonably well the effect of the process parameters on the transmembrane flux.

Finally the mathematical model of osmotic membrane distillation is simulated in the present work to predict the theoretical transient response with aid of computer program using FORTRUN program. The theoretical results are compared with the experimental result of osmotic membrane distillation.

5.1 Effect of osmotic agent concentration and type

The experimental data illustrated by the volume of permeate solution with time and the values of fluxes calculated from the relation between the collected volume of permeate and time are represented in Tables (5.1) to (5.5) for $CaCl_2$ solution and Tables (5.6) to (5.9) for NaCl solution.



Volume	Time (sec)								
(ml)	Zero	0.5 hour	1 hour	1.5 hour	2 hour	2.5 hour	3 hour		
	hour								
0	0	0	0	0	0	0	0		
0.1	119.3333	126	132.3333	138	140.6667	158.6667	158.6667		
0.2	259	280.6667	280.6667	276.6667	278.6667	276	276		
0.3	389	381.3333	402.3333	411	412	391.6667	391.6667		
0.4	531.6667	535	523.6667	541	534.3333	555.3333	555.3333		
Flux (L/m ² .h)	0.9982	0.986528	0.982386	0.961052	0.961544	0.961031	0.954387		
Standard deviation	2.08E-05	7.79E-06	8.87E-06	2.50E-05	1.67E-05	1.47E-05	1.26E-05		

Table (5.2) Volume of permeate solution collected with time for 2M CaCl₂.

Volume	Time (sec)								
(ml)	Zero	0.5 hour	1 hour	1.5 hour	2 hour	2.5 hour	3 hour		
	hour								
0	0	0	0	0	0	0	0		
0.1	54.33333	70.66667	71	61	66	63	61.666666		
0.2	121.6667	138.3333	137.6667	130.3333	158	132.3333	135.33333		
0.3	201.3333	198.6667	207.6667	195	230.6667	207	201		
0.4	280.6667	275	289	272	312	283.3333	276.33333		
Flux	1.934998	1.920436	1.846339	1.96995	1.72538	1.88378	1.923617		
$(L/m^2.h)$	1.754776	1.920+30	1.0+0557	1.90995	1.72550	1.00570	1.925017		
Standard									
deviation	6,11E-05	8.29E-05	4.95E-05	7.87E-05	0.000229	5.53E-05	6.531E-05		

Volume	Time (sec)								
(ml)	Zero	0.5 hour	1 hour	1.5 hour	2 hour	2.5 hour	3 hour		
	hour								
0	0	0	0	0	0	0	0		
0.1	38	39.66667	41	38.33333	44.66667	41	48.666666		
0.2	78	78.66667	82.33333	76.66667	87.66667	85	81		
0.3	121.3333	122	123.6667	123.3333	129.6667	132	118.33333		
0.4	162.6667	164.3333	153.3333	166.3333	167.6667	178.3333	165.66666		
Flux (L/m ² .h)	3.24926	3.218546	3.291779	3.198198	3.075879	2.97188	3.153		
Standard									
deviation	9.49E-05	6.05E-05	4.93E-05	7.09E-05	0.00016	6.85E-05	7.08E-05		

Table (5.4)	Volume of	permeate s	solution	collected	with	time for	· 4M	CaCl ₂ .
				concerea				

Volume	Time (sec)								
(ml)	Zero	0.5 hour	1 hour	1.5 hour	2 hour	2.5 hour	3 hour		
	hour								
0	0	0	0	0	0	0	0		
0.1	29	30	32	32	26.66667	35.66667	39.333333		
0.2	58.66667	62.66667	59.66667	62.33333	60.33333	68.66667	73.666666		
0.3	91.66667	92	98	99	90.33333	110	107.33333		
0.4	123.6667	123.3333	132.3333	134	129.3333	152	143.33333		
Flux	4.283	4.24828	4.02582	3.969346	4.1847	3.52089	3.6435		
$(L/m^2.h)$	4.205	4.24020	4.02302	3.707540	4.1047	3.32007	5.0455		
Standard									
deviation	5.97E-05	9.09E-05	0.000142	0.000173	4.69E-05	0.000116	0.0002331		

Volume	Time (sec)								
(ml)	Zero	0.5 hour	1 hour	1.5 hour	2 hour	2.5 hour	3 hour		
	hour								
0	0	0	0	0	0	0	0		
0.1	26.33333	27.33333	26	26.66667	28.66667	27	30		
0.2	50.66667	48.66667	50.66667	51.66667	56	58.33333	60.666666		
0.3	75.66667	76.33333	78.33333	81.66666	84.66667	89	92		
0.4	100.3333	105	104.3333	109	113	118.6666	120.66666		
Flux (L/m ² .h)	5.197208	5.066519	5.0254	4.84007	4.6416	4.428667	4.3165898		
Standard									
deviation	0.000198	6.99E-05	8.25E-05	6.88E-05	0.000129	0.000141	9.66E-05		

 Table (5.6) Volume of permeate solution collected with time for 2M NaCl.

