Membrane Distillation Enhanced by an Asymmetric Electric Field

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A novel membrane distillation (MD) process enhanced by an asymmetric electric field was proposed. By applying an asymmetric electric field across a membrane with an increased field intensity in the direction of permeation, the permeation of water molecules through the membrane will be facilitated by the electric potential gradient, resulting in a higher flux than that achieved with the conventional process. The effectiveness of using asymmetric electric field to enhance mass transport was confirmed experimentally, and the effects of electric field parameters (including voltage, electrode distance, and electrode geometry) on water permeation were investigated. The novel process was tested for water desalination by MD and the results showed that both permeation flux and salt rejection were increased by the electric field. © 2014 American Institute of Chemical Engineers *AIChE J*, 60: 2307–2313, 2014
Keywords: electric field, electric potential gradient, polar dielectric, vacuum membrane distillation, desalination

Introduction

Membrane distillation (MD) is a water treatment process based on the differences in the volatility of the components to be separated, and the mass transport is driven by a partial vapor pressure gradient generated by a temperature difference across the membrane, or by vacuum or sweep gas on the permeate side. Membranes for water treatment by MD are microporous and hydrophobic, which prevent the feed liquid from passing through the membrane pores while only allowing volatile molecules (mainly water) to pass as a vaporous permeant.[1] Due to the very high rejection to nonvolatile dissolved components (e.g., salt ions or macromolecules), MD can be used to produce ultrapure water. It is not limited by osmotic pressure as in reverse osmosis, and thus it is capable of treating highly concentrated aqueous solutions or reverse osmosis concentrates, leading to an increased water recovery and lowered brine discharge.[2,3] MD is also well-suited for wastewater treatment,[4,5] food processing,[6,7] and biomedical applications.[8]

Although the potential uses of MD are well recognized, it is not yet commercially available for industrial applications, primarily due to its relatively low permeation flux compared to other membrane processes. To make MD commercially competitive for use in industry, high permeation fluxes must be achieved with moderate energy consumptions.[9–11] To improve the performance of MD, different approaches have been attempted, including manufacture of membranes with large pore sizes and high porosities,[12] modification of hollow fiber membrane geometries,[13,14] and the use of hydrophobic/hydrophilic composite membranes.[15] In addition, the mass transfer can also be enhanced through process engineering by optimization of operating conditions[16,17] and module and spacer designs[18,19] for proper management of fluid flow hydrodynamics.

To increase the driving force for permeation, MD is often operated at an elevated temperature. However, the feed temperature should not be too high from the standpoint of energy consumption. Certain applications dealing with the concentration of heat sensitive compounds cannot be operated at high temperatures either to minimize thermal degradation. This study seeks to develop a nonthermal approach to enhance the

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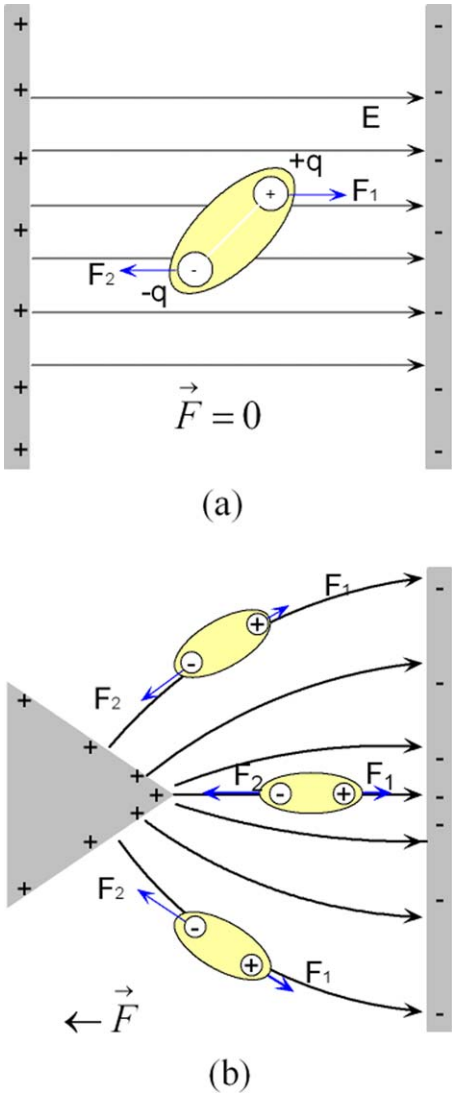

