Permeate Flux and Rejection Behavior in Submerged Direct Contact Membrane Distillation Process Treating a Low-Strength Synthetic Wastewater

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Abstract: The effects of operational conditions such as permeate recirculation velocity, mixing intensity, and trans-membrane temperature on the performances of hydrophobic polyethylene (PE) hollow-fiber membrane were investigated by operating the submerged direct contact membrane distillation (SDCMD) process treating a synthetic low-strength wastewater. Permeate flux of the membrane increased with increasing a permeate recirculation velocity through the fiber lumen. However, the effectiveness was less pronounced as the velocity was higher than 0.5 m/s. Increasing rotational speed to 600 rpm, which can lead to mixing intensity from a bulk wastewater toward hollow-fiber membrane, enhanced permeate flux. Feed temperature played a more significant role in enhancing permeate flux rather than a permeate temperature under constant trans-membrane temperature. The SDCMD process treating a synthetic low-strength wastewater achieved an excellent rejection efficiency which is higher than 97.8% for both chemical oxygen demand (COD$_{Cr}$) and total phosphorus (T-P) due to the hydrophobic property of membrane material which can allow water vapor through membrane. However, the rejection efficiency of the ammonia nitrogen (NH$_3$-N) was relatively low at about 87.5% because ammonia gas could be volatilized easily through membrane pores in SDCMD operation. In a long-term operation of the SDCMD process, the permeate flux decreased significantly due to progressive formation of inorganic scaling on membrane.

Keywords: direct contact membrane distillation; organic wastewater; nutrients; inorganic scale

1. Introduction

Membrane distillation (MD) is a thermal-driven separation process that uses hydrophobic membranes as an excellent way to treat water and wastewater. Unlike pressure-driven membrane process, the driving force to produce membrane permeate is the vapor pressure difference between the two sides of the membrane pores, allowing for the mass transfer of the volatile solution components [1–3]. In addition, hydrostatic pressure encountered in MD is lower than that used in pressure-driven membrane processes such as reverse osmosis (RO) and nanofiltration (NF) [4]. One of the great advantages of the MD technology in water and wastewater treatment is that the industrial sources and renewable energies such as waste heat or solar energy can be used as the heat energy to operate MD system [3,6]. Interests in MD technology have been growing rapidly in the areas of seawater desalination due to relatively low fouling potentials and energy requirements compared to high-pressure driven membrane process [7,8]. Nevertheless, the MD process has disadvantages such as low permeate flux compared to NF or RO and high susceptibility permeate flux to the concentration and temperature of the feed conditions due to the concentration and temperature polarization phenomena [4]. Furthermore, few studies have been done to better understand the performances of MD processes in wastewater treatment in comparison to seawater desalination [9,10].
Direct contact membrane distillation (DCMD) is the most commonly used MD module configuration. In DCMD, both feed wastewater and permeate are in direct contact with a hydrophobic membrane. The water vapor transferred across the membrane is directly condensed in the permeate produced by the membrane module from which temperature is lower than feed one [4]. The DCMD is attractive particularly for wastewater treatment because it does not require external equipment such as vacuum pump or condensation plate, which may be necessary for other types of MD configuration such as air gap membrane distillation (AGMD), sweeping gas membrane distillation (SGMD) or vacuum membrane distillation (VMD) [11–13]. For DCMD, a submerged configuration using a bundle of hydrophobic hollow-fiber membranes is preferentially used [14–16]. Furthermore, the DCMD configuration is especially suitable for hybrid membrane technologies such as membrane distillation bioreactor (MDBR) or integrated forward osmosis-membrane distillation (FO-MD) [17–19].