Volume	Time (sec)								
(ml)	Zero	0.5 hour	1 hour	1.5 hour	2 hour	2.5 hour	3 hour		
	hour								
0	0	0	0	0	0	0	0		
0.1	116.6667	119.3333	107.6667	119	86.33333	95	117.3333		
0.2	239.6667	195.3333	219.3333	235	203	211	256.3333		
0.3	385	289.3333	334	341	314.3333	332.3333	409.3333		
0.4	521.3333	375.3333	450.3333	457.6667	426.6667	474	537.3333		
Flux	1.0226	1.1478	1.173	1.138	1.2769	1.184	0.9787		
$(L/m^2.h)$	1.0220	1.1470	1.175	1.150	1.2709	1.104	0.9707		
Standard									
deviation	2.43E-05	4.00E-05	4.40E-05	4.27E-05	0.0002	0.000217	2.965E-05		

Volume	Time (sec)								
(ml)	Zero	0.5 hour	1 hour	1.5 hour	2 hour	2.5 hour	3 hour		
	hour								
0	0	0	0	0	0	0	0		
0.1	77.66667	80.66667	101	77.33333	84	83	80.333333		
0.2	172	180	195	161.6667	187	173.6667	168.33333		
0.3	248	269.3333	288.3333	258.3333	280.6666	266.3333	265.33333		
0.4	354.6667	354.6666	436.3333	354	364	366.6667	363.66666		
Flux	1.52247	1.471278	1.4524	1.457018	1.4238	1.4536	1.4426		
$(L/m^2.h)$									
Standard									
deviation	4.59E-05	2.91E-05	0.000225	4.56E-05	1.55E-05	6.61E-06	1.28E-05		

Table (5.8) Volume of permeate solution collected with time for 4M NaCl.

Volume	Time (sec)						
(ml)	Zero	0.5 hour	1 hour	1.5 hour	2 hour	2.5 hour	3 hour
	hour						
0	0	0	0	0	0	0	0
0.1	53	58	59	56	59.33333	62	52
0.2	114.3333	125	126.3333	120	123	127.6667	123.6667
0.3	178.3333	188.6666	195.6667	195.6667	195.6667	202	198
0.4	244	267.3333	265.3333	271.3333	273.6667	283.6667	275
Flux	2.18618	2.022	2.0004	1.9897	1.975	1.898	1.9466
$(L/m^2.h)$	2.10010	2.022	2.0004	1.9697	1.975	1.070	1.7400
Standard							
deviation	4.47E-05	0.000102	4.65E-05	5.64E-05	8.05E-05	1.64E-05	1.58E-05

Volume	Time (sec)							
(ml)	Zero	0.5 hour	1 hour	1.5 hour	2 hour	2.5 hour	3 hour	
	hour							
0	0	0	0	0	0	0	0	
0.1	28.33333	42.66667	46.66667	44.66667	50.66667	47.33333	43.666667	
0.2	67	91	83	94.33333	105.6667	100.3333	99.6666667	
0.3	126.6667	146.6667	139.3333	150.3333	159.6667	156	158.33333	
0.4	179.6667	202.3333	204	191	230.6667	215.6667	220	
Flux	3.17855	2.6611	2.69439	2.710187	2.363	2.4408	2.456	
$(L/m^2.h)$								
Standard								
deviation	0.000594	0.000103	9.36E-05	8.62E-05	0.000143	9.22E-05	0.0001522	

Table (5.9) Volume of	permeate solution	collected with	time for 5M NaCl.
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The concentration of osmotic agent (OA) solutions was varied over 1-5M calcium chloride and 2-5M sodium chloride. The concentration of sodium chloride begins from 2M because in 1M there is no flux. The relationship between transmembrane flux and time is represented in Figures (5.1) to (5.5) for CaCl₂ and Figures (5.6) to (5.9) for NaCl. The transmembrane flux decreased with time and the best transmembrane flux is found at the first minutes of the experiment. This is attributed to the membrane fouling occuring in the first few minutes of the experiment. It means that, the concentrations of CaCl₂ and NaCl solutions are important factor affecting strongly the speed of membrane fouling [90, 91].



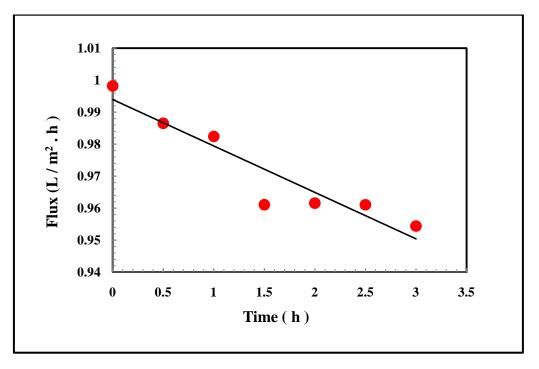


Figure (5.1) Flux of permeate solution with time for 1M CaCl₂.

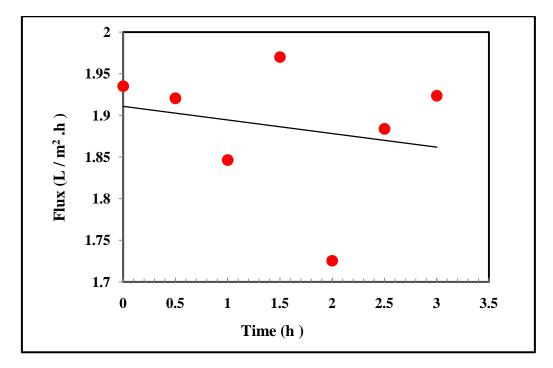


Figure (5.2) Flux of permeate solution with time for 2M CaCl₂.



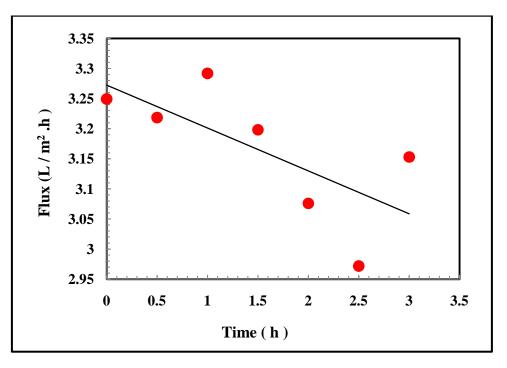


Figure (5.3) Flux of permeate solution with time for 3M CaCl₂.