Figure 1. Electric force on water molecules in a uniform (a) and nonuniform (b) electric field. ($\vec{F}$: net force).

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driving force for mass transport across the membrane using an electric field with a nonuniform intensity to facilitate water permeation. The enhancement in water permeation in MD by the electric field was demonstrated, and the effects of parameters involving the electric field on the mass transport were investigated. The mechanism of mass transport facilitated by the electric gradient force was explored.

Theory

Water is a polar dielectric molecule having a permanent dipole moment. The dipoles of polar molecules are randomly oriented in the absence of electric field. However, when an electric field is present, these dipoles will be stretched to align with the electric field, with the positive pole facing the cathode and the negative pole toward the anode. This process is commonly known as the orientation polarization of dielectric molecules. In a uniform electric field, a polar molecule feels no net force as both dipoles experience equal forces in an opposite direction (see Figure 1a). However, in such a nonuniform electric field as those generated by two electrodes of different shapes (e.g., needle-plate), the electric field intensity is higher near the needle electrode. Each dipole will experience a different force, resulting in a net force on the molecule, that is, the electric gradient force (which is also called dielectrophoretic force). The direction of the electric gradient force depends on the electric field gradient, but not on the direction of the electric field. This force will motivate polar molecules to move to the region in the electric field with a higher intensity (see Figure 1b), which is known as dielectrophoresis.[20,21]

The phenomenon of directional migration of polar molecules in a nonuniform electric field has been utilized to increase the transport rate of polar molecules. For example, the evaporation rates of water and ethanol were reported to be enhanced in the presence of a wire-plate electric field.[22] Electrohydrodynamic drying of fruits and vegetables is another example. It has been reported that when food was placed on the plate electrode in a needle-plate electric field, the water molecules inside the food oriented along the external electric field and migrated along the direction of increasing field intensity thereby accelerating water evaporation from a higher concentration internal to a lower concentration external of the food.[23]

In a MD process for water treatment, the water vapor pressure gradient across the membrane determines the direction of water transport. Applying an electric gradient force in the same direction of permeation is expected to facilitate water transport through the membrane. Providing an electric field across the membrane with an increasing field intensity along the direction of permeation, water molecules will be "pulled" to the direction of increasing field intensity. As such, the driving force for water permeation will be a combination of the vapor pressure gradient and the electric potential gradient, and the permeation flux will be the sum of the flow induced by the pressure field and the flow induced by the electric field. Therefore, the mass transport of water will be accelerated in the nonuniform electric field. Conversely, the transport of salt molecules or macromolecules will be hardly affected by the electric field as they cannot evaporate and permeate through the membrane. In this study, the distribution of the nonuniform electric field along the membrane thickness was asymmetric, and the integration of molecular transport by migration in the electric field and the transmembrane diffusion in the pressure field is referred to as MD enhanced by asymmetric electric field.

Experimental

Materials

Hydrophobic microporous polytetrafluoroethylene (PTFE) flat sheet membrane (supported on a nonwoven fabric, thickness of the PTFE layer 20 μm, and effective pore size 0.22 μm) supplied by Taoyuan, China, was used for the experiments. For the purpose of proof of concept, two types of feed solutions were used in the MD studies: pure water (conductivity 14.6 μs/cm) was used as the feed to confirm the effectiveness of asymmetric electric field on water transport; and an aqueous sodium chloride solution (0.1 M) was used to demonstrate the application of the process for water desalination.