Hollow-fiber membrane is considered more favorable than flat sheet membrane in submerged direct contact membrane distillation (SDCMD) process due to its high packing density as well as relative easiness in membrane module design. Nevertheless, thermal efficiency can often be lower as membrane packing density is higher because the permeate can be heated up through the geometry of hollow-fiber membrane [20–22]. Most of all, hydrodynamic conditions should affect trans-membrane temperature and permeate flux because they can influence temperature polarization. Recently, the interest in MD processes such as submerged configuration have grown rapidly for resource recovery from brine solution [23–25]. In a submerged MD configuration, temperature polarization across the membrane should occur more easily due to the lack of turbulence on the membrane surface compared to the cross-flow MD system [26]. However, the submerged MD system requires less energy and a smaller foot-print than that needed to generate a high cross-flow velocity from a side-stream of MD process. In addition, the submerged MD can be integrated with a bioreactor to obtain energy more easily, for example, anaerobic MD bioreactor process [19]. In spite of many advantages, there is still lack of effort applying the SDCMD process to understand the effect of operational conditions on MD performances in treatment of low-strength wastewater. The objectives of this study are to investigate the effect of operational conditions including permeate recirculation velocity into the lumen of hollow-fiber MD membrane and mixing intensity in SDCMD treating low-strength wastewater. Permeate flux was investigated systematically with respect to operational conditions. Membrane performances, such as the rejection of organics and nutrients including ammonia nitrogen (NH$_3$-N) and total phosphorus (T-P), were also observed by conducting short-term and long-term operation with SDCMD.

2. Materials and Methods

2.1. Feed Wastewater and Membrane

In this study, synthetic wastewater consisting of sodium acetate anhydrous (C$_2$H$_3$NaO$_2$, Duksan Chemicals, Korea) and sodium propionate (C$_3$H$_5$NaO$_2$, TCI Chemicals, Korea) yielding chemical oxygen demand (COD$_{cr}$) was prepared. Ammonium chloride (NH$_4$Cl, Duksan Chemicals, Korea), potassium phosphate monobasic (KH$_2$PO$_4$, Daejung Chemicals, Korea), and sodium bicarbonate (NaHCO$_3$, Duksan Chemicals, Korea) were prepared and applied as feed wastewater for SDCMD experiments. The synthetic wastewater tested in this study was based upon the low-strength feed wastewater prepared as feed for laboratory-scale anaerobic bioreactors [27,28]. The characteristics of the synthetic wastewater used in this study are shown in Table 1.

| Chemical Oxygen Demand (mg COD/L) | Ammonia Nitrogen (mg NH$_3$-N/L) | Total Phosphorus (mg PO$_4^{3-}$-P/L) | Conductivity (µS/cm) | pH |
|----------------------------------|----------------------------------|--------------------------------------|---------------------|----|
| 282.6 ± 5.7                     | 32.2 ± 1.3                       | 5.3 ± 0.1                            | 971.4 ± 4.0         | 7.7 ± 0.1 |
The polyethylene (PE) hollow-fiber membrane (Econity Co. Ltd., Yongin, Korea) was available with high hydrophobicity (contact angle is about 130°) and used for operating SDCMD system. The mean pore size of the commercially available PE membrane was customized as 0.1 μm and the porosity of the membrane was 50%. The outer and inner diameter of PE hollow-fiber membrane was 1100 and 750 μm, respectively. In this study, twenty fibers were assembled into a membrane holder specially designed and sealed by epoxy resin. The effective surface area of the hollow-fiber membrane tested was 84.3 cm². The fiber length and distance between each fiber were 9.9 and 1.6 cm, respectively.

2.2. Experimental Set-Up and Performance Tests

A schematic diagram of experimental set-up of SDCMD process is illustrated in Figure 1. The synthetic wastewater prepared was placed into a feed tank with 5 L of effective volume made by tempered glass and stainless steel, in which a magnetic bar was stirred for generating mixing intensity in bulk wastewater. The feed temperature was controlled by using a digital hotplate (MSH-20D, Daihan Scientific, Wonju, Korea). The hollow-fiber membrane module was submerged horizontally and located at about two third of depth from the free surface of wastewater in the feed tank. Digital pressure gauges were connected at inlet and outlet tubing from MD membrane to monitor hydraulic pressure of fiber lumen. Two thermometers were submerged into both feed and permeate tank to monitor water temperatures. Permeate produced by the hollow-fiber membrane was recirculated into the permeate tank with 0.5 L of volume through a water chiller (RBC-20, JEIO Tech, Daejeon, Korea) using a gear pump (WT3000-1FA, Longer Pump, Hebei, China). Deionized (DI) water was filled initially in the permeate tank. The weight of permeate accumulated into the permeate tank with time was measured by using an electric balance (FZ-5000i, A&D Company, Tokyo, Japan). The SDCMD tests were performed over both the short-term (1 h) and long-term period (180 h) in this study.