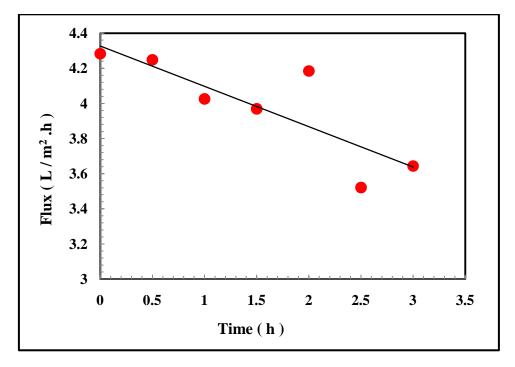


Figure (5.4) Flux of permeate solution with time for 4M CaCl₂.



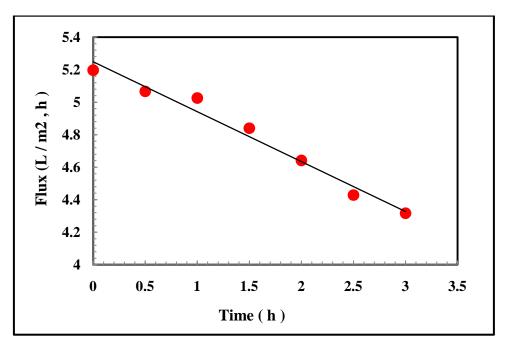


Figure (5.5) Flux of permeate solution with time for $5M CaCl_2$.

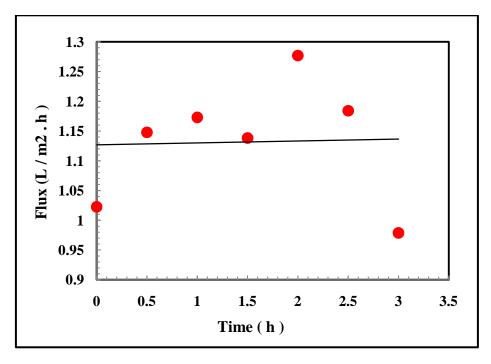


Figure (5.6) Flux of permeate solution with time for 2M NaCl.



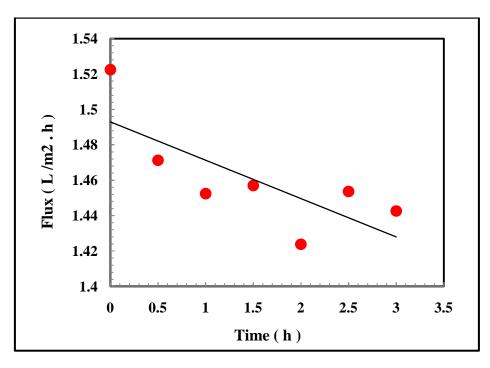


Figure (5.7) Flux of permeate solution with time for 3M NaCl.

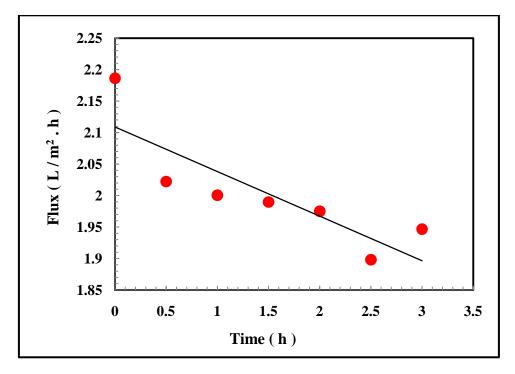


Figure (5.8) Flux of permeate solution with time for 4M NaCl.



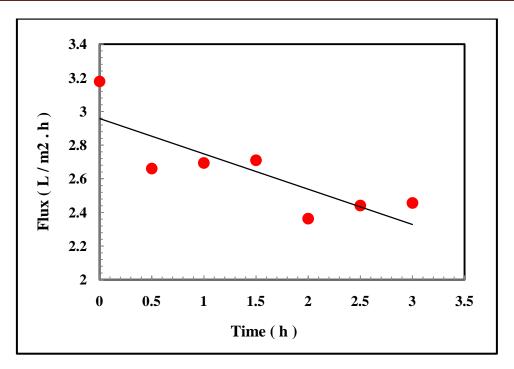


Figure (5.9) Flux of permeate solution with time for 5M NaCl.

The values of transmembrane flux observed at different concentrations of osmotic solution are shown in Figures (5.10) and (5.11). In both cases CaCl₂ and NaCl the transmembrane flux is increased with an increase of osmotic agent (OA) concentration. This is expected and attributed to the increase in vapor pressure difference across the membrane with an increase in the concentration of osmotic agent solution, which results in an increase in the driving force for water transport through the membrane as reported by Naveen et al [46]. The calcium chloride has showed higher transmembrane flux at all the concentrations when compared to that of sodium chloride. This is mainly due to the higher osmotic activity (ratio of its water solubility to its equivalent weight) of CaCl₂, which is resulted in higher vapor pressure gradient across the membrane [46]. Table (5.10) gives a list of membranes used either on the laboratory scale or on a pilot plant scale and the values of flux obtained. In laboratory studies flat membranes are usually chosen



whereas in industrial equipments hollow fiber or spiral wound modules are generally preferred.

Table (5.10)	Membrane	used	and	fluxes	obtained	in	osmotic	evaporation
process.								