MD enhanced by asymmetric electric field

The MD performance of the novel process was evaluated using a bench-scale membrane test system under vacuum

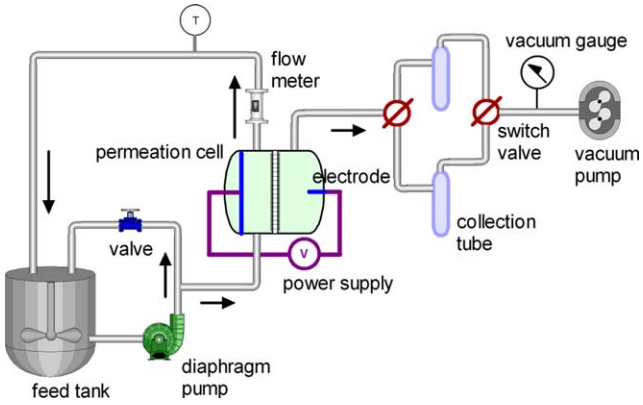

Figure 2. Schematic diagram of experimental setup for asymmetric electric field enhanced MD.

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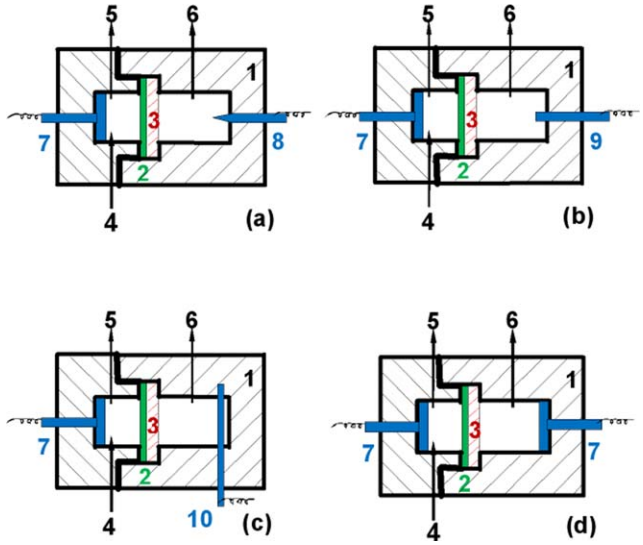

Figure 3. Membrane permeation cell with different configurations of electric field.

(a) Needle⊥plate electric field; (b) rod⊥plate electric field; (c) wire||plate electric field; and (d) plate||plate electric field. 1, permeation cell; 2, membrane; 3, porous plate; 4, feed; 5, retentate; 6, permeate (vacuum); 7, plate electrode; 8, needle electrode; 9, rod electrode; and 10, wire electrode. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

MD mode. The apparatus mainly consisted of three parts: asymmetric electric field membrane permeation cell, feed recycle, and permeate collection system (see Figure 2).

Design of the Asymmetric Electric Field Membrane Permeation Cell. The electric field permeation cell is the key part of this novel MD system. Such a permeation cell design has two functions: (1) it allows water molecules to permeate from the feed side to the permeate side as in a conventional MD process and (2) water permeation is facilitated by the electric gradient force due to an asymmetric electric field. The asymmetric characteristics of the electric field was controlled by a pair of electrodes installed on the two sides of the permeation cell. The charges are distributed over the surface of a conductor and tend to concentrate in areas of maximum curvature, resulting in a high field intensity in areas of sharp curvatures. Therefore, an electrode with a rounded needle tip was placed on the permeate side, whereas a flat disk of electrode (diameter 45 mm and thickness 50 mm) was placed on the feed side. Such a needle⊥plate electric field is a typical asymmetric field with the electric field intensity increasing from the disk electrode to the needle electrode and forming an electric potential gradient between the two electrodes (see Figure 3a). For comparison purposes, three other electrodes of different geometries were used on the permeate side in lieu of the needle electrode: a rod electrode (diameter 8 mm) placed perpendicular to the disk electrode (Figure 3b), a wire electrode (diameter 2 mm and length 45 mm) positioned parallel to the disk electrode (Figure 3c), and a disk electrode (diameter 45 mm) located parallel to the disk electrode on the feed side. All electrodes were made of stainless steel.

