MEMBRANE DISTILLATION

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Summary

Membrane distillation is a membrane separation process which may overcome some limitations of other membrane technologies. In particular, high solute concentrations can be reached, overcoming concentration polarization phenomena and ultrapure water can be produced as a permeate.

The process uses microporous hydrophobic membranes, impermeable to the transport of liquid water, while water vapor can be transported across them, having as driving force a vapor pressure difference at the two solutions membrane interfaces. Various polymeric hydrophobic membranes have been prepared with an appropriate microporosity of the order e.g. of 0.2 mm, of interest for this process.

The influence of the feed temperature, transmembrane temperature gradient, feed concentration, fluodynamic conditions, etc., have been studied theoretically and tested experimentally. Various membrane configurations and operation techniques have been also suggested for optimizing transmembrane fluxes and energy consumption.

Membrane distillation has shown interesting potential in water desalination, fruit juice concentrations and in other various industrial productions. Furthermore, by using membrane distillation in integrated operations it is possible to achieve higher concentration values and better overall performance of the processes.

1. Introduction

Membrane distillation is a relatively new membrane separation process which might overcome some limitations of the more traditional membrane technologies. In particular high solute concentrations can be reached and ultrapure water can be produced in a single step. The possibility of an industrial development of this technology is related to the growing commercial availability of membranes of potential interest.

When a microporous hydrophobic membrane separates two aqueous solutions at different temperatures, selective mass transfer across the membrane occurs: this process takes place at atmospheric pressure and at temperatures which may be much lower than the boiling point of the solutions. The hydrophobicity of the membrane prevents the transport of the liquid phase across the pores of the partition while water vapor can be transported across them from the warm side, condensing at the cold surface. The driving force is the vapor pressure difference at the two solution membranes interfaces.

Because the process can take place at normal pressure and low temperature, membrane distillation could be used to solve various wastewater problems, to separate and recover chemicals, and also to concentrate to high osmotic pressures aqueous solutions of

substances sensitive to high temperatures. The possibility of using solar, wave or geothermal energy, or existing low temperature gradients typically available in industrial processing plants is particularly attractive.

The fundamental simplicity of traditional distillation is compromised by various factors such as the need for complete removal of all noncondensable gases. The use of vacuum pumps, high pressure vessels, deaeration devices, etc. are required for removing the effects of the noncondensable gases, with a significant energy consumption. A number of distillation processes have been proposed with the aim of eliminating the need for creating a vacuum.

A relatively large number of evaporators based on the circulation or convection of water-saturated air, from an evaporation surface to a condenser, have been designed. An interesting result was found in the study of this kind of systems: the production of pure water per unit area and per unit time was inversely proportional to the gap existing between the evaporation and condensation surfaces. Obviously, it is impossible to reduce this gap without causing contamination of the distillate by the feed water, but this fact suggested the use of membrane systems.

Hydrophobic microporous membranes allow easy passage of water vapor, but completely block the flow of liquid water. Surface tension of the water prevents its passage through the pores of the hydrophobic material. If feed water is in contact with one of the surfaces of the membrane, the gap distance between the evaporation and condensation surfaces could be reduced to the thickness of the membrane, thus preventing contamination by the feed water.

Early work was presented by Findley (1967) and Findley et al. (1969) where transport in vapor phase across porous partitions was studied. The membrane materials used in these works were paper hot cup, glass fibers, aluminum foil and similar.

The efficiency of the process was related to the stability of membrane materials. In the 1980s new microporous hydrophobic membranes became commercially available and membrane desalination (MD) again attracted significant attention.

1.1 Principle of Membrane Distillation

Non-isothermal transport of fluids, especially water, through membranes has been studied since the beginning of the century . The first experiments in the field were based on dense membranes. The measured fluxes were small and the process of diffusive nature, was called “thermal osmosis” or “thermoosmosis”. The description of the phenomenon was generally carried out within the framework of the Thermodynamics of Irreversible Process.

In the 1960s (Findley 1967; Findley et al. 1969) a much bigger non-isothermal water transport was found in porous hydrophobic partitions, and called “membrane distillation”. The magnitude of fluxes were 1000 times greater than those found in dense membranes, at the same conditions.