Reference	Distillation	Liquid system	Membranes	Temperature	Hydrodynamic	Fluxes
	type				conditions	obtained
						(L/m ² .h)
Sheng [65]	OE	Fruit(orange,app	Plate and frame	ambient	Counter-	0.4-7.9
		le,grape)juices/c	modules		current feed	(laboratory)
		onc. NaCl	(membrane		stream	0.02-2.8
		solutions	surface area up			(pilot plant)
			to 18 cm2 pore			
			radius,0.1< r			
			<0.45 µm;			
			polypropylene			
			hollow fiber			
			membranes			
Sheng et al.	OE	Fruit(orange,app	PTFE	29 – 40 °C	Counter-	$0 \rightarrow \approx 2.2$
[53]		le,grape)juices/c	membrane (r =		current flow,	for osmotic
		onc. NaCl	0.1		juice flow rate	pressure
		solutions (> 280	µm,thickness		5.8 l/min,	differences
		g NaCl per kg	l=100 μm),		brine flow rate	between 280
		solution)	syrinx plate and		1 l/min	and 416 atm
			frame			
			configuration,			
			membrane size			
			0.7 m ²			
Mengual et	OE	Pure water /	Millipore	10 - 60 °C	Agitation,	0 - ≈ 0.5
al. [58]		NaCl solutions	PVDF	0 – 350 rpm	stirring rates	
		(0-5 mol/l)	(GVHP),r = 0.2	Lewis cell		
			μm ,l = 125 μm			
			, porosity p =			
			70 % ;			
			Millipore PTFE			
			(FHLP),r = 0.2			