![Figure 1. Experimental set-up of laboratory scaled SDCMD process (PI: pressure indicator, TI: temperature indicator).](image)

2.3. Analytical Methods

Permeate flux (N) from the hollow-fiber membrane in operating the SDCMD process was calculated by measuring the weight of permeate with time on electric balance and effective surface area of membrane using Equation (1) as below [29].

$$N = \frac{\text{Weight increase of permeate tank (kg)} \div \text{density of water (kg/L)}}{\text{membrane effective area (m}^2\text{)} \times \text{operation time (h)}}$$  \hspace{1cm} (1)

The density of membrane permeate was considered as the same as to the one of the pure water ($\rho_w = 10^3$ kg/m³). The permeate temperature was maintained as 20 °C by using the chiller yielding experimental error of permeate flux caused by the change of permeate temperature was within 0.2%.
The rejection of organic contaminants and nutrients such as ammonia nitrogen and total phosphorus were estimated by using Equation (2) [30].

\[
\text{Rejection (\%)} = \left(1 - \frac{C_p}{C_f}\right) \times 100
\]  

(2)

where \(C_p\) and \(C_f\) are concentration of target compound to be removed in permeate at the end of SDCMD operation and feed wastewater, respectively. A permeate pH was measured by using a pH meter (Orion VersaStar Pro, Thermo Scientific, Waltham, MA, USA). The COD of the feed wastewater was measured by Standard Methods (APHA, 2012) while ammonia nitrogen and total phosphorus were determined by using reagent kits and spectrophotometer (DR3900, Hach Lange, Loveland, CO, USA. Concentration ranges detected by reagent kits to measure ammonia nitrogen and total phosphorus were 0.4–50.0 mg/L NH\textsubscript{3}-N, 0–3.5 mg/L PO\textsubscript{4}\textsuperscript{3–}-P, respectively.

3. Results and Discussion

3.1. Effect of Permeate Recirculation Velocity and Trans-Membrane Temperature

Figure 2A illustrates the permeate flux with respect to the permeate recirculation velocity at feed and permeate temperature of 55 and 20 °C, respectively. The permeate recirculation velocity was calculated by dividing recirculating permeate flow rate by cross-sectional area of fiber lumen. The results show that permeate flux increases with increasing permeate recirculation velocity but approaches an asymptotic value when the velocity is higher than 0.5 m/s. An increase in the permeate recirculation velocity could prevent temperature drop in MD permeate while it was flowed through fiber lumen. Thus, vapor pressure difference can be maintained so that it can result in stable permeate flux [31,32]. As the permeate recirculation velocity increases, the Reynolds number (Re) also increases along the hollow-fiber membrane. When the permeate recirculation velocity increased from 0.2 to 0.8 m/s, the value of Re was increased from 332 to 1162 based upon water density and viscosity at 20 °C. Since the conductivity in MD permeate was very low (less than 30 µS/cm) as observed in this study, the experimental error in estimating Re at 20 °C was within 0.2% only. The permeate flux increased proportionally with Re number, but the flux approached to the steady value as it was higher than 664. The tendency of experimental result shows that the thickness of thermal boundary layer in permeate stream is reduced by increasing the permeate recirculation velocity even under laminar region (Re < 2100). As a result, an increase of permeate recirculation velocity enhances transverse vapor flux through membrane [33–35].

