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REVIEW

Direct Contact Membrane Distillation

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ABSTRACT

This review looks at the work carried out over the past 15 years on membrane distillation and reports the conditions utilized for research. The process is still used mainly at the laboratory scale, but a few pilot plants have been built across the world, mostly for desalination and the production of potable water. Studies into membrane distillation have been concerned with the effect of mass transfer, heat transfer, and stirring rate, but the most important effect that has to be considered with this process is temperature polarization. A section on temperature polarization and the effect of boundary layers is included in this review.

INTRODUCTION

Membrane distillation has been in use for around thirty years and is currently used mostly at the laboratory scale, with relatively few pilot plants in use around the world. This current review is concerned with research carried out in the last 15 years on the process of direct contact membrane distillation. For a look at the historical perspective of earlier work and other forms of membrane distillation, the review by Lawson (1) should be consulted.

Direct contact membrane distillation uses a porous, hydrophobic membrane with liquids in direct contact with both surfaces of the membrane. The driving force is a temperature-induced vapor pressure difference caused by having a

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hot feed and a cold permeate. The mass transfer is accompanied by the transfer of the corresponding latent heat plus the conductive heat leak through the membrane.

The mechanism of transport from the feed to the permeate can be split into three steps. As the membrane is hydrophobic, liquid cannot enter the pores of the membrane, therefore the first step is evaporation of the volatile components at the hot membrane interface. The second is the transport of vapor through the microporous system of the membrane. The final stage is condensation of the vapor at the cold membrane interface (2). The main developments in the process were made in the early 1980s when newer, more suitable membranes became available, for instance hydrophobic polytetrafluoroethylene (PTFE).

Separations which can be achieved using membrane distillation include cases where the permeate is the product (desalination, water reclamation), the concentrated feed is the product (liquid foodstuffs), and both feed and permeate are products (azeotropic separation).

This review considers aspects of direct contact membrane distillation other than those mentioned in the review by Lawson (1). These include a look at the experimental work carried out on this process, the membranes actually used, and a section on the effect of boundary layers.

THEORETICAL MODELING

Much work has been carried out on the modeling of membrane distillation. The modeling falls into two distinct areas. Membrane modeling, which looks at the mechanism of transport of permeate through the membrane, and overall modeling which deals with predicting the permeate flux produced at specific operating conditions.

Membrane Modeling

Resistance to mass transfer through the membrane comes from the presence of air trapped in the membrane and the membrane structure, i.e., pore size, porosity, and tortuosity (6). The resistance to the flow of vapor from the membrane structure can be described by Knudsen diffusion or Poiseuille flow or a combination of both (7). In most applications of membrane distillation, water is the component transported through the membrane. The molecular mean free path for water vapor at 60°C is approximately 0.3 μm (7), which is around the pore size distribution of the membranes used for membrane distillation. This means that both Knudsen and Poiseuille flows have to be considered for describing the flow of vapor through the membrane (6). Poiseuille flow is only dominant when the pore size is larger than the mean free molecular path. The equations developed to describe the above vapor transport suggest Eq. (1) as an overall, basic equation.

$$N = C(P_1 - P_0) \tag{1}$$

Schofield et al. (7) produced a model that is a combination of Knudsen and Poiseuille flows, which shows that the membrane mass transfer coefficient, C, is slightly temperature dependent, decreasing 3% with a 10°C increase in mean temperature. Peña et al. (9, 10) also found that the coefficient decreased with increasing mean temperature. This suggests that for water flux through 0.2 to 1.0 μm pores, Knudsen flow is dominant. The membrane mass transfer coefficient can also be affected by pressure, but for most cases the membrane mass transfer coefficient is mainly constant (11, 12).

The morphology of a microporous membrane can be difficult to measure, which leads to inaccuracy when calculating the mass transfer (13). Schofield et al. (7) state the morphology term must be estimated experimentally. Experimental values can be obtained by measuring the evaporation rate of water at varying temperatures and pressures (6). Martínez-Díez and Vázquez-González (12) looked at mercury porometry and the liquid displacement methods for measuring the pore sizes of suitable membranes. Both methods can produce good results but are not consistent. The appropriate method is determined by the type of membrane used.

Assumptions of tortuosity are more common. Tortuosity is a measure of the straightness of a pore from one side of the membrane to the other (1.0 means a straight pore with the actual length of the pore equal to the membrane thickness). Calabrò et al. (14) showed that assuming a tortuosity value of 1.2 for calculating the theoretical flux leads to a correlation of around 99.6% with experimental results.

Overall Modeling

Mass Transfer

In membrane distillation only water and volatile solutes can cross the membrane in the vapor phase (15). Because the entrainment of dissolved particles is avoided, a permeate with high purity is obtained (16).

Mass transfer in membrane distillation occurs by diffusive transport of water vapor as specified by the ratio of pore size to molecular mean free path, and the convective transport due to the temperature-induced driving force.

The vapor pressure difference term in Eq. (1) can be replaced by a temperature difference term to give, for dilute solutions,

$$N = C \left.\frac{dP}{dT}\right|_{T_m} (T_1 - T_0) \tag{2}$$

and, for concentrated solutions,

$$N = C\frac{dP}{dT}[(T_1 - T_0) - \Delta T_{\text{th}}](1 - x_{\text{m}}) \tag{3}$$

where

$$\Delta T_{\text{th}} = \frac{RT^2}{M\lambda}\frac{x_1 - x_0}{1 - x_{\text{m}}} \tag{4}$$

If $(T_1 - T_0)$ is less than ΔT_{th}, a negative flux is produced (3, 7), which inhibits the membrane distillation process.

When membrane distillation is run under very high concentration conditions, the behavior is very different from that for a dilute solution. Mass transfer decreases as the concentration of the feed increases, and this can lead to membrane distillation crystallization. If the solute is not easy to crystallize, the viscosity will increase until the flux decreases to zero. If the solute is easy to crystallize, then the flux will continue until supersaturation concentrations are reached, and crystals will then start to precipitate. These crystals can then be collected outside the module, and the membrane distillation continued (20).

A nonvolatile solute in the feed reduces the vapor pressure according to Raoult's law (4),

$$p = P^{\circ}(1 - x) \tag{5}$$

It also alters the fluid dynamics through effects on density and viscosity, and influences heat transfer through thermal conductivity and specific heat (17).

Aerated systems are often mass transfer limited. A number of studies have looked at deaerating the feed solutions and have been able to produce flux improvements (18). Deaerating the membrane decreases the molecular diffusion resistance which makes Knudsen flow dominant (1). This means that an improvement will not be evident when Knudsen flow is already dominant (19).

Heat Transfer

In direct contact membrane distillation, the evaporation and condensation surfaces are close to each other. As a consequence of this, a high conductive heat flux parallels the mass flux, and the effective temperature difference across the membrane is greatly reduced with respect to the bulk temperature difference (21).

Heat transfer occurs by heat conduction, Q_{c}, and latent heat, Q_{v}, across the membrane according to

$$Q_{\text{c}} = \frac{k_{\text{m}}}{\delta}(T_1 - T_0) \tag{6}$$

and

$$Q_v = N\lambda = C\frac{dP}{dT}\lambda(T_1 - T_0) \tag{7}$$

where k_m is the membrane thermal conductivity given by

$$k_m = \varepsilon k_g + (1 - \varepsilon)k_s \tag{8}$$

The membrane thermal conductivity is required in order to describe heat and mass transfer through the membrane as the membrane contains both polymer and vapor. The values of membrane thermal conductivity found by Eq. (8) agree with measured values within 10% (19).