The permeation cell was constructed from nylon and consisted of two detachable parts. The PTFE membrane was mounted on a PTFE porous sintered plate embedded in the permeate chamber. The disk electrode was placed in the feed chamber, parallel to the membrane surface. The disk electrode size was the same as the effective membrane area for permeation (15.9 cm^2). The distance between the disk electrode and the membrane surface was fixed as 11 mm. The tip of the needle or rod electrode was positioned in the center of the permeate chamber, perpendicular to the membrane surface, whereas the wire electrode was parallel to the membrane surface (see Figure 3). The distance between the electrodes in the feed and the permeate side was adjustable. A DC power supply (0–20 kV, maximum 1 mA) manufactured by Tianjin Dongwen of China was used to generate an electric voltage between the feed and permeate sides. Caution was exercised during the experiments to ensure that all connections between the electrodes and the power supply were well isolated using a high voltage electrical insulation tape.

Feed Recycle and Permeate Collection. The feed stream was circulated by a diaphragm pump from a feed tank to the permeation cell, and the retentate was recycled back to the feed tank. The permeation cell was vertically oriented with retentate withdrawn from the top of the cell. The feed was allowed to flow parallel to the membrane surface at a flow rate of 60 L/h and at a temperature of 20°C.

The temperature of the permeate side was the same as the feed (20°C). The vapor pressure gradient across the membrane was generated by lowering the permeate pressure on the downstream side using a vacuum pump. The permeate pressure was maintained at 20 kPa absolute. The permeate vapor was condensed and collected in a cold trap immersed in liquid nitrogen.

The performance of the MD process was characterized by permeation flux and salt rejection. The permeation flux J was determined gravimetrically by measuring the quantity of the permeate sample Q collected over a time interval t with an effective membrane area A

$$J = \frac{Q}{A \cdot t} \tag{1}$$

To study the effect of the electric field on water permeation, two comparative experimental runs were carried out: one was without applying the electric field, and the other with an electric field at a voltage in the range of 0–4 kV. In the second series of experiments, the permeation flux after removing the electric field was measured again to check if

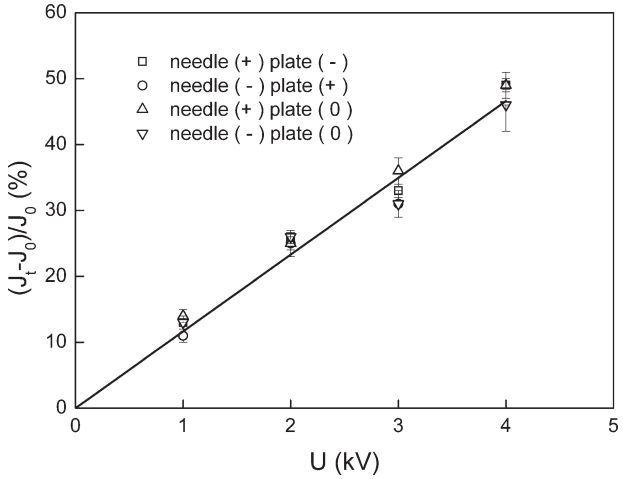

Figure 4. Effect of asymmetric electric field on the enhancement in water permeation.

(+): Anode, (−): cathode, (0): grounded; needle⊥plate electric field; and electrode distance: 23.5 mm. Feed and permeate temperature: 20°C; feed flow rate: 60 L/h; permeate pressure: 20 kPa absolute. J_0: 0.06 kg/(m^2 h). Solid line represents the calculated value.