Figure 2A also shows the effect of mixing intensity driven by the rotational speed using a magnetic bar in the feed tank on MD performances. Enhancing the mixing intensity of feed wastewater tends to reduce concentration and temperature polarization layer on membrane surface [36,37]. Nevertheless, the beneficial effect of mixing intensity on the permeate flux was not observed as the rate was higher than 600 rpm. In the SDCMD process, inducing mixing intensity in feed wastewater should be the only way to control thermal polarization formed on membrane due to temperature gradient. Higher mixing in feed wastewater is associated with more turbulence on the membrane surface, thereby improving the permeate flux across membrane. However, there was no beneficial effect on enhancing permeate flux as the mixing intensity was higher than the critical value probably due to the limitation of the size of the wake region in fluid mixing which creates secondary flows and/or local mixing on membrane.

The permeate fluxes through the hydrophobic membrane in SDCMD process vary dependent upon the feed temperature due to water vapor pressure change [38,39]. Figure 2B shows that the permeate flux increases as trans-membrane temperature increases from 15 to 40 °C almost exponentially under constant permeate recirculation velocity and mixing intensity of 0.5 m/s and 600 rpm, respectively. The extent to which the enhancement of permeate flux was more pronounced by higher feed temperature rather than that at lower permeate temperature. Permeate flux increased linearly with the permeate temperature as the feed temperature was fixed at 55 °C, indicating that controlling feed temperature
should affect permeate flux more dominantly. The theoretical value of permeate flux was calculated by using the MD coefficient from the preparatory experiment using DI water under the corresponding operational condition with Equations (3) and (4).

![Graph A](image1)
![Graph B](image2)

**Figure 2.** Permeate flux changes with operational parameters (A) permeate recirculation velocity and mixing intensity, (B) trans-membrane temperature.

The mass transfer coefficient was estimated as an indicator to observe the impact of temperature on permeate flux using Equation (3) as below.

\[ N_v = C \times \Delta P = C \times (P_{fm} - P_{pm}) \]  

where \( N_v \) is vapor flux in SDCMD process, \( C \) is MD coefficient, \( \Delta P \) (kPa) is the pressure difference, \( P_{fm} \) and \( P_{pm} \) are vapor pressure on the membrane surface at feed and permeate side, respectively. Water vapor pressure was calculated by using Antonine equation as below [40].

\[ P = \exp\left(23.238 - \frac{3841}{T_m - 273.15}\right) \]  

where \( P \) is the vapor pressure of solution, \( T_m \) is the temperature of solution near membrane surface as unit of Kelvin (K). Permeate fluxes calculated by using mass transfer coefficient agreed very well with experimental observations, as shown in Figure 2B. Overall MD coefficient calculated by Equations (3) and (4) was within 6.8% of experimental error, suggesting that the membrane permeability in SDCMD be remained at an average 0.4 L/m²-hr-kPa.

### 3.2. Variations of Solution pH with Operational Conditions

Figure 3 shows the changes of solution pH measured in both feed wastewater and permeate produced by SDCMD process with changes of operating parameter. Experiments were performed under the permeate recirculation velocity of 0.5 m/s, mixing intensity of 600 rpm, feed temperature of 55 °C and permeate temperature of 20 °C for each experimental condition. As shown in Figure 3A, increasing a permeate recirculation velocity as mixing intensity, feed and permeate temperature are constant reduces a permeate pH from 7.1 to 6.7. This can be explained by the transport of CO₂ from the feed wastewater toward permeate side through hydrophobic MD membrane [41]. Since higher recirculation velocity through fiber lumen can mitigate the increase in permeate temperature during MD operation, it allows more CO₂ dissolved in MD permeate. Furthermore, mass transfer through the membrane walls can be improved by increasing permeate recirculation velocity. It has been reported that enhancing mass transfer allows the passage of volatile fatty acid (VFA) such as sodium acetate and sodium propionate in feed wastewater through membrane easily [42]. However, there was no significant impacts of mixing intensity on solution pH when other operational parameters
were constant as shown in Figure 3B. This may be caused by the fact that mixing in feed wastewater through SDCMD process can reduce the concentration or thermal polarization on the outer surface of membrane more rather than mass transfer of CO$_2$ towards the fiber lumen.