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um, l = 175 µm , porosity p = 70 %; Gelman PTFE (TF- 1000), r = 1 µm J = 178 µm, porosity p = 80 %; Gelman PTFE (TF- 450), r = 0.45 µm, l = 178 µm , porosity p = 80 %; Gelman PTFE (TF- 200), r = 0.45 µm, l = 178 µm , porosity p = 80 %; Gelman PTFE (TF- 200), r = 0.2 µm, l = 178 µm , porosity p = 80 %; Gelman PTFE (TF- 200), r = 0.2 µm, l = 178 µm , porosity p = 80 %; membrane surface area : 27.5 cm²Co-current flow, 0.02- depending depending dependingVahdati et al. [61]Coupled Water or dilute distilland / saturatedParts and the solution membrane (material not indicated)20-33 °C flow, 0.02- flow, 0.02- depending depending distilland / hydrophobic0.20-3.3 °C flow, 0.02- depending depending depending distilland / hydrophobic0.18 l/min. on the absolute (0.18 l/min.Godino et al. [62]Coupled Water / NAC1Millipore PTFE m mean temp. Agitation, 30 - 50 °C. stirring rates:0.2 in both							
$\begin{tabular}{ c c c c c c c c c c c c c c c c c c c$				μm ,l = 175 μm			
Vahdati et Coupled Water / NaCl Flat sheet of 60 indicated) 20 – 33 °C μm thick co-current flow, 0.02- temp. and on the applied temperature difference (-0-10 K)				, porosity p =			
Vahdati etCoupledWater or diluteFlat sheet of 6020 – 33 °Cco-current0.2–3.3dependingMillipore PTFEMean temp.Agitation,0-2 in both				70 % ; Gelman			
Vahdati etCoupledWater or diluteFlat sheet of 60 istilland / saturated20 – 33 °C mmbrane surface area : 27.5 cm²co-current no the absolute0.2–3.3 depending distilland / istilland / solutions0.2–3.3 indicated)Vahdati etCoupledWater or diluteFlat sheet of 60 mmbrane surface area : 27.5 cm²20 – 33 °C no the absoluteco-current depending on the absoluteVahdati etCoupledWater or dilute istilland / solutionsFlat sheet of 60 indicated)20 – 33 °C no the 				PTFE (TF-			
Vahdati etCoupledWater or diluteFlat sheet of 60 hydrophobic $20 - 33$ °Cco-current flow, 0.02- depending $0.2-3.3$ dependingVahdati etCoupledWater or diluteFlat sheet of 60 hydrophobic $20 - 33$ °Cco-current flow, 0.02- depending $0.2-3.3$ dependingVahdati etCoupledWater or diluteFlat sheet of 60 hydrophobic $20 - 33$ °Cco-current flow, 0.02- depending $0.2-3.3$ dependingVahdati etCoupledWater or diluteFlat sheet of 60 hydrophobic $20 - 33$ °Cco-current flow, 0.02- dependingal. [61]OE/MDaqueous food distilland / saturatedmembrane membrane surface area : 27.5 cm ² flow, 0.02- dependingMgSO ₄ (material not indicated)number: 2-53 itemp. and on the applied temperature difference (0-10 K)o.18 l/min. down the applied temperature				1000), r = 1 μm			
Vahdati et Coupled Water or dilute Flat sheet of 60 hydrophobic 20 – 33 °C co-current 0.2–3.3 depending Vahdati et Coupled Water or dilute Flat sheet of 60 hydrophobic 20 – 33 °C co-current 0.2–3.3 depending al. [61] OE/MD aqueous food distilland / bydrophobic Flat sheet of 60 hydrophobic 20 – 33 °C co-current flow, 0.02- bydrophobic 0.2–3.3 depending MgSO ₄ (material not solutions indicated) membrane bydrophobic Reynolds number: 2-53 temp. and on the applied temperature difference (0-10 K)				$,l = 178 \ \mu m$,			
Vahdati etCoupledWater or diluteFlat sheet of 60 um distilland / $20 - 33 \ ^{\circ}\text{C}$ co-current flow, 0.02- $0.2 - 3.3$ dependingVahdati etCoupledWater or diluteFlat sheet of 60 um tick $20 - 33 \ ^{\circ}\text{C}$ co-current flow, 0.02- $0.2 - 3.3$ dependingVahdati etCoupledWater or dilute um tickFlat sheet of 60 um tick $20 - 33 \ ^{\circ}\text{C}$ co-current flow, 0.02- $0.2 - 3.3$ dependingVahdati etCoupled um tickWater or dilute issurface area : $27.5 \ cm^2$ flow, 0.02- um tickdepending depending0.16 I/min.on the aqueous food distilland / saturated membranemembrane issurface areaReynolds absolute0.2 - 3.3Godino etCoupledWater / NaClMillipore PTFEMean temp.Agitation , o.2 in both				porosity p = 80			
$\begin{tabular}{ c c c c c c c c c c c c c c c c c c c$				% ; Gelman			
Vahdati et Coupled Water or dilute Flat sheet of 60 20 – 33 °C co-current 0.2–3.3 dageous food μm thick flow, 0.02- depending on the al. [61] OE/MD aqueous food μm thick flow, 0.02- depending MgSO4 (material not membrane saturated membrane saturated number: 2-53 temp. and on MgSO4 indicated) MgSO4 (material not number: 2-53 temp. and on the applied Godino et Coupled Water / NaCI Millipore PTFE Mean temp. Agitation, 0-2 in both				PTFE (TF-			
vahdati et Coupled Water or dilute Flat sheet of 60 20 – 33 °C co-current 0.2–3.3 al. [61] OE/MD aqueous food µm thick flow, 0.02- depending distilland / hydrophobic 0.18 1/min. on the saturated membrane absolute number: 2-53 temp. and on MgSO4 indicated) indicated) number: 2-53 temp. and on MgFrence Water / NaCl Millipore PTFE Mean temp. Agitation , 0-2 in both				450), r = 0.45			
Water / NaCl Water / NaCl<				μm ,l = 178 μm			
Vahdati etCoupledWater or diluteFlat sheet of 60 indicated)20 - 33 °Cco-current0.2-3.3 dependingVahdati etCoupledWater or diluteFlat sheet of 60 indicated)20 - 33 °Cco-current0.2-3.3 dependingVahdati etCoupledWater or diluteFlat sheet of 60 indicated)20 - 33 °Cco-current0.2-3.3 dependingVahdati etCoupledWater or diluteFlat sheet of 60 indicated)20 - 33 °Cco-current0.2-3.3 dependingVahdati etCoupledWater or diluteFlat sheet of 60 indicated)20 - 33 °Cco-current0.2-3.3 dependingVahdati etCoupledWater or diluteFlat sheet of 60 indicated)20 - 33 °Cco-current0.2-3.3 dependingon the saturatedmembraneReynoldsabsoluteMgSO4(material not indicated)number: 2-53temp. and on the applied temperature difference (0-10 K)Godino etCoupledWater / NaClMillipore PTFEMean temp.Agitation , 0-2 in both				, porosity p =			
$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$				80 % ; Gelman			
$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$				PTFE (TF-			
, porosity p = 80 %; membrane surface area : 27.5 cm², porosity p = 80 %; membrane surface area : 27.5 cm², porosity p = 80 %; membrane 27.5 cm², porosity p = 80 %; membrane 27.5 cm², porosity p = 80 %; membrane, poros				200), r = 0.2			
Wahdati et Coupled Water or dilute Flat sheet of 60 20 – 33 °C co-current 0.2–3.3 Vahdati et Coupled Water or dilute Flat sheet of 60 20 – 33 °C co-current 0.2–3.3 al. [61] OE/MD aqueous food µm thick flow, 0.02- depending distilland / hydrophobic 0.18 l/min. on the saturated membrane Reynolds absolute MgSO4 (material not number: 2-53 temp. and on solutions indicated) indicated) temperature difference Godino et Coupled Water / NaCl Millipore PTFE Mean temp. Agitation, 0-2 in both				μm ,l = 178 μm			
Mathematical Ma				, porosity p =			
Vahdati etCoupledWater or diluteFlat sheet of 6020 – 33 °Cco-current0.2–3.3al. [61]OE/MDaqueous foodµm thickflow, 0.02-dependingdistilland /hydrophobic0.18 l/min.on thesaturatedmembraneReynoldsabsoluteMgSO4(material notnumber: 2-53temp. and onsolutionsindicated)indicated)itemperatureGodino etCoupledWater / NaClMillipore PTFEMean temp.Agitation ,Ocino etCoupledWater / NaClMillipore PTFEMean temp.Agitation ,0-2 in both				80 % ;			
Vahdati et al. [61]Coupled OE/MDWater or dilute aqueous food distilland / saturatedFlat sheet of 60 µm thick20 – 33 °C co-currentco-current flow, 0.02- depending 0.18 l/min.0.2–3.3 dependingal. [61]OE/MDaqueous food distilland / saturatedµm thick hydrophobicflow, 0.02- 0.18 l/min.depending absoluteal. [61]OE/MDaqueous food distilland / saturatedhydrophobic membrane0.18 l/min. number: 2-53on the temp. and on the applied temperature difference (0-10 K)Godino etCoupledWater / NaClMillipore PTFEMean temp.Agitation , 0-2 in both				membrane			
Vahdati et al. [61]Coupled OE/MDWater or dilute aqueous food distilland / saturatedFlat sheet of 60 µm thick20 – 33 °C co-currentco-current flow, 0.02- dependingal. [61]OE/MDaqueous food distilland / saturatedµm thick hydrophobicflow, 0.02- 0.18 l/min.depending absoluteMgSO4 solutions(material not indicated)membrane indicated)Reynolds temp. and on the applied temperature difference (0-10 K)Godino etCoupledWater / NaClMillipore PTFEMean temp.Agitation , ol-2 in both				surface area :			
al. [61]OE/MDaqueous food distilland / saturatedμm thick hydrophobicflow, 0.02- 0.18 l/min.depending on the absoluteal. [61]OE/MDaqueous food distilland / saturatedhydrophobic membraneflow, 0.02- 0.18 l/min.depending on the absoluteal. [61]OE/MDsaturatedmembraneReynoldsabsoluteMgSO4 solutions(material not indicated)number: 2-53temp. and on the applied temperatureal. [61]CoupledWater / NaClMillipore PTFEMean temp.Agitation ,0-2 in both				27.5 cm^2			
distilland / hydrophobic 0.18 l/min. on the saturated membrane Reynolds absolute MgSO4 (material not number: 2-53 temp. and on solutions indicated) the applied temperature difference (0-10 K) (0-10 K) 0-2 in both	Vahdati et	Coupled	Water or dilute	Flat sheet of 60	20 – 33 °C	co-current	0.2–3.3
Saturated membrane Reynolds absolute MgSO4 (material not number: 2-53 temp. and on solutions indicated) temp. and on the applied Low Low Low Low Low Godino et Coupled Water / NaCl Millipore PTFE Mean temp. Agitation , 0-2 in both	al. [61]	OE/MD	aqueous food	μm thick		flow, 0.02-	depending
MgSO4 (material not indicated) number: 2-53 temp. and on the applied temperature difference (0-10 K) Godino et Coupled Water / NaCl Millipore PTFE Mean temp. Agitation , 0-2 in both			distilland /	hydrophobic		0.18 l/min.	on the
solutions indicated) the applied indicated) indicated) the applied indicated) indicated) indicated) indicated) inditindicated) indicated)			saturated	membrane		Reynolds	absolute
Godino et Coupled Water / NaCl Millipore PTFE Mean temp. Agitation , 0-2 in both			$MgSO_4$	(material not		number: 2-53	temp. and on
Godino et Coupled Water / NaCl Millipore PTFE Mean temp. Agitation , 0-2 in both			solutions	indicated)			the applied
Godino etCoupledWater / NaClMillipore PTFEMean temp.Agitation ,0-2 in both							temperature
Godino et Coupled Water / NaCl Millipore PTFE Mean temp. Agitation , 0-2 in both							difference
							(0-10 K)
al. [62] OE/MD solutions (0-5 , $r = 0.25 \mu m$, 1 30 – 50 °C. stirring rates: directions	Godino et	Coupled	Water / NaCl	Millipore PTFE	Mean temp.	Agitation,	0-2 in both
	al. [62]	OE/MD	solutions (0-5	,r = 0.25 μ m ,1	30 – 50 °C.	stirring rates:	directions
mol/l) = 175 μ m, Temp. 0-350 rpm depending			mol/l)	$= 175 \ \mu m$,	Temp.	0-350 rpm	depending
porosity $p = 80$ differences: Lewis cell on the sense				porosity $p = 80$	differences:	Lewis cell	on the sense
% $-20 < \Delta T <$ of the				%	$-20 < \Delta T <$		of the
20 °C applied					20 °C		applied
temp.							temp.
difference							difference