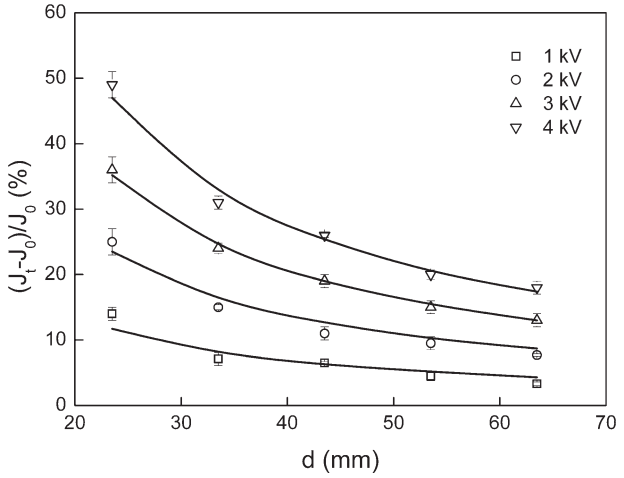

Figure 5. Effect of electrode distance on the enhancement in water permeation.

Needle⊥plate electric field; needle electrode: anode; plate electrode: grounded. Operating conditions same as specified in Figure 4. Solid line represents the calculated value.

there was any change in the membrane by the electric field. To analyze the contribution of the electric field on water permeation, the enhancement in water permeation G was defined as

$$G=\frac{J_t-J_o}{J_o}\cdot 100\% \tag{2}$$

where subscripts t and o represent the total water flux with and without the electric field, respectively. Because the study focused on using an electric field to accelerate water transport through the membrane, pure water was initially used as the feed. Then, an aqueous NaCl solution at 0.1 M was used as the feed to test the performance of the MD process for desalination applications. In the latter case the salt rejection R was defined as

$$R=\left(1-\frac{C_\mathrm{p}}{C_\mathrm{f}}\right)\cdot 100\% \tag{3}$$

where C_f and C_p are the concentrations of the salt in the feed and permeate, respectively. The salt concentration was determined using a conductivity meter (Ohaus, STARTER 3C). All the experiments were repeated at least three times to ensure reproducibility of the measurements.

Results and Discussion

Verification of asymmetric electric field on enhancement in water permeation

To verify that the asymmetric electric field can enhance mass transport of water across the membrane, experiments were carried out using pure water as the feed with and without the asymmetric electric field. Figure 4 shows that water permeation is indeed enhanced by the asymmetric electric field. When a voltage was applied to the needle and disk electrodes on the two sides of the membrane, water in the feed side was subjected an electric force to the same direction of permeation, leading to a faster mass transport. The direction of the electric gradient force is from the region with a lower field intensity to the region with a higher field intensity. The direction of the electric gradient force depends on the gradient of field intensity rather than the field direction which is from the anode to the cathode. Therefore, with the needle⊥plate electric field, the direction of electric force acting on water is in the direction from the feed to the permeate side. The data in Figure 4 show that no matter how the electrodes were connected to the power supply, at a given voltage, the enhancement in water permeation is essentially the same. This suggests that the direction of migration of water molecules in an asymmetric electric field is not correlated to the direction of the electric field, but is determined by the electric potential gradient only.

It needs to mention that PTFE is a very good insulator and thus have a high breakdown voltage, and the electric field strength in this study was not sufficiently high to cause sparkover. When the voltage applied on the electrodes was removed, the water permeation flux gradually decreased to the same level as the initial flux in the absence of the electric field (i.e., conventional vacuum MD), indicating that the membrane was intact in the electric field, at least within the experimental range of field intensities tested.

Optimization of the asymmetric electric field

The thermal motion of molecules tends to prevent the orientation and directed migration of dipoles along the electric field. Therefore, increasing the strength of the electric field or the gradient of electric potential will make dipoles aligning more orderly and migrating faster. The electric potential gradient is determined by such parameters as the magnitude of voltage applied, the distance between the electrodes, and the shapes of the electrodes used. With an increase in the voltage applied, water molecule was subjected a higher electric gradient force, which led to a higher permeation rate, as shown in Figure 4. When the applied voltage reached to 4 kV, the water permeation flux was increased by about 50% as compared to conventional vacuum MD under the same operating conditions in the absence of the electric field.