![Figure 3](image)

**Figure 3.** Variations of pH in feed wastewater and permeate after 1 h of operation time with respect to the permeate recirculation velocity (A), mixing intensity (B), feed temperature (C) and permeate temperature (D) (background operational condition; permeate recirculation velocity of 0.5 m/s, mixing intensity of 600 rpm, feed temperature of 55 °C and permeate temperature of 20 °C).

The results shown in Figure 3C indicates that the increase of feed temperature leads to an increase of the solution pH in both feed wastewater and permeate as recirculation velocity and mixing intensity are 0.5 m/s and 600 rpm, respectively. However, as shown in Figure 3D, increasing the permeate temperature did not change the pHs in both feed wastewater and MD permeate. Ammonia transfer is driven by the difference in partial pressure of ammonia gas between feed wastewater and permeate. An explanation is that higher feed temperature can allow more ionization of the bicarbonate ions (HCO$_3^-$) into carbonate ions (CO$_3^{2-}$), leading to increase a feed wastewater pH [43]. In addition, an increase in temperature of feed wastewater should facilitate the volatilization of ammonium ion (NH$_4^+$) into ammonia gas which can be passed through the hydrophobic membrane easily. With synthetic wastewater tested in this study, ammonia nitrogen can exist mainly as two forms of nitrogen consisting of volatile ammonia and ammonium ions. The solubility of ammonia in feed wastewater can be decreased by increasing its temperature. The vaporization of dissolved ammonia across the membrane should reduce the rejection of ammonium nitrogen. As a result, permeate pH should be increased due to the formation of ammonia nitrogen as shown below [44].

$$\text{NH}_3 + \text{H}_2\text{O} \leftrightarrow \text{NH}_4^+ + \text{OH}^-$$ (5)
At the 25 °C feed temperature, the rate constant in forward reaction \( (k_A) \) in Equation (5) is \( 1.8 \times 10^{-5} \) while the constant in backward reaction \( (k_B) \) is \( 5.6 \times 10^{-10} \). This means that the rate of forward reaction with the formation of ammonium ion is almost \( 3.2 \times 10^4 \) times faster than that occurring toward the formation of ammonia gas [45]. Consequently, the ammonia gas passed through the membrane can be ionized into ammonium ion quickly, thereby increasing the permeate pH [43,46].

3.3. Membrane Rejections

Figure 4 shows rejection efficiencies of organics and phosphorus at the same operational conditions applied in Figure 3. Results in Figure 4A through D show that the rejection efficiency of organics and phosphorus are very high as 98.3 and 99.9%, respectively regardless of all operational conditions such as permeate recirculation velocity, mixing intensity, feed temperature and permeate temperature. The phosphate ions \( (PO_4^{3-}) \) can lead to the formation of inorganic precipitation which can be associated with magnesium and calcium ions such as \( Mg_3(PO_4)_2, Ca_3(PO_4)_2 \) or \( NH_4MgPO_4 \) and this can contribute to high phosphorus rejection efficiency [47–49]. However, the rejection of ammonia nitrogen was relatively low, yielding 80.6%–91.3% depending upon operational conditions. Figure 4C shows that increasing feed temperature reduces the rejection of ammonia significantly. A lower rejection of ammonia nitrogen is caused by more fraction of NH3 gas formed in MD permeate at higher feed temperature due to its more vaporization through the membrane in the SDCMD process as discussed above [50,51].