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Chapter Five

Results & Discussion

Durham and	OE	Water / aqueous	PTFE	ambient	Counter-	0.6-1.4
Nguyen [64]		solutions	membranes:Gor		current flow	depending
		containing 28%	e-Tex 10387 ,r		(500 ml/min)	on
		NaCl	$= 0.1 \ \mu m$,l =			membrane
			8.5 μm ,			material and
			porosity p = 78			cleaning
			% ; Gelman			procedures
			11104/2 TPR, r			
			$= 0.1 \ \mu m$, $l = 9$			
			μm , porosity p			
			not indicated			

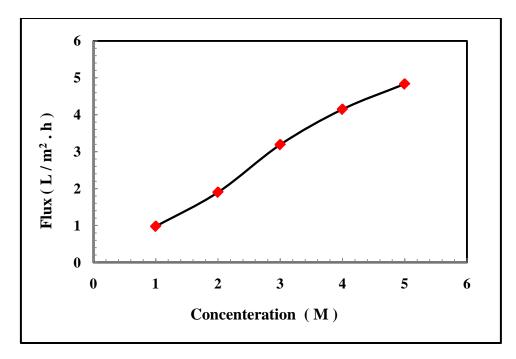


Figure (5.10) Effect of osmotic agent (OA) calcium chloride $CaCl_2$ concentration on transmembrane flux.



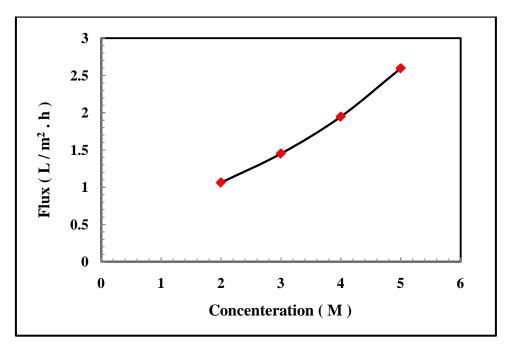


Figure (5.11) Effect of osmotic agent (OA) sodium chloride NaCl concentration on transmembrane flux.

5.2 Model validation

To validate the model in case of real systems, theoretical fluxes were estimated by accounting the individual mass transfer coefficient for boundary layers (feed and osmotic agent OA) as well as for membrane as shown in Table (5.10). In order to estimate the water transport through the boundary layers (feed as well as osmotic agent OA side), empirical correlation comprising of dimensionless numbers (Equation (4.7)) was used and the membrane module employed is flat. The values of the constants in Equation (4.7) are considered as $b_1 = 0.027$, $b_2 = 4/5$ and $b_3 = 0.4$ [47]. Mass transport of water through membrane has been estimated based on mode of diffusion mechanism in the pores by Knudsen or molecular diffusion (Equation (4.3) or (4.4)). It may also be noted that the mechanism of mass transfer in the membrane could not be clearly pointed out to be either Knudsen or molecular [22, 45]. This may be mainly due to the fact that the membranes employed by those researchers are composite type where mechanism will be

different in the active layer compared to the support layer. In the present case of supported membranes, it was observed that, the molecular to be the mode of diffusion when Knudsen number < 0.01 when calculated from equation (4.5).