The electric potential gradient also depends on the distance between the electrodes. At a given applied voltage, increasing the distance between the electrodes will decrease the intensity of the electric field, resulting in a reduction in

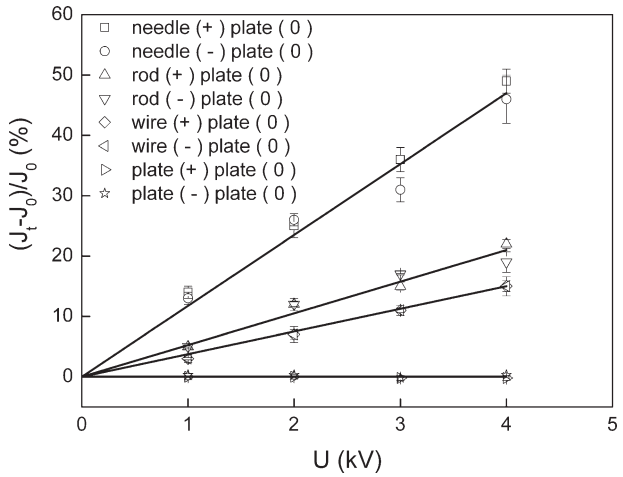

Figure 6. Effect of electrode configuration on enhancement in water permeation.

Needle, rod, wire, or plate electrode in the permeate side: anode or cathode; plate electrode in the feed side: grounded. Operating conditions same as specified in Figure 4.

the electric potential gradient. This will reduce the enhancement in water permeation by the electric field, as shown in Figure 5. The distance between the electrodes should not be too far away in order to maintain a strong electric field, but conversely, the two electrodes cannot be too close to each other in order to prevent voltage breakdown. This should be considered in equipment design for practical applications.

Concerning the electrodes used on the permeate side, two other different geometries (i.e., rod and wire) were also tested. All three configurations of electrodes (i.e., needle-plate, rod⊥plate, and wire∥plate) can generate an asymmetric electric field across the membrane with the field intensity increasing from the feed side to the permeate side to enhance water permeation. As shown in Figure 6, the water permeation flux was enhanced with applied voltage for all the electric field configurations. As the field intensity near a conducting surface increases with the surface curvature, the needle electrode with a thin sharp point can produce a higher electric potential gradient than other configurations (i.e., rod and wire). Therefore, for a given electrode distance at a given voltage applied, the needle⊥plate configuration yielded the best performance for enhancing water transport across the membrane among the three electric field configurations, whereas the wire∥plate configuration was the least efficient. The electric field between a pair of parallel plates is a uniform field, and there is no net force exerted on polar molecules. Therefore, water permeation was not accelerated in the electric field produced by the plate∥plate configuration. This result proves that a nonuniform electric field is essential to obtain an additional driving force for water transport.

Mass transport in asymmetric electric field enhanced MD

The permeation flux J of a membrane process can be expressed by a phenomenological equation

$$J = D\left|\frac{\partial \Phi}{\partial x}\right| \tag{4}$$

where D is the overall mass-transfer coefficient, $\partial\Phi/\partial x$ the driving force, that is, gradient of Φ (e.g., pressure, concentration, or electrical potential) in the direction x perpendicular to the membrane surface.[24]