Q_c is the heat conducted through the membrane from the hot to the cold side and has the effect of reducing the driving temperature difference. Q_c should be minimized as it impedes the process of membrane distillation. Johnsson et al. (22) stated that using PTFE membranes with high porosity can reduce the membrane thermal conductivity, therefore the heat lost by conduction constitutes only a relatively small part of the total rate of heat transfer.

Q_c and Q_v can be combined to obtain an effective heat transfer coefficient, H, to enable the heat transfer occurring in membrane distillation to be defined as

$$Q = \left[C\frac{dP}{dT}\lambda + \frac{k_m}{\delta}\right](T_1 - T_0) = H(T_1 - T_0) \tag{9}$$

As the mean temperature of the module increases, the effective heat transfer coefficient also increases (4, 7) due to dP and λ increasing with temperature. The heat transfer in the hot boundary layer can also be found by relating dimensionless groups (10).

Schofield et al. (18) utilized an iterative procedure to solve mass and energy balances in the direction of the feed to calculate the permeate flux. This was found to accurately predict the fluxes obtained with direct contact membrane distillation.

Ugrozov et al. (23) also combined heat and mass transfer to develop equations that could accurately model a membrane distillation module. The main conclusion from their work was that the mean temperature across the membrane decreases with increasing distance along the module. This means that the production rate of membrane distillation will not increase directly with module length.

The majority of models assume a linear temperature gradient over the membrane, but Gryta and Tomaszewska (24) showed that this would lead to inaccuracies in calculating the permeate flux. They applied a temperature gradient that would better match the actual temperatures inside the membrane and obtained more accurate data from their modeling.

BOUNDARY LAYERS

When modeling membrane distillation, consideration needs to be made of the influence of boundary layers on the permeate flux. The modeling of boundary layers has been a major activity in fluid mechanics. Most studies have been concerned with flow through pipes and over objects, such as aircraft wings. In membrane processes, however, the flow is always enclosed, with transport occurring through one or two porous walls.

A simple boundary layer is a layer of fluid where the velocity approaches zero at a solid surface, the no-slip condition, and approaches the bulk velocity at the edge of the boundary layer. The velocity gradients are large near the surface and smaller in the direction of flow. Heat and mass transfer rates can be predicted because the major resistances lie in the thermal and concentration boundary layers. The velocity, thermal, and concentration boundary layers can develop and exist simultaneously.

Belfort (25) pointed out that as mass transport to a membrane surface is dependent on the flow above the surface, understanding the fluid mechanics is a prerequisite to studying mass transport problems such as concentration polarization and fouling. This reasoning can be extended to heat transfer and temperature polarization.

Dubinski et al. (26) decided that understanding the hydrodynamic conditions in their equipment would aid in the optimum construction of a module for membrane distillation. Their design was a circular disk with the permeate outlet in the center. The fluid flows from one side to the other as shown in Fig. 1. Their work concentrated on predicting where the fluid would flow using

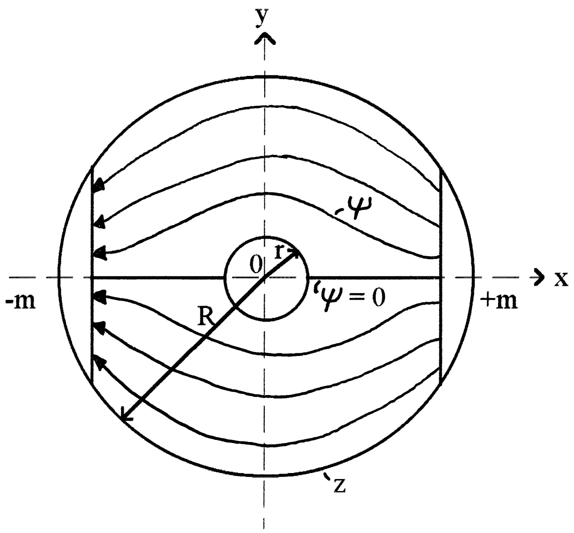

FIG. 1 Diagram of flow over a circular plate for membrane distillation (26).

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Laplace's equation (Eq. 10) and not the effect of the module geometry on permeate flux.

$$\Delta\psi = 0 \tag{10}$$

ψ is the flow through a cross section between ψ equal to 0 and any local value of ψ, i.e., it is a stream function.

The effects of boundary layers cannot be ignored. Sudoh et al. (28) did a comparison by using an analogy analysis to model the boundary layers and found that the fluxes obtained when ignoring boundary layers were larger than those actually obtained. When the concentration of the lithium bromide used in their study was less than 5%, the effect of the concentration boundary layer was considered negligible. However, as is to be expected, the thermal boundary layer was important over the complete concentration and temperature range used.

Burgoyne et al. (29, 30) utilized boundary layer analysis to predict the development of the thermal boundary layers in a flat-plate module. A set of integral equations of the form given in Eq. (11) were developed and solved as a set of first-order differential equations.

$$C_p\rho \frac{d}{dx}\int_{y=0}^{y=y_1} v(y)[T(y) - T(y_1)]dy = k\left.\frac{dT}{dy}\right|_{y=y_1} - k\left.\frac{dT}{dy}\right|_{y=0} \tag{11}$$

The work showed that increasing the channel length decreased the permeate flux produced. This was due to the temperature of the feed and permeate decreasing and increasing, respectively, reducing the driving force.

Agashichev and Sivakov (31) used mass and energy balances to define the hydrodynamic, temperature and concentration boundary layers. All the boundary layers were related by fixed ratios to the velocity boundary layer, assuming a flat membrane, and incompressible, continuous laminar flow. The velocity profile used was

$$v_\eta = \frac{v_z}{v} = \sin\left(\frac{\pi}{2}\eta\right) \tag{12}$$

in which η is 1. The corresponding temperature and concentration boundary layers were then

$$\frac{t_\theta - t_{1m}}{t_1 - t_{1m}} = \sin\left(\frac{\pi}{2}\theta\right) \tag{13}$$

where

$$\theta = \eta\left(\frac{C_p\mu}{\lambda}\right)^{1/2} \tag{14}$$

and

$$\frac{x_{\beta} - x_1}{x_{1\mathrm{m}} - x_1} = e^{\alpha\beta} \tag{15}$$

where

$$\beta = \left(\frac{\mu}{\rho D}\right)^{1/3} \left(\frac{C_{\mathrm{p}}\mu}{\lambda}\right)^{-1/2} \tag{16}$$

The model formed determines the physical properties of the fluid, and the temperature and concentration in both the feed and permeate channels. They stated that the velocity boundary layer is larger than the temperature boundary layer, which is in turn larger than the concentration boundary layer, as shown in Fig. 2. Their model took into account the energy interdependence between the flow in the feed and permeate channels.

Gryta and Tomaszewska (24) developed a much easier model based on differential element analysis to follow the temperatures and concentrations along a module. Their experimental work utilized a capillary membrane, but the analysis could be used for most module configurations. The equations were formed to calculate the interfacial conditions using heat transfer correlations. The model was shown to accurately predict the permeate fluxes obtained when using the most suitable heat transfer correlation. One important result from their work was that increasing the membrane area decreased the permeate flux obtained because of the reduction in temperature difference over the membrane. This agrees with the work of Burgoyne et al. (29, 30).