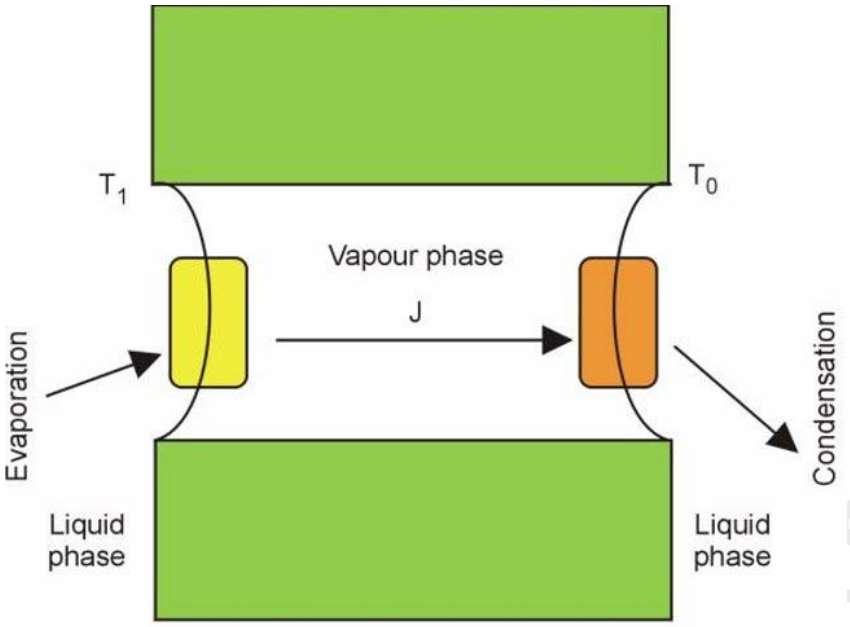

Figure 1. A schematic representation of the membrane distillation process: T_1, temperature at the hot side; T_0, temperature at the cold side; J, flux of the vapor phase.

Up to now membrane distillation has referred to the non-isothermal transport of water, via vapor phase, through the pores of a hydrophobic partition. The system consists of a porous hydrophobic membrane, separating two aqueous solutions of a non-volatile component maintained at different temperatures. Due to the liquid-rejecting properties of the membrane material, the liquid phase is prevented from penetrating the pores, as long as the pressure of liquid does not exceed the minimum entry pressure of the porous partition. Liquid-vapor interfaces are formed on both sides of the membrane pores and, due to the temperature difference, a vapor pressure difference is created between sides of each pore. Evaporation takes place at the warm interface and, after vapor is transported through the pores, condensation takes place at the cold interface. In this way a water flux occurs through the membrane in the direction from warm to cold. Obviously, for membrane distillation to proceed, it is essential that liquid water is excluded from the pores. In this sense, the role of the membranes is somewhat peculiar, since it acts as a physical support for the liquid-vapor interfaces. A schematic representation of the membrane distillation process is shown in Figure 1.

The origin of water transport in this kind of process is a difference in water chemical potential created by a vapor pressure difference. This is produced by a temperature difference between the two solutions facing the membrane, but could also be produced by different concentrations between the two aqueous solutions (Mengual 1993; Sheng et al. 1991). The process is called “osmotic distillation” if the system is kept in isothermal conditions and the difference in concentration is produced by non-volatile solutes.

The systems described above are the so-called “direct contact membrane distillation” and “osmotic membrane distillation” methods where the distance between the evaporation and condensation surfaces is reduced to the membrane thickness. But, following the description of the membrane transport, it is possible to imagine other

possible set ups, designed with the aim of increasing process efficiency. All these different configurations, for example "gas-gap membrane distillation" or "vacuum membrane distillation" will be considered in the next sections.

1.2 Future Development

Membrane distillation is an alternative to the traditional evaporative distillation systems used for desalination or water purification processes. On the other hand, membrane distillation can be compared with other membrane techniques, e.g. reverse osmosis.