The thermal boundary layers (feed and osmotic agent OA) were estimated from empirical correlation of dimensionless numbers (equation (4.17)), and the heat transfer of the membrane was estimated from equation (4.13) where the total thermal conductivity was estimated from equation (4.12), where the thermal conductivity of the gases $k_{gas}^{T} = 0.027$ W/m.K and the thermal conductivity of the membrane polymer $k_{polymer}^{T} = 0.22$ W/m.K [88, 89].

Concentration (M)	$K_{p} (\times 10^{4} \text{ ms}^{-1})$					
a. For $CaCl_2$	· · · ·					
1	2.501959					
2	3.092461					
3	3.936354					
4	5.476957					
5	8.627225					
b. For NaCl						
2	5.425339					
3	5.371074					
4	5.220535					
5	5.028386					
$K_{mm} = 1.196276E-02 \text{ kg m}^{-2} \text{ h}^{-1} \text{ Pa}^{-1}.$						
$K_{\rm f} = 4.834239 * 10^{-5} {\rm m s^{-1}}$						

Table (5.11) Valu	es of mass	s transfer	coefficient a	at different	concentrations of
osmotic agent.					



5.3 Simulation Results

Theoretical values of the transmembrane flux were estimated after calculating the overall mass transfer resistance (membrane plus boundary layers) and water vapour pressure within the membrane. Simulation was done with the aid of FORTRUN computer program as shown in appendix (A).

The theoretical results were compared with the experimental results for the process variable. Figures (5.12) and (5.13) represent the comparison between experimental and theoretical transmembrane flux for calcium chloride $CaCl_2$ and sodium chloride NaCl respectively. It can be seen that, there are small deviations between the theoretical and experimental results. Naveen Nagaraj, reported that the observed deviations of the predicted values from the experimental values of the transmembrane flux could be attributed to uneven pore distribution, geometry of the membrane and complex hydrodynamic nature of the boundary layer (feed and OA) [46].



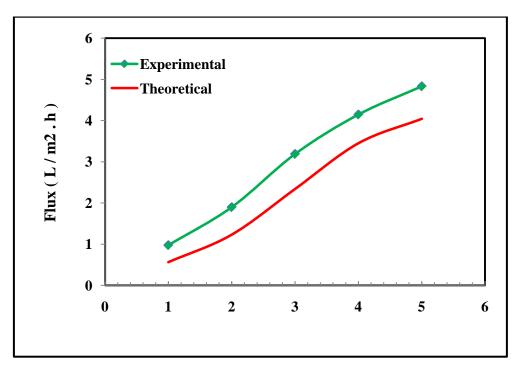


Figure (5.12) Comparison between theoretical and experimental transmembrane flux for $CaCl_2$.

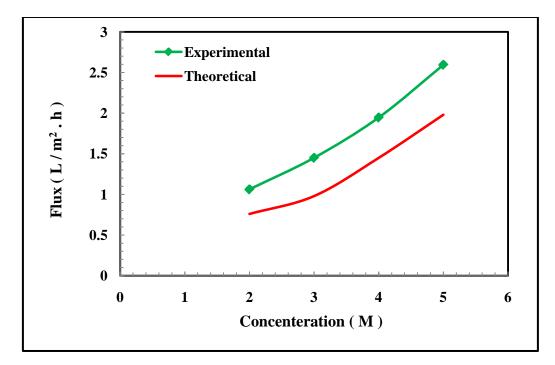


Figure (5.13) Comparison between theoretical and experimental transmembrane flux for NaCl.



Chapter Six

Conclusions and Recommendations

6.1 Conclusions

1. Classical gas and liquid mass transfer mechanisms were tested to simulate the performance of OD of pure water at 30 $^{\circ}$ C with commercial membrane TF200 and with calcium chloride solutions of (1- 5) M salt concentration and sodium chloride of (2-5) M salt concentration.

2. The influence of the process parameters, such as osmotic agent concentration, on transmembrane flux was studied for real system. In the range of operating conditions and maximum temperature difference was only about 0.2 °C.It was found that the transmembrane flux increased with increasing the osmotic agent concentration and the calcium chloride has showed higher transmembrane flux from sodium chloride.

3. The membrane mass transfer coefficient was described by molecular diffusion model and was estimated to be 1.196276E-02 kg m⁻² h⁻¹ Pa⁻¹. It was observed that the mass transfer mechanism was in the molecular diffusion region when Knudsen number < 0.01.

4. The empirical correlation comprising of dimensionless numbers was used to estimate the mass transfer coefficient for boundary layers (feed and osmotic agent OA).

5. Theoretical values of the transmembrane flux could be estimated after calculating the overall mass transfer resistance (membrane plus boundary layers) and water vapour pressure within the membrane .The values of experimental transmembrane flux were found to be in good agreement with the predicted values.



6.2 Recommendations for Future work

1. Studying the effect of flow rate of osmotic agent solution and feed flow rate on transmembrane flux.

2. Other types of membrane can be investigated and the effect of pore size distribution on transmembrane flux can be studied.

3. Different types of membrane module can be employed such as hollow fibre membrane, shell and tube membrane and spiral wound membrane.

4. Studying the effect of concentration polarization and temperature polarization.

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*****FORTRUN PROGRAM******

IMPLICIT NONE REAL::K,Kf,Km,KP REAL::P,KB,T,SEC,Lmd,r,Kn REAL::PIA REAL::VA,FIA,meu,MW,Dwa,Zet,X,del,PaL REAL::b1,b2,b3,Re,Sc,ROU,Dh,U,Ph,S,Sh REAL::Dwas,MEUs,MWs,Res,ROUs,Scs,Shs,Ki REAL::Ct,Xs,GAMA,PS REAL::dHV,DPDT,Ks,Kg,Kt,NP,hf,hp,hm,DTm,N REAL::CP,CPs,Prf,Prp,Nup,Nuf INTEGER::I,NI OPEN(1,FILE='INPUT.DAT') OPEN(2,FILE='OUT.PLT') OPEN(3,FILE='OUT2.PLT') NI=5 PIA=22.0/7.0 P=1E5 KB=1.3807E-23 T=303.0 SEC=2.641 r=198.96E-9 !-----MW=18.0 R=8.314 meu=7.708E-7*995.0 CP=75.282 FIA=2.26 VA=0.0756 Zet=0.69 X=2.0 del=55.E-6 !-----U=0.7 ROU=995. S=2.75E-3 Ph=0.1858 Dh=4*S/Ph b1=0.027 b2=4./5. b3=0.4 !-----MEUs=1.35E-3 MWs=58.5 ROUs=1.26E3 !----dHV=2220.8975E3 Kg=0.027