One method of reducing these effects is to introduce turbulence. Martínez-Díez et al. (27) used netlike spacers to introduce turbulence at the membrane surface. It was found that coarse spacers are better than fine spacers and that spacers can cause the transport to be controlled by mass transfer effects.

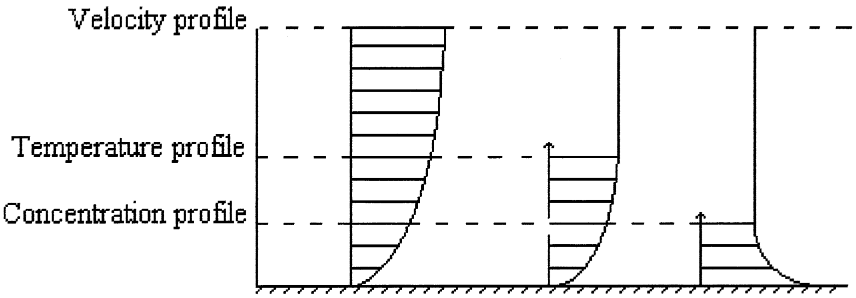

FIG. 2 Relation of velocity, temperature, and concentration boundary layer profiles (31).

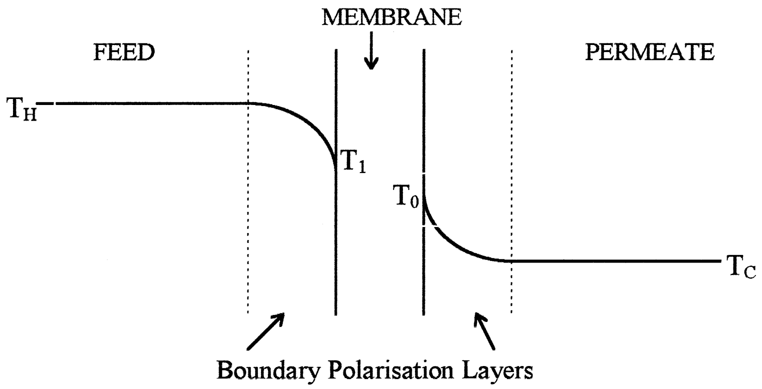

FIG. 3 Relation of the interfacial temperatures to the bulk temperatures.

TEMPERATURE POLARIZATION

Temperature polarization is a loss of driving force brought about by thermal gradients in the fluids bounding the membrane (7) and is a major problem for membrane distillation. Figure 3 shows the relation of the interfacial and bulk temperatures due to temperature polarization. Temperature polarization becomes more significant at higher temperatures (19). It is possible to characterize temperature polarization by a heat transfer analysis.

The membrane distillation system can be characterized as a system of heat resistances as shown in Fig. 4, so that the overall heat transfer can be expressed as

$$Q = h_1(T_H - T_1) = h_0(T_0 - T_c) = H(T_1 - T_0) \tag{17}$$

As the driving force is the temperature difference between the interfacial temperatures, T_1 and T_0, rearranging Eq. (17) and defining the temperature polar-

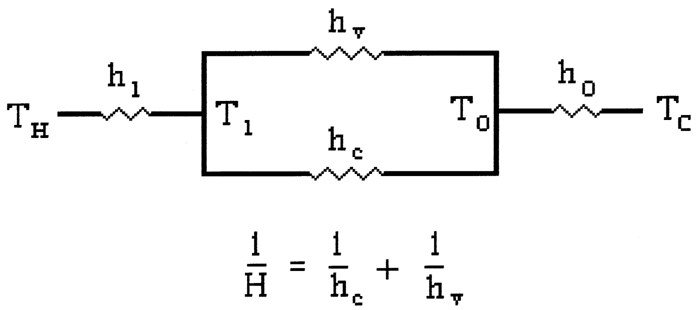

FIG. 4 Membrane distillation heat resistances.

ization coefficient (TPC) as

$$\mathrm{TPC} = \frac{1}{1 + \frac{H}{h_1} + \frac{H}{h_0}} \tag{18}$$

gives (4, 7)

$$(T_1 - T_0) = \mathrm{TPC}(T_{\mathrm{H}} - T_{\mathrm{C}}) \tag{19}$$

Ideally, TPC should equal 1, but usually it is closer to 0. If the TPC is less than 0.2, then the process is heat transfer limited, and would indicate a poor module design. If the TPC is greater than 0.6, then the process is mass transfer limited due to a low membrane permeability (18). Chmielewski and Zakrzewska-Trznadel (32) used this method to compare experimental values of TPC with theoretical values and found them to agree quite well. They stated that as the temperature increased, the TPC decreased. For example, with their flat-sheet module, increasing the temperature from 298 K to 323 K decreased the TPC from 0.75 to 0.59. The TPC is the fraction of external applied thermal driving force that contributes to the mass transfer (17).

Schofield et al. (7) used the TPC as a tool in designing membrane distillation systems. They looked at various module configurations and calculated the TPC. The closer the TPC was to 1.0, the better the heat transfer in the module, and therefore more suitable for membrane distillation (Table 1). They found

TABLE 1
Temperature Polarization Coefficient (TPC) for Various Module Configurations (6)

Membrane geometry	Flow characteristics	Nu	d (mm)	h (W/m^2·K)	TPC
1 mm i.d. tube	Re = 5,000	29	1.0	19,000	0.9
	Re = 3,000	20	1.0	13,000	0.85
	Re = 1,000	4.4	1.0	2,900	0.54
0.3 mm i.d. tube	Re = 300	4.4	0.3	9,700	0.8
0.6 o.d. tube bundle	Close-packed laminar	≈5	0.9	3,700	0.6
Channel 0.5 m long	v = 2 m/s	970	500	1,300	0.51
Conducting film:					
2 mm thick	Laminar	5.4	8	450	0.15
0.5 mm thick	Laminar	5.4	2	1,800	0.4
0.1 mm thick	Laminar	5.4	0.4	8,900	0.7
Stirred cell	Re = 8,000	54	50	710	0.2
	Re = 32,000	120	50	1,600	0.4

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that the three best module configurations were

1. 1 mm tube, Re ~ 5000 in the tube
2. 0.3 mm tube, Re ~ 300 in the tube
3. 0.1 mm film with laminar flow

Temperature polarization can be reduced by increasing the velocities of the liquids, using turbulence promoters, and decreasing the height of liquid channels (19).

Usually there is no variation of TPC with temperature difference or mean temperature (10), it only strongly depends on membrane characteristics, fluid dynamics, and feed concentration (3). However, Ortiz de Zárate et al. (33) found that the TPC and flux increased with stirring rate. Stirring the fluids either side of the membrane in test cells has been shown to increase flux due to the effect of fluid shear on the temperature polarization layer. Stirring increases the film heat transfer coefficient, which therefore decreases temperature polarization. Martínez-Díez and Vázquez-González (34) expanded this concept to an actual polarization coefficient, f (Eq. 20). This takes account of both temperature and concentration polarization and is useful for real solutions.

$$f = \frac{P_1 - P_2}{P_H - P_C} \tag{20}$$

The interfacial temperatures cannot be measured directly, but they can be found from a knowledge of the heat transfer coefficients throughout the membrane distillation module and bulk fluid temperatures (11).

Gryta and Tomaszewska (24) looked at the correlations available for determining heat transfer coefficients. They specifically looked at correlations for laminar flow as this is the area for the greatest error as any turbulence etc. could alter the heat transferred. It was shown that up to 94% modeling accuracy could be achieved by choosing the most suitable heat transfer correlation. The heat transfer effects were studied using a heat exchanger with a similar configuration to the membrane modules used.