Reverse osmosis is industrially used in desalination processes (nearly 30 per cent of desalinated water in the world is produced by this membrane technique), production of ultrapure water and food concentration (juices, sugars and milk, for example). RO is a pressure driven membrane process based on the solution-diffusion of the solvents, mainly water, across the membrane. Reverse osmosis efficiency is strongly affected by the osmotic pressure of the highly concentrate feed solutions (the osmotic pressure of seawater is about 25 bar) and by the concentration polarization phenomena that occurs on the pressurized membrane-solution interface. In addition, the membrane rejection is generally of the order of 98-99 per cent, and some salts can diffuse in the permeate.

In contrast, membrane distillation is a thermally driven membrane process where efficiency shows a slight decrease with increasing salt (or other inorganic solutes) concentration, because of a decrease in vapor pressure. In principle, MD can also produce ultrapure water from feeds at quite high concentrations where RO cannot practically operate.

In addition, the quality of the permeate (the separation efficiency) is virtually independent on the feed concentration. Mass transport in fact takes place in the vapor phase; non-volatile solutes are completely rejected by the membrane and only volatile solutes can be transported. This point has great relevance because it permits to design separation systems, with efficiencies near 100 per cent. But MD has some disadvantages. Compared to RO, the MD fluxes of permeate are usually lower and, being a thermally driven process there is necessarily a higher energy consumption. In addition, some membrane materials do not present a sufficiently high chemical resistance in the presence of salt, which implies a loss in the process efficiency, or they are still too expensive. These possible disadvantages could be overcome in different ways. In Table 1, a summary of the advantages and disadvantages of membrane distillation is shown.

Advantages	Disadvantages (⇒Possibilities To Overcome)
The continuous vapor permeation increases the evaporation in the warm liquid solution. Reduction of non-condensable species vapor phase. High concentration at low pressures and temperatures. Integration with other membrane	Energy consumption Use of solar, wave or geothermal energy. Use of existing temperature gradients available in industrial plants. Use of MD in the treatment of solutions discharged at high temperature. Fluxes are lower than in other membrane processes for industrial applications.

Advantages	Disadvantages (⇒Possibilities To Overcome)
operations. Reduction of osmotic limits.	Integration with other membrane processes or traditional technologies. Engineering improvement of the process.

Table 1. Advantages and disadvantages of membrane distillation operation.

The applications of membrane distillation, described in Section 5, are not restricted to the desalination field. In fact, the applications are only determined by the wettability of the membrane, which implies that mainly aqueous solutions containing inorganic or dilute aqueous solutions of organic compounds could be treated.

2. Fundamentals of Membrane Distillation

2.1 Process Description and Terminology

Different names exist to identify the mass transport, in vapor phase, across porous hydrophobic partitions, e.g.: "Gore-Tex® membrane distillation" (Gore 1982), "trans membrane distillation" or thermo-pervaporation. The most suitable, "membrane distillation", is however used by the majority of the authors and it cannot be confused with other membrane operations (Franken 1988). The main reason for choosing this name, regarding the nature of transport, is because membrane distillation is a process in which the membrane itself has no influence on the vapor-liquid equilibrium of the liquids to be separated. This is the main difference with pervaporation in which liquids diffuse across the membrane and the separation characteristics are determined by sorption into and diffusion through the membrane.

In this way, the characteristics of membrane distillation are:

- the membrane must be porous;
- the membrane must be not wettable by the liquids transported;
- only vapor will be transported across membrane matrix;
- there is not capillary condensation inside the pores of membrane;
- the vapor-liquid equilibrium must not be altered by the membrane;
- at least one side of the membrane must be in direct contact with the liquids to be processed;
- the driving force of this membrane operation is a partial pressure gradient in the vapor phase for each component.

Membranes can be characterized by the following performance parameters:

1. *Membrane (polymer) material*. The membrane material must be not wettable by process liquids. The thermal conductivity has a relatively high influence on the magnitude of membrane distillation fluxes (and osmotic distillation fluxes) in relation to temperature polarization effects. At present the membranes used are all made of polymer but this does not exclude other types of materials.
2. *Membrane thickness*. This parameter is important considering that the mechanical

strength and the membrane fluxes depend of it (Schofield 1987; Schofield et al. 1990a; Schofield et al. 1990b; Xu et al. 1994).