 $K_{s=0.22}$!NP=3.6*0.00198129/(2.75*10e-3) !Ct=1. !Xs=1.768 !GAMA=0.65937 !PS=4.10367 DO I=1.NI READ(1,*)Ct,GAMA,PS,Xs,CPs,NP NP=NP*3.6/(2.75*10e-3) ******* Lmd=KB*T/(P*SQRT(2.)*PIA*SEC**2) PRINT*,"Lmd=",Lmd Kn=Lmd/2.0/r PRINT*,"Kn=",Kn Dwa=117.3E-18/MEU*(FIA*MW)**0.5*T/VA**0.6 PaL=(LOG10(1E5)+LOG10(3.4E5))/2.0 PRINT*,"Dwa=",Dwa IF(Kn>=10)THEN Km=Mw/R/T*2*Zet*r/(3*X**del)*SQRT(8*R*T/PIA/MW) ELSEIF(Kn<=0.01)THEN Km=MW/R*T*Dwa*Zet/(X**del)*P/paL **ENDIF** PRINT*,"Km=",Km ******* Re=U*Dh*ROU/MEU Sc=MEU/ROU/Dwa Sh=b1*Re**b2*Sc**b3 PRINT*,"Sh=",Sh Kf=Sh*Dwa/Dh

PRINT*,"KfDPDT=",Kf

Dwas=117.3E-18/MEUs*(FIA*MWs)**0.5*T/VA**0.6 Res=U*Dh*ROUs/MEUs Scs=MEUs/ROUs/Dwas Shs=b1*Res**b2*Scs**b3 PRINT*,"Shs=",Shs Ki=Shs*Dwas/Dh PRINT*,"KI=",KI KP=Ki*Ct*MWs/(Xs*GAMA*PS) K=(1./Kf+1./Km+1./Kp)**(-1) PRINT*,"KP=",KP

DPDT=dHV/R/T**2*EXP(23.238-3841/(T-45)) PRINT*,"DPDT=",DPDT



KT=Zet*Kg+(1-Zet)*Ks hm=KT/del

Prf=meu*CP/KT PRINT*,KT

Nuf=0.027*Re**(4/5.)*Prf**0.3 hf=Nuf*KT/Dh

Prp=meus*CPs/KT Nup=0.027*Re**(4/5.)*Prp**0.3 hp=Nup*KT/Dh

DTm=NP*DHV/(hm+(1/hf+1/hp)**(-1))

N=K*DPDT*DTm/1000. PRINT*,"DTm=",DTm WRITE(3,*)Ct,Km,dpdt,k,DPDT WRITE(2,"(5F15.8)")Ct,K,DTm,N,NP ENDDO

END PROGRAM NISREEN



تقطير الاغشية التنافذية هو عملية مبتكرة باستخدام الاغشية لإزالة الماء من المحاليل المائية المخفّفة مثل الأطعمة السائلة أو الألوان الطبيعية بتركيزها مع الاحتفاظ بخصائصها الغذائية والعضوية. تم تطبيق التجارب باستخدام نظام حقيقي (ماء نقي) على وحدةِ غشاءِ مستوى نوع (TF200). تم در اسة تاثير محلول الكاشف التنافذي على اداء جريان الغشاء حيث تم استخدام نوعين من المحاليل الملحية وهي كلوريد الكالسيوم بتراكيز مختلفة (من 1 الي 5 مولاري) وكلوريد الصوديوم بتراكيز (من 2 الى 5 مولاري) وقد لوحظ ان اعلى قيم للجريان تكون عند التراكيز العالية وبالمقارنة بين كلا الملحين فان الجريان في حالة كلوريد الكالسيوم يكون اعلى في حالة كلوريد الصوديوم. تم ايجاد معامل انتقال الكتلة للمحلول الداخل (feed) ومحلول الكاشف التنافذي (osmotic agent) باستخدام العلاقات التجريبية الكلاسيكية . وقد تم تطبيق الانتشار الجزيئي وانتشار ندسن لانتقال البخار خلال الغشاء التنافذي باستخدام الخصائص النوعية للغشاء وجد ان ميكانيكية انتقال الكتلة يكون ضمن منطقة الانتشار الجزيئي وذلك لان قيمة عدد ندسن يكون اقل من 0.01 وتم تطبيق انتقال الحرارة المكملة لمعادلات انتقال الكتلة المصاحب لانتقال بخار الماء خلال الغشاء لايجاد القيم النظرية للجريان وبالمقارنة مع القيم العملية وجد ان هناك تطابق جيد بين القيم النظرية والعملية.



وزارة التعليم العالي والبحث العلمي الجامعة التكنولوجية قسم الهندسة الكيمياوية

تناظر انتقال الكتلة والحرارة في عملية تقطير الاغشية التنافذية

رسالة مقدمة إلى قسم الهندسة الكيمياوية في الجامعة التكنولوجية وهي جزء من متطلبات نيل شهادة دكتوراه فلسفة في الهندسة الكيمياوية

من قبل نسرين صباح على العزاوي (ماجستير هندسة كيمياوية)

۵۵۵۹م کویکی 1430 کری کی کی کی کی کی کی کی ک