In summary, temperature polarization is a major problem for membrane distillation as it is a thermally driven process and can severely reduce the fluid temperatures.

MEMBRANE TYPES

Various membranes are used for membrane distillation. The most popular include PTFE (polytetrafluoroethylene), PVDF (polyvinyldifluoride), and PP (polypropylene). The membrane is used only as a physical barrier between the feed and permeate streams and is not directly involved in the separation. The hydrophobic nature of these membranes prevents bulk liquid transport of the

TABLE 2
Membranes Used for Research of Membrane Distillation

Membrane type	Origin	Polymer	Pore size (μm)	Thickness (μm)	Porosity (%)	Reference
Flat	Enka	PP	0.1	100	75	7
Flat	Enka	PP	0.2	140	75	7, 18
Flat	Gelman	PTFE	1.0	178	80	4, 10, 17, 35, 43
Flat	Gelman	PTFE	0.45	178	80	27, 35, 44
Flat	Gelman	PTFE	0.2	178	80	9, 10, 17, 45
Flat	Gelman	PTFE	0.2	60	60	21, 34, 42, 44, 46
Flat	Taflen	PTFE	0.8	60	50	4, 47
Flat	Vladipor		0.25	120	70	48
Flat	Millipore	PVDF	0.45	110	75	3, 7, 18, 44
Flat	Millipore	PVDF	0.11	140	75	14
Flat	Millipore	PTFE	0.2	130	70	15, 32
Flat	Millipore	PTFE	0.5	175	85	9, 45
Flat	Teknokrama	PTFE	0.2		80	8
Flat	Teknokrama	PTFE	0.5		80	8
Flat	Teknokrama	PTFE	1.0		80	8
Flat		PTFE	0.1	178	80	15, 33, 50
Flat		PTFE	0.2	178	80	15, 33, 50
Flat		PTFE	0.45	178	80	15, 33, 50
Flat		PTFE	0.2	178	70	15, 50
Flat		PTFE	0.2	80	75	15, 28, 50
Flat		PVDF	0.22	80	75	33
Flat		PTFE	0.3	80		51
Flat		Silicone	Dense	250	Nonporous	37
Flat		Fluorocarbon	Dense	130	Nonporous	38
Capillary	Enka	PP	0.43	150	70	20, 52
Capillary	Enka	PP	0.2		70	2
Capillary	Accurel	PP	0.5	150	66	4
Capillary	Accurel	PP	0.6	400	74	4
Capillary	Accurel	PP		155	75	49
Capillary		PP	0.2	150		19
Capillary		PP	0.25	800	75	24
Capillary		PP	0.5	300	70	24
Capillary		PVDF	0.03	100	81	45
Capillary		PP	0.45	100	70	3

liquid phase across the membrane (2). The choice of membrane is a compromise between high flux (thin membrane) and low thermal conductivity (thick membrane) (35). Details of membranes used in studies of membrane distillation are given in Table 2.

According to the definition of the process (36), membrane distillation is only possible with hydrophobic membranes, but Ohta et al. (37, 38) conducted an experimental study using hydrophilic membranes but still called the process membrane distillation. The dense, hydrophilic membranes used were silicone and a fluorocarbon composite. Both were applied to seawater desalination. Fluxes of a similar magnitude to those obtained with hydrophobic membranes were achieved (37, 38). No details were given on why the authors thought that the process used could be defined as membrane distillation or how the transport mechanism could have taken place. The phenomenon observed can therefore only be acknowledged as closely related to membrane distillation.

Membranes with a narrow pore size distribution and high porosity [$> 50\%$ (7)] are best for membrane distillation (2). Porosity can be induced by mechanical stretching and/or thermal phase separation techniques. Also, due to temperature polarization, the thermal conductivity of the polymer is a controlling parameter for the process. Thermal conductivities of commercial membranes lie between 0.04 and 0.06 W/m·K, increasing with decreasing porosity (7).

Membrane distillation is usually carried out with commercial microfiltration membranes, but a number of studies have produced their own specific membranes in order to improve the flux and separation. Ortiz de Zárate et al. (39) looked at using phase polymerization with PVDF as the polymer and dimethylacetamide (DMA) and dimethylformamide (DMF) as the solvents. They observed that pore diameters and porosity increased as the PVDF content fell, though there was no improvement in flux as compared to commercial membranes. Another study by Tomaszewska (40) also used PVDF polymer and DMA and DMF solvents, but introduced a lithium chloride additive to the casting solution. This had the effect of increasing porosity and pore size. In the membrane distillation of a 1–2% sodium chloride solution, the permeate flux increased as the lithium chloride content was increased, as shown in Fig. 5. It was therefore concluded that the characteristics and properties of the membrane were affected by the composition of the casting solution and by the tem-

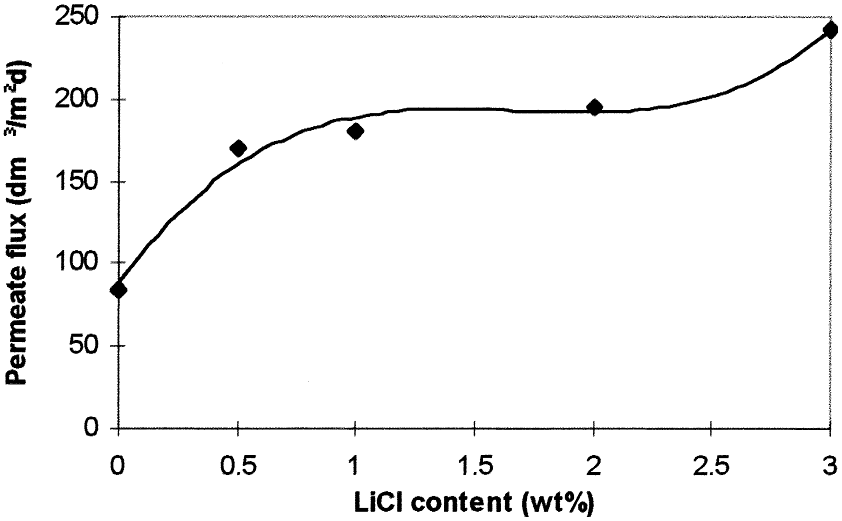

FIG. 5 Effect of lithium chloride in the membrane casting solution on permeate flux (40).

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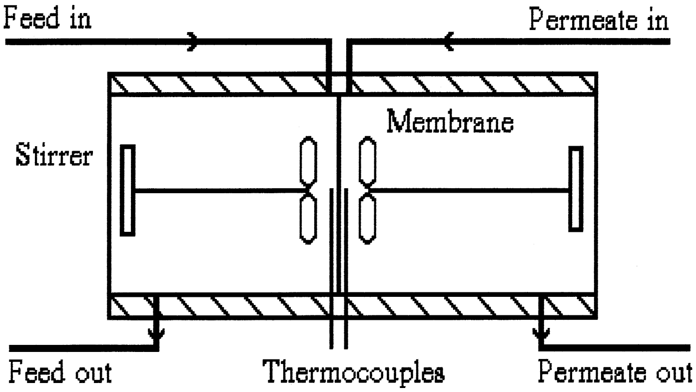

FIG. 6 A Lewis cell.

perature of the coagulation bath used in the phase polymerization. Elkina et al. (41) modified normal hydrophobic membranes by forming a hydrophilic film on the surface. This introduced a diffusion step to the process and enhanced selectivity with volatile compounds while maintaining dry pores in the hydrophobic part of the membrane.