![Figure 4](image_url)

**Figure 4.** Rejection efficiency of COD\(_{Cr}\), NH\(_3\)-N and T-P with operational parameters of SDCMD process after 1 h of operational time with respect to permeate recirculation velocity (A), mixing intensity (B), feed temperature (C) and permeate temperature (D) (background operational condition; permeate recirculation velocity of 0.5 m/s, mixing intensity of 600 rpm, feed temperature of 55 °C and permeate temperature of 20 °C).
3.4. Long-Term Operation of SDCMD Process

Long-term operation of the SDCMD process was conducted at 0.5 m/s of permeate recirculation velocity, 600 rpm of mixing intensity, 55 °C of feed temperature, and 20 °C of permeate temperature. A temperature in feed wastewater was replenished regularly to maintain a constant water level in the feed tank. Permeate flux from the membrane decreased slowly with operation time, but it was dropped rapidly after 72 h of operation (Figure 5A). Mass transfer coefficients of organics and nutrients are also shown in Figure 5B. The concentration of each component was measured immediately after taking the samples. As mentioned above, ammonia nitrogen in feed wastewater can be passed through the membrane as form of ammonia gas which can be continuously transferred into permeate water. The mass transfer coefficient associated with ammonia nitrogen was estimated about 131.9 ± 12.1 mg/m²·h during 180 h operation. The concentration of ammonia nitrogen in MD permeate was similar to the concentration of it from the feed wastewater after 24 h continuous SDCMD operation.

Figure 5. Variation of permeate flux (A) and mass transfer coefficients (B) in long-term operation.

From the SDCMD process, the transfer of organic compounds such as VFA can occur through membrane. The results show that the mass transfer of VFA through MD membrane slightly increased and reached at 59.5 ± 4.8 mg/m²·h at the end of the long-term experiment due to volatilization of VFA as discussed [29]. In contrast, the SDCMD process showed excellent rejection of phosphorus yielding relatively low mass transfer coefficient which is about 1.3 ± 0.4 mg/m²·h as compared to organics and ammonia nitrogen. Therefore, it appeared that membrane wetting did not affect membrane performances significantly during the long-term operation of SDCMD.

A reduction in permeate flux from the MD membrane is caused by membrane fouling formed, as evidenced by scanning electron microscope (SEM) images and energy dispersive X-ray (EDX) analysis. External fouling on the membrane surface became more serious during longer SDCMD operation. Figure 6 shows the presence of carbon (C), nitrogen (N), oxygen (O), magnesium (Mg), phosphorus (P), and calcium (Ca) ions on the membrane surface as detected by EDX analysis using fouled membrane (Figure 6D). The presence of C, O, and N peaks is often representative of organic fouling [52]. The detection of Ca, P, and Mg ions on membrane can also support the presence of inorganic precipitates such as CaCO₃, MgCO₃, and Ca₃(PO₄)₂ [53]. Among these inorganic scales, the CaCO₃ and Ca₃(PO₄)₂ are thought to be dominant as external resistance against permeate flux through MD membrane. Obviously, high rejection of phosphorus across SDCMD process as shown in Figure 5B is not only caused by the membrane rejection but also due to the formation of inorganic precipitates on the membrane surface [54,55].
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Figure 6. SEM/EDX analysis of raw (A,C) and fouled (B,D) membrane surface.

4. Conclusions

Attempts were made by applying the SDCMD process for improving the rejection of contaminants such as nutrients present in low-strength synthetic wastewater. The results obtained show that the increase in permeate recirculation velocity through the lumen of the hollow-fiber membrane treating synthetic wastewater increased permeate flux. However, there was no beneficial effect on permeate flux as the velocity was higher than 0.5 m/s. Similarly, permeate flux was enhanced by increasing a mixing intensity in the SDCMD process, but the effectiveness was limited as the intensity was higher than 600 rpm. Permeate flux was affected by controlling the feed temperature more dominantly than permeate temperature. For all operational conditions, the rejection of organics and phosphorus were higher than 98.3% and 97.8%, respectively, but the rejection of ammonia nitrogen was relatively low (87.5%). Increasing feed wastewater temperature decreased the rejection of ammonia nitrogen due to its volatilization across the membrane into ammonia gas. Further research is needed to fully understand the performances of SDCMD process using real wastewater such as domestic sewage. More research should also be directed at developing hybrid MD combined to improve the removal of nutrients and energy saving.

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