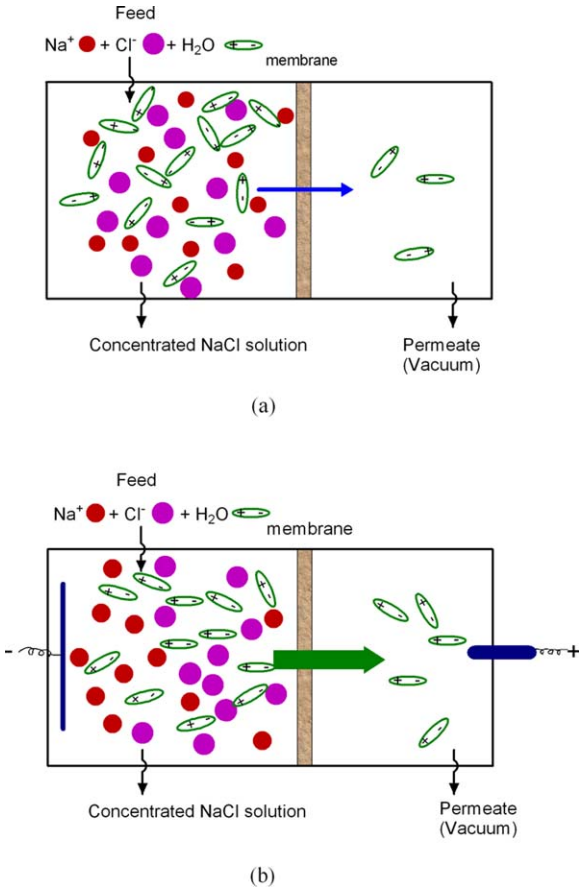

Figure 7. Schematic diagram of MD.

(a) Conventional system and (b) asymmetric electric field enhanced system. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

The mass transport in conventional MD has been well documented and can be summarized as follows: (1) evaporation of water at the liquid–vapor interface immobilized at the pore entrance and (2) mass transfer of vapor through the membrane pores.[25] The driving force for mass transport across the membrane is the transmembrane vapor pressure gradient, and the water permeation flux is J_0

$$J_0 = -D_p \frac{\partial p}{\partial x} \tag{5}$$

where D_p is the mass-transfer coefficient of water across the membrane driven by the pressure gradient $\partial p/\partial x$ (see Figure 7a). When the pore size of the membrane is sufficiently small relative to the mean free path of the permeant molecules, Knudsen diffusion will dominate and the mass-transfer coefficient will be related to the Knudsen diffusivity.

In the presence of an asymmetric electric field, water molecules not only permeate to the downstream side under the pressure gradient, but also migrate to the direction of increasing field intensity under the electric potential gradient. The total permeation flux J_t can thus be expressed as

$$J_t = -D_p \frac{\partial p}{\partial x} + D_e \frac{\partial U}{\partial x} \tag{6}$$

where D_e is the mass-transfer coefficient due to migration of water molecules under the electric potential gradient $\partial U/\partial x$.

Obviously, the coefficient D_e depends upon the dielectric constant of water. The first term on the right side of the equation represents the permeation driven by the pressure gradient, and the negative sign represents the pressure gradient is in the opposite direction of the permeation. The second term represents the contribution of molecular migration to the permeation under the electric potential gradient, and the positive sign represents that the molecular migration is along the direction of increasing electric potential. When the electric potential across the membrane increases from the feed side to the permeate side, the migration of water driven by the electric potential gradient is in the same direction with the permeation driven by the pressure gradient, and as a result, the mass transport of water molecules across the membrane is intensified (see Figure 7b).

As an approximation, Eq. 6 can be rewritten to give the following integrated form if the electric potential gradient is a constant in the electric field (i.e., $\partial U/\partial x = U/d$)

$$J_t = J_0 + D_e \frac{U}{d} \tag{7}$$

where U is the applied voltage on the electrodes across the membrane and d the distance between the electrodes. Equation 7 suggests that plotting water flux vs. voltage gradient will yield a straight line with a slope equal to D_e. This has been shown to be the case in Figures 4 and 6. However, it should be mentioned that strictly speaking in the electric fields studied here, the electric potential gradient perpendicular to the membrane surface is not uniform (see Figure 1) and $\partial U/\partial x$ along the permeation direction is not a constant over the entire membrane surface. Nevertheless, for the small membrane area used in this study, the above approximation is shown to work well, and the D_e values can be determined to be 1.8×10^{-4}, 8×10^{-5}, and 6×10^{-5} kg/(m^2 h kV/m) for the needle⊥plate, rod⊥plate, and wire∥plate configurations, respectively.