MODULE CONFIGURATIONS

The majority of the work carried out on direct contact membrane distillation using flat-plate modules has involved the use of a Lewis test cell (Fig. 6) (8, 10, 42, 45, 46, 50) which has stirring capabilities. Schofield et al. used the thin channel device shown in Fig. 7. Other flat modules of the type shown in Fig. 8 were used by Ortiz de Zárate et al. (15, 33), Ohta et al. (37, 38), and Sarti et al. (45). The other modules used were of the hollow fiber type (2–5, 20, 49, 52, 53) shown in Fig. 9. In Fig. 9 the feed is shown to pass through the shell, but the arrangement could be switched to allow the feed to flow through the

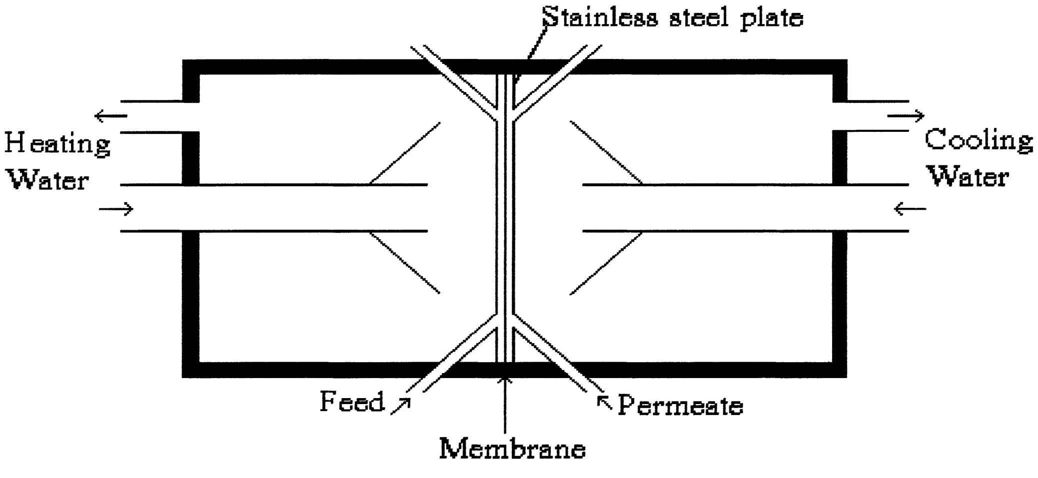

FIG. 7 A thin channel module.

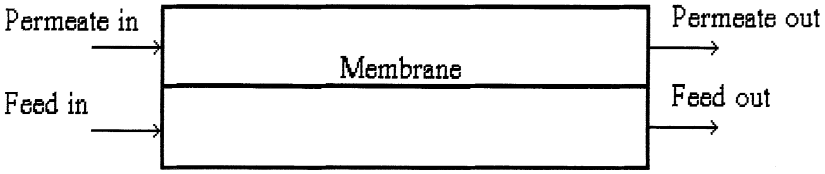

FIG. 8 Basic flat plate direct contact module.

fibers. Having the fibers twisted or braided instead of straight produces more uniform flow in the module and allows for any thermal expansion of the membranes (49).

A problem with hollow fiber modules is that as the feed solution travels along the module, the process of membrane distillation will cool the feed, thereby reducing the temperature driving force. Work has been carried out in order to overcome this by using countercurrent flow (54), and Schneider et al. (49) found that in turbulent flow a single capillary in a tube suffered less than a 3°C drop in temperature from inlet to outlet. With flat-plate modules it is accepted that having the warm feed on the bottom of the module is best, as free convection of heat enhances the heat transfer rate (47).

Overall, the Lewis cell is only suitable for laboratory scale work, but the principles involved can be used in developing larger modules. At the current time the other module types all have similar capabilities of performing membrane distillation.

APPLICATIONS

The main applications of membrane distillation are for the purification and reclamation of water. Desalination has been the main application, with various studies looking at the effectiveness of membrane distillation both at the laboratory and pilot scale (11, 37, 38, 45, 56). Hogan et al. (5) built a pilot plant for desalination by membrane distillation, and incorporated energy savings by collecting power from solar panels for use in heating the feed. Schneider et al. (49) stated that at present, membrane distillation will not be able to

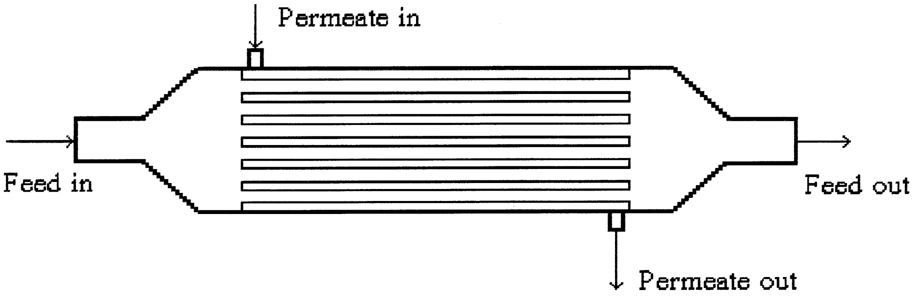

FIG. 9 A hollow fiber module.

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compete with the current large scale multieffect evaporation units used for desalination. Membrane distillation, however, would be competitive if waste heat generated from an industrial plant is used (49). Lawson and Lloyd (55) compared membrane distillation with reverse osmosis for desalination, and although their current work is limited to the lab scale, they reported permeate fluxes two to three times higher than those attainable with reverse osmosis. Also, salt rejection can be as low as 90% with reverse osmosis, whereas the membrane distillation rejection was approximately 100%.

Production of ultrapure water is an extension of desalination (7, 52) as the permeate product is very pure and is suitable for use in the medical and pharmaceutical sectors.

Utilizing membrane distillation to manage effluent streams from processes is another viable option. The process has been used to separate and recover chemicals, such as nonvolatile salts (52, 56), taurine (53), sodium chloride (53), dyes (3), and volatile solutes such as acids (57–59). Fujii et al. (60) looked at the removal of low concentration organics from water. These types of separations are possible, but only at low concentrations because of membrane wetting. For example, a 5% ethanol/water solution was separated by membrane distillation with a flux of 0.7 $kg/m^2 \cdot h$. Tomaszewska et al. (40, 57–59) carried out a large amount of work on acids. A recent finding (59) is that the variation of the partial vapor pressure of HCl with concentration influences the flux of HCl. They also confirmed that when a salt is added to a mixture of volatile components, the salt raises the partial pressure of one component while lowering that of another, allowing better separation.

A new application for membrane distillation is removing water from azeotropes. Membrane distillation, like pervaporation, would remove the need for an entrainer which is required in conventional azeotropic distillation. Udriot et al. (61) looked at separating the propanoic acid/water azeotrope. The low diffusivity of the heavy propanoic acid molecules, relative to water, reduces its apparent volatility compared to its actual value according to vapor/liquid equilibrium. The effect of diffusion on the vapor/liquid equilibrium is shown in Fig. 10. Complete azeotropic distillation is not possible using membrane distillation, but mixtures containing large amounts of water can be separated above and below the azeotropic point.

Another application of membrane distillation is for concentration of the feed. The majority of concentration applications are for the food industry, where membrane distillation has been used to concentrate orange juice, milk, sugar, and gelatine. Currently, multistage vacuum evaporation is the most widely used technology for these separation processes, but the product loses flavor and color, and the concentrate can acquire a cooked taste (14).