3. *Porosity of membranes*. Defined as the volume of pores divided by the total volume of membrane. The transmembrane fluxes are directly proportional to porosity (Peña 1993; Schofield 1987; Schofield et al. 1990b).
4. *Nominal pore size*. This parameter refers to pore sizes estimated from bubble-point tests, gas permeation experiments, etc.; it can be used for approximate calculations of the fluxes.
5. *Liquid-entry-pressure of water (LEPw)*, Also called wetting pressure, the LEPw is the pressure that must be applied to pure water before it penetrates into a non-wetted membrane and indicates the highest pressure difference that could be applied (Drioli et al. 1984; Kim et al. 1987; Pagliuca et al. 1983; 1987; Sarti et al. 1985; Udriot et al. 1990), through a membrane, in a membrane distillation operation. For a given pore size a critical penetration pressure P could be defined. Following the Kelvin law, this pressure is:

$$P_c = \frac{2\gamma \cos\theta}{r} \qquad (1)$$

where γ is the surface tension of the liquid, θ is the contact angle between liquid and membrane and r is the radius of the pore.

At present, a great number of membranes have been used in membrane distillation but no membrane has been made with the special aim to be used in this kind of process. The membranes are shown in Tables 2 and 3, in Section 3, as well as their principal performance characteristics.

Company	**Name**	**Material**	**Porosity (%)**	**Nominal pore size**	**LEP_W (bar)**	**Thickens (μm)**
Schleicher and Schuell	TE 35	PTFE Supported on polyester	75	0.2	4.0	120
	TE 36	PTFE Supported on polyester	75	0.45	1.8	120
Gore-Tex	-	PTFE	50	0.02	24	80
	-	PTFE	78	0.2	2.75	60
	-	PTFE	84	0.45	1.35	80
	-	PTFE	91	1.00	0.48	80
	-	PTFE	95	3.00	0.13	25
	-	PTFE	95	5.00	0.07	25
Millipore	AP-20	Borosilicated with acrylic glue	50	-	-	300
	GVHP	PVDF	75	0.22	-	125
	HVHP	PVDF	70	0.45	-	125
	LCWP	PTFE	68	5.0	2	125
	LSWP	PTFE	60	10.0	-	125
	FSLW	PTFE Supported on polyester	75	3.0	-	200

	FALP	PTFE Supported on polyester	75	1.0	-	145
	FHLP	PTFE Supported on polyester	75	0.5	-	175
	FHLP	PTFE	75	0.5	-	60
	FGLP	PTFE Supported on polyester	70	0.2	-	175
Gelman Instruments	TF200	PTFE Supported on polyester	80	0.20	2.70	175
	TF450	PTFE Supported on polyester	80	0.45	1.40	175
	TF 1000	PTFE Supported on Polyester	80	1.00	0.50	150
	TF5000	PTFE Supported on Polyester	80	5.00	0.14	150
	P5PQ	PTFE Supported on PTFE	80	0.5	2	178
	P5PL	PTFE Supported on PTFE	80	1.0	-	165
	P5PJ	PTFE Supported on PTFE	-	2.0	-	152
	P5P1	PTFE Supported on PTFE	-	3.0	-	152
	P4PH	PTFE	-	5.0	-	127
	R2PQ	PTFE	80	0.5	-	76
	R2PL	PTFE	80	1.0	-	76
	R2PJ	PTFE	-	2.0	-	25
	R2P1	PTFE	-	3.0	-	25

Table 2. Main performance characteristics of the flat membranes used for membrane distillation.

Company	**Name**	**Material**	**Number of capillaries**	**Nominal pore size (μm)**	**Inner diameter (mm)**
ENKA AG	LM-2P 1 2	Polypropylene	22	0.2	1.2
	LM-2P06	Polypropylene	85	0.2	0.6
	MD020TP2N	Polypropylene	3	0.2	5.5
	MD020CP2N	Polypropylene	40	0.2	1.8

Table 3. Main performance characteristics of tubular membranes used for membrane distillation.

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