It was also observed during the experiments that the electric current increased with an increase in the electric field strength, but maximum current was below 0.1 mA. This suggests that the electric field enhanced MD process consumes very little power and is energy efficient. This process intensification is especially advantageous for treating heat sensitive products where increasing operating temperature to enhance the permeation flux is not viable.

It may be pointed out that using an asymmetric electric field to enhance membrane separation is not only limited to MD, and it can in principle be applied to any separation processes (e.g., gas separation and pervaporation), as long as the components to be separated have different polarities, in order to improve the separation efficiency. The acceleration effect of the electric field on the permeation flux depends on the polarity of the permeating components.

Asymmetric electric field enhanced MD for desalination

Previous results show that the permeation of water molecules was enhanced remarkably by an asymmetric electric field across the membrane. The effects of the electric field on the water flux and salt rejection for water desalination are investigated here. Salts soluble in water dissociate into ions. When an electric field is applied, the plate electrode on the feed side, which directly contacts with the salt solution, tends to attract counterions from the feed. An electric double layer will be formed at the interface between the plate electrode and the bulk feed solution. The electric potential of the double layer can weaken the electric potential gradient of the electric field generated externally, and therefore, the electric gradient force acting on water molecules will be reduced. As shown in Table 1, although the asymmetric electric field was effective in enhancing water permeation from a salt solution, the enhancement in water flux was about 10% lower than that in the case of pure water permeation. Although the salt is nonvolatile and only water vapor is expected to pass through the membrane, any leakage or crossover of feed through the membrane due to defects (large pores or pinholes) on the membrane will lower the salt rejection. Because of the enhanced water permeation by the electric field, the salt concentration in the permeate become lower than that obtained by conventional MD in the absence of electric field, resulting in a better salt rejection, as shown in Table 1. The increased salt rejection indicates again that the electric field did not have any destructive effect on the membrane. These results demonstrate that by applying an asymmetric electric field across the membrane, both the water flux and the product water quality can be improved.

Table 1. Comparison of Asymmetric Electric Field Enhanced Membrane Distillation and Conventional Membrane Distillation on the Water Flux and Salt Rejection

		R (%)	
Feed	$(J_t - J_0)/J_0$ (%)	Conventional Process	Coupled Process
Aqueous NaCl solution (0.1 M)	28 ± 1	99.22 ± 0.01	99.62 ± 0.09
Pure water	31 ± 1	–	–

Needle⊥plate electric field; needle electrode: anode; plate electrode: grounded; voltage applied: 4 kV; electrode distance: 33.5 mm. Feed and permeate temperature: 20°C; feed flow rate: 60 L/h; and permeate pressure: 20 kPa absolute.

Conclusions

The concept of using asymmetric electric field to enhance mass transport in MD was demonstrated, and the following conclusions can be drawn:

1. An asymmetric electric field across the membrane was so established that the direction of the electric gradient force was the same as the permeation, which was shown to enhance water transport through the membrane.
2. The electric potential gradient depended on the voltage applied, the distance between the electrodes and the geometry of the electrodes. The enhancement in water permeation was more significant with increasing the applied voltage, decreasing the electrode distance or using an electrode with a sharp point on the permeate side.
3. The driving force for mass transfer under the pressure field and the driving force for mass transfer under the electric field were independent, and the electric field-enhanced MD was effective especially for water permeation in applications operated at a low pressure gradient (e.g., a low feed temperature or a low permeate vacuum).
4. The electric field enhanced MD was shown to perform better for water desalination than conventional vacuum

MD in terms of water productivity and salt rejection. There was very little power consumption in creating the electric field that could enhance water transport significantly.

Acknowledgment

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