A specialist application is for the nuclear industry. Chmielewski and Zakrewska-Trznadel (32) successfully enriched deuterium and oxygen-18 from water and achieved higher separations than obtained with normal fractionation.

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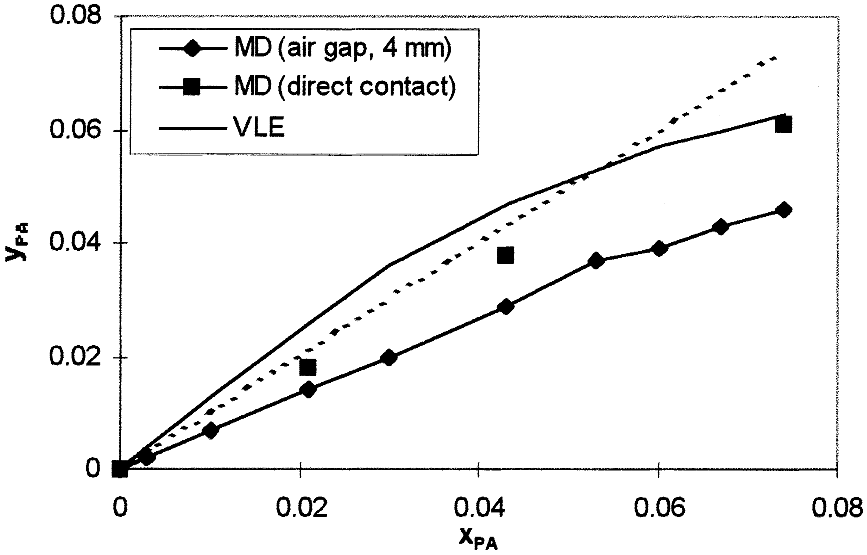

FIG. 10 Vapor–liquid equilibrium diagram for membrane distillation of propanoic acid/water mixtures (61).

Membrane fouling can be a severe problem with food concentrations. An ultrafiltration pretreatment unit could be used for heavy fouling feeds to remove the larger particles that could increase the viscosity of the stream through the membrane distillation unit (14). An example of this is given in Fig. 11 for orange juice, and shows that the ultrafiltration results in the flux remaining almost constant in the membrane distillation unit. The initial flux

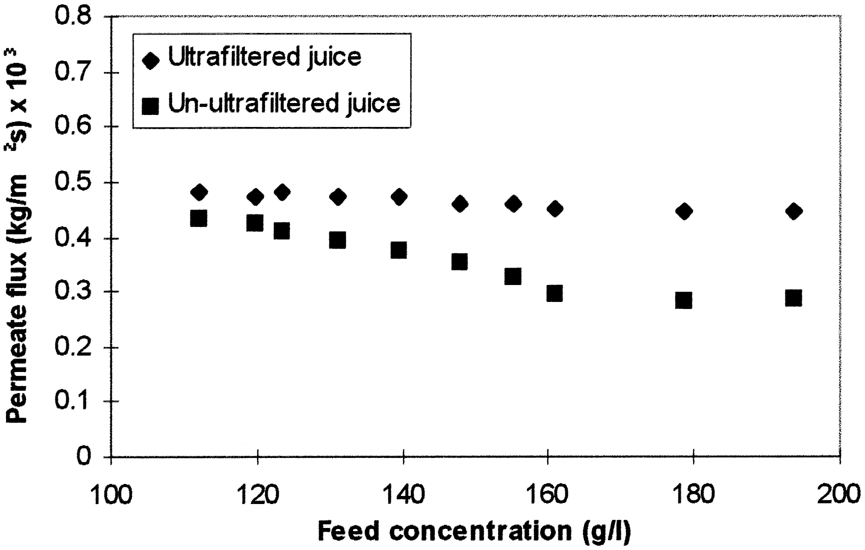

FIG. 11 Permeate mass flux vs feed concentration, with and without ultrafiltration pretreatment (14).

was restored after cleaning the membrane with either sodium hydroxide or hydrochloric acid.

EXPERIMENTAL WORK

Various experiments have been carried out in order to investigate the effectiveness of membrane distillation. These can be split into two groups. The first group uses either pure water or a sodium chloride solution as the feed, and is concerned with the basic process trends of membrane distillation. The second group uses different types of feeds and is concerned with specific applications for the process.

Group one type experiments are the most extensive, and have been investigated by Schofield et al. (5, 11, 18), Drioli et al. (20, 52), Ortiz de Zárate et al. (15, 33, 42, 50), as well as many others (37, 38, 45, 46). A review of the experimental conditions used for these experiments can be found in Table 3.

These experiments have allowed the investigation of the effect of the operating conditions on membrane distillation, and a number of trends have been

TABLE 3
Experimental Conditions Used for Basic Research of Membrane Distillation

Feed	Initial concentration (M)	Feed temperature (°C)	Permeate temperature (°C)	Stirring rate (rpm)	Temperature difference	Maximum flux ($kg/m^2 \cdot h$)	References
Water	n/a	20–60	20–60	200	0–10	7	10
Water	n/a	25–90		n/a	10–55	40	18
Water	n/a	45–50	40–45	0–360	5		33
Water	n/a	40–50	10–20	0–350	30	72	50
Water	n/a			200	5–20	45	35
Water	n/a	65–75	25–35	n/a	0–30	7.2	46
Water	n/a	30–70	20–60	150–350	10	14.4	8
Water	n/a	50	20	n/a	30	6.2	32
Water	n/a	20–50	10–40	n/a	10	0.12	27, 34, 43, 44
Water	0–5			50–350	0, 6	7.2	45
NaCl	0–5	20–60	20–60	200	0–10	9	10
NaCl	0–2.5	61, 71, 81	21	n/a	41, 51, 61	60	11
NaCl	0–5.3	30			10–15	5	5
NaCl	0.05–0.5	50		n/a	5, 9.5	1.7	20, 52
NaCl	0.1–0.3	40–50	10–20	0–350	30	16 mol/s	15, 50
NaCl	0–4			0–300	5–30	11.8	17
NaCl		40–60	20–40	n/a		4	37, 38
NaCl	0–0.9	40–70	20–50	n/a	0–20	0.72	42
NaCl	0–5			50–350	0, 6	6.5	45
NaCl	1–2 wt%	50, 60	20	n/a	30, 40		47
NaCl	0.55–1.67	20–50	10–40	n/a	10	0.10	34
NaCl	0.5	65–75	25–35	n/a	0–30	5.5	46
NaCl	0–30 wt%	100	42	n/a	30–60	9	49

found to exist. The trends can be summarized in the following way:

1. Flux increases with increasing feed temperature
2. Flux increases with increasing temperature difference across the membrane
3. Flux increases with increasing fluid flow over the membrane surface
4. Flux decreases with increasing feed concentration

Other trends found show that dearation increases flux by roughly 50% (11), increasing the stirring rate produces higher fluxes (50), and flux increases with decreasing membrane thickness (54). Also, as the hot and cold side heat transfer coefficients increase, flux increases and, as the membrane area increases, flux decreases due to closer temperature differences across the membrane (18). Drioli et al. (2) found that, generally, as the porosity of the membrane is increased, the flux increases.

A full list of specific feed solutions of the Group 2 experiments can be found in Table 4.

MEMBRANE WETTING

Membrane wetting is a serious problem in membrane distillation as the process relies on the pores of the membrane being filled only with vapor. This means that the membrane must be highly hydrophobic and the wetting power of the liquids low (16). Wetting causes membrane distillation to slow down, and, in the worst cases, distillation is halted due to a liquid flux occurring in the liquid-filled pores, in opposition to the vapor flux through the vapor-filled pores. Once the membrane is wetted, some pores will always contain liquid. Membrane wetting can occur due to various operating conditions. Repeated heating and cooling tends to wet the membrane (2), as do feeds containing organics (16), and membranes also become less hydrophobic with use (42). A main reason for membrane wetting is the pressure of liquid on the feed side of the membrane. For a given pore size there is a critical penetration pressure, above which the liquid will penetrate the membrane (2). This pressure is known as the liquid entry pressure of water (LEPW). The mechanism for membrane wetting is that water enters the larger pores of the membrane by breaking the surface tension at the interface between liquid and vapor on the membrane surface (2). The relationship between the pore size and pressure, given by the Kelvin law, is

$$P = 2\sigma \frac{\cos \phi}{r} \tag{21}$$

Schofield et al. (62) stated that pore sizes must be less than 0.5 μm to avoid wetting.

TABLE 4
Experimental Conditions Used for Specific Solutions

Feed	Initial concentration (M)	Feed temperature (°C)	Permeate temperature (°C)	Stirring rate (rpm)	Temperature difference	Maximum flux (kg/m^2·h)	References
Glucose	0–5.3	30			10–15	5	5
Glucose	10, 30 g/L	50		n/a	4.5, 8.5	5	2, 20, 52
Sucrose	0–5	61, 71, 81	21		41, 51, 61		11
LiBr	0–55 wt%	35–100	15	200–800	20–85	30	28
NH_3Cl	0.1–0.5	40–70	20–50	n/a	0–20	0.36	42
Sulfuric acid	12 wt%	60	30	n/a	30	12.5	47
Sulfuric acid	10 wt%	60	20	n/a	40	12.5	4
H_2SO_4/NaCl	10–60 wt%/1–20 wt%	40–80	20	n/a	20–60	8.3	24
HCl	0–30 wt%	60	20	n/a	40	10.5	4
HCl	0.5–7	60, 70	20	n/a	40, 50	8.3	59
Citric acid	12 wt%	55	25	n/a	30	10.4	3
$NiSO_4$	0.1	40, 50, 60	15	n/a	25–45	18.7	48
$NiCl_2 \cdot 6H_2O$	0.1	40, 50, 60	15	n/a	25–45	18	48
$Ni(NO_3)_2 \cdot 6H_2O$	0.1	40, 50, 60	15	n/a	25–45	20.1	48
Ethanol/water	0–7 wt%	30–60	0–20	n/a	10–40	4.7	21
Propanoic acid/water		60	30	n/a	30	22.1	56
Ethylene glycol		65	25–45	n/a	20–40		64
Gelatine	3–10 wt%	90	20	n/a	55–70	50	49
Orange juice	108–300 g/L	25–45	20	n/a	5–25	10.8	14

Organics have the effect of reducing the LEPW. Water and solutions of inorganics have high surface tensions ($> 72 \times 10^{-3}$ N/m), but when organics are present the surface tension falls rapidly. For example, Gostoli and Sarti (21) found that for mixtures of water and ethanol, the LEPW decreased linearly with ethanol concentration until the membrane was freely wetted at an ethanol concentration of 75 wt%. This is shown by Fig. 12.

Franken et al. (16) looked at methods for calculating the penetration surface tension. The penetration surface tension is the surface tension of the liquid on the verge of penetrating into the membrane. From comparing experimental results with theoretical calculations, it was discovered that contact angle measurements on homogeneous smooth materials were unsuitable for determining the possibility of membrane wetting occurring during membrane distillation. However, the wettability criteria determined by the penetrating drop method showed good agreement with the calculated values of the critical penetration surface tension.

Lawson et al. (63) also conducted research on membrane wetting, and they looked at the effect of membrane compaction and permeability on wetting. To compact the membrane in membrane distillation, the feed pressure can be increased, which increases the permeability and reduces the thermal energy requirement without affecting the temperature driving force. The results obtained showed that permeability could increase up to 11% with membrane compaction.

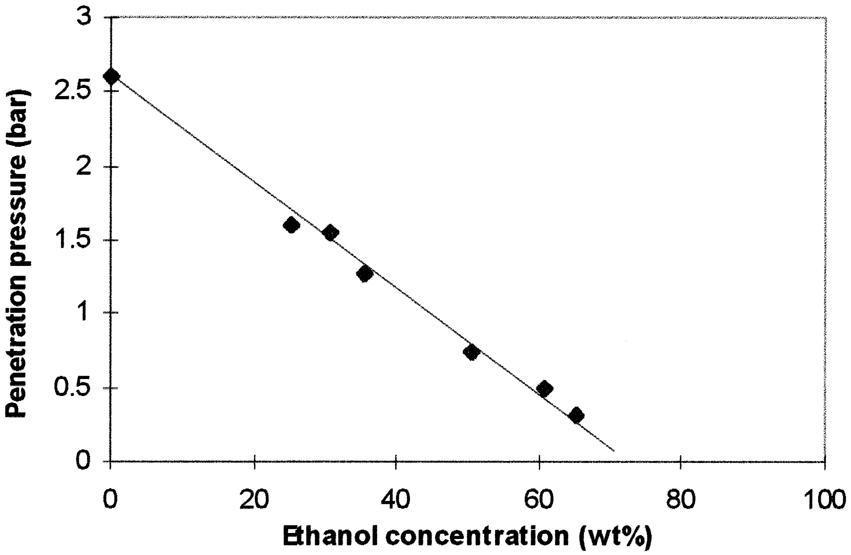

FIG. 12 Minimum entry pressure of ethanol–water mixtures in a PTFE membrane, pore size = 0.2 μm (21).

ENERGY ANALYSIS

Membrane distillation is energy intensive, and so heat economy is a most important issue (56). Energy recovery up to 75% can be achieved by heat exchange between the feed and warmed distillate in countercurrent flow (62).

Zotolarev et al. (48) carried out a preliminary economic analysis which showed that under some conditions membrane distillation was competitive with reverse osmosis for the production of distilled and potable water. The possibility of using solar and geothermal energy, or the existing low temperature gradients available in industry, is attractive, and it was used by Hogan et al. (5). Jonsson et al. (22) suggested that as membrane distillation requires the same heat addition for evaporation as a conventional one-stage evaporator, low-grade or waste heat should be used.

The advantage of membrane distillation is the ability to recover latent heat for reuse (5). Ohta et al. (37) looked at the amount of heat brought into a membrane distillation system and the heat actually used for the evaporation of the feed, and derived Eq. (22) for the thermal efficiency.

$$\text{Thermal efficiency } \% = \frac{\text{effective heat for evaporation}}{\text{heat input excluding emanation heat}} \tag{22}$$

The effective heat for evaporation is found from the permeate flux. The emanation heat is a loss that emanates from outside the module. An increase in flow rate does not increase the thermal efficiency very much. However, a higher feed temperature does result in a significant rise in the thermal efficiency. Efficiencies of up to 40% were calculated (38). An expression for efficiency similar to that given by Ohta et al. (37) was given by Calabrò et al. (3):

$$\mathrm{e} = \frac{NA\lambda}{mC_p(T_{in} - T_{out})} \tag{23}$$

Equation (23) shows that to increase the efficiency, the driving force must be increased. This can be accomplished at constant feed temperature by reducing the permeate temperature. The efficiency has been increased from 8 to 14% by this method. In terms of modules, larger membrane surface areas and lower flow rates increase the contact time in the module, which gives closer approach temperatures and therefore more recoverable heat (5).

Schofield et al. (18) suggested various methods to minimize the heat lost by conduction across the membrane. The first was to increase the thickness of the membrane, which lowers the heat lost by conduction, but also affects the latent heat, and therefore the overall effect is negligible. Another suggestion was to introduce an air gap which would act as an insulating layer, but this could reduce the flux due to the lengthened diffusion path. The final suggestion was dearation, which would increase the latent heat. Normal heat loss in a membrane distillation module is 20–40% of the heat input. Dearation could reduce this to less than 10%.

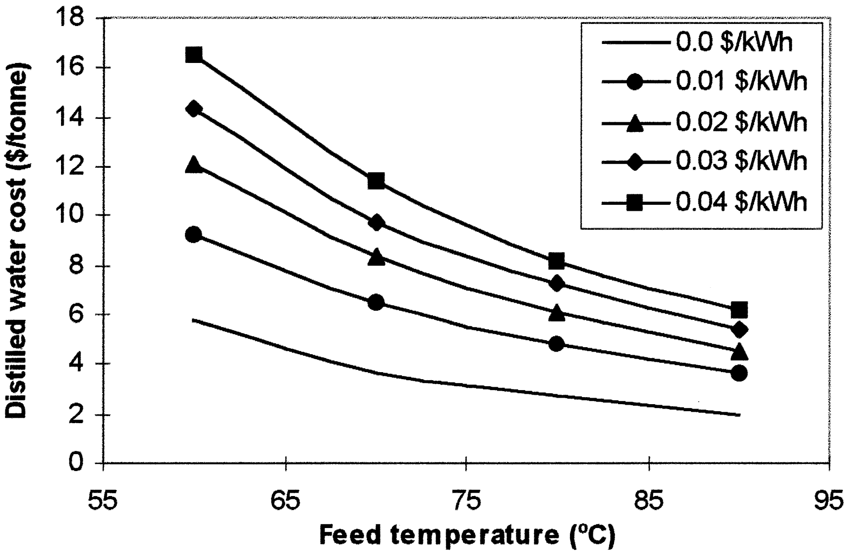

FIG. 13 Effect of energy cost and feed temperature on distilled water cost (5000 kg/h distilled water) (19).

Schofield et al. (19) found that the energy cost of membrane distillation falls with increasing temperature as shown in Fig. 13. For example, at 90°C the cost could be as low as $2/tonne, using the heat recovery system shown in Fig. 14.

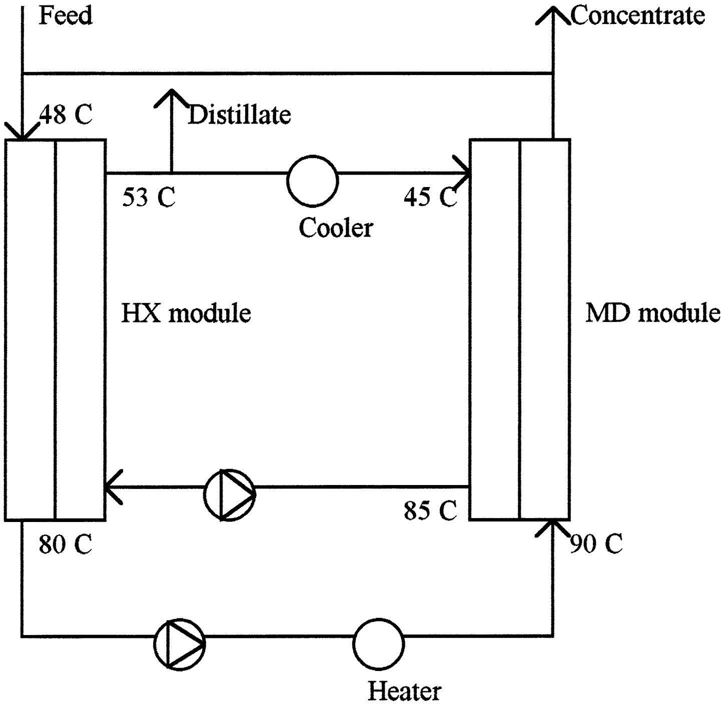

FIG. 14 Membrane distillation with energy recovery by heat exchanger (19).

CONCLUSIONS

A review of the research carried out on membrane distillation over the past 15 years has shown that the modeling of membrane distillation has been the main area of investigation. Studies have been concerned with the effect of mass transfer, heat transfer, and stirring rate. The initial feed concentration, temperature difference, membrane temperature, and boundary layers have all been shown to be factors that have to be accounted for in modeling membrane distillation. The most important effect that has to be considered with membrane distillation is temperature polarization, which results in a reduction in the driving temperature difference. Any model for membrane distillation must include the effect of temperature polarization.

Two types of module have been used for experimental work on membrane distillation. These are flat plate and hollow fiber modules. The majority of flat plate work has used a Lewis cell which has facilities for stirring the bulk fluids. Membranes used in the modules must be hydrophobic, with a porosity of 70–80% and a mean pore size of around 0.2 μm. Membrane distillation is suitable for a range of separations. These include processes where

1. The permeate is the product (e.g., desalination, water reclamation)
2. The concentrated feed is the product (e.g., foodstuffs)
3. Both feed and permeate are products (e.g., azeotropic separation)

Studies which involved experimental work were concerned with two different systems. The first involved using water and sodium chloride to study the basic trends of membrane distillation and to confirm the theoretical modeling. The second involved using specific feed solutions to test the suitability of membrane distillation for each separation.

A number of studies found that when dealing with organics the problem of membrane wetting could be significant, producing a liquid flux in opposition to the required permeate flux.

Membrane distillation has been found to be most suitable when the energy required is supplied by waste heat. This is because membrane distillation is energy intensive. Introducing a separate heat exchanger to recover heat has also been shown to improve the efficiency of the process.

NOMENCLATURE

A	membrane surface area (m^2)
C	membrane mass transfer coefficient ($kg/m^2 \cdot s \cdot Pa$)
C_p	specific heat ($J/kg \cdot K$)
D	diffusion coefficient (m^2/s)
e	distillation efficiency (—)

f	polarization coefficient (—)
h	heat transfer coefficient ($W/m^2 \cdot K$)
H	overall heat transfer coefficient ($W/m^2 \cdot K$)
k	thermal conductivity ($W/m \cdot K$)
M	molecular weight (kg/mol)
N	mass flux ($kg/m^2 \cdot s$)
P	pressure (Pa)
$P°$	vapour pressure (Pa)
Q	heat transfer rate ($W/m^2 \cdot K$)
r	membrane pore radius (m)
R	gas constant ($J/mol \cdot K$)
T	temperature (K)
v	velocity (m/s)
x	mole fraction (—)
y	physical distance (m)
z	physical distance (m)
β	dimensionless channel distance (—)
δ	membrane thickness (m)
ε	porosity (—)
ϕ	contact angle (°)
η	dimensionless channel distance (—)
λ	latent heat of vapourization (J/kg)
μ	viscosity ($Pa \cdot s$)
θ	dimensionless channel distance (—)
ρ	density (kg/m^3)
σ	surface tension (N/m)
ψ	stream function (—)

Subscripts

c	conduction
C	cold bulk
g	gas
H	hot bulk
m	membrane
s	solid
v	latent heat
l	interfacial hot side
0	interfacial cold side

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