

# Modification of Vacuum Membrane Distillation with Humid-air Recirculation and Double-stage Condensation

Nitcha Kaewseng, Lalinee Tubtimthong, Monthon Thanuttamavong\*, Chart Chiemchaisri

**Abstract**— Enhancement of water productivity from conventional vacuum membrane distillation (CVMD) was achieved by applying recirculation vacuum membrane distillation (RVMD) using tubular membrane module. With a mild condition of 50°C feed water temperature, 30°C cooling water temperature and -60 kPa transmembrane pressure, the maximum water flux of RVMD system was found at 4.04 L/m<sup>2</sup>·h while the highest water flux of CVMD system was reached only 2.89 L/m<sup>2</sup>·h at relatively lower cooling water temperature of 10°C. As the results, the water productivity in RVMD was significantly enhanced, according to recirculation of remained humid air back into the lumen membrane module could increase transmembrane temperature and permeate vapor transport. Moreover, double-stage condensers was introduced to assure complete dew formation in CVMD system and extended extra water condensation in RVMD system. In addition, effects of feed salt concentration on permeate flux were also observed. The results showed that increases of feed concentration from 0 to 30 g/L had decreased permeate fluxes from 4.04 to 2.33 L/m<sup>2</sup>·h, in spite of change in salt rejection efficiencies achieved as high as 99.4% throughout the steady stages. Therefore, the modification of humid-air recirculation coupled with double-stage condensation could accomplish productivity enhancement for the innovative RVMD.

**Index Terms**— Dew-point temperature, double-stage condensation, humid-air recirculation, vacuum membrane distillation, water productivity

## I. INTRODUCTION

Membrane distillation (MD) process has been known since the late 1960s. It is a technology contributed in the research for seawater desalination and emerging technology, which can be used as a substitute for conventional desalination processes such as MSF and RO [1-3]. The MD is a relatively new membrane separation process which might overcome some limitations of the more traditional membrane technologies. In recent years, MD has been developed for processing high salinity solution due to its particular separation effects and promising application prospects, it has been paid more and more attention. In particular, high solute concentrations can be reached and ultrapure water can be produced in a single step [4]. A typical MD system involves salt solution at a higher temperature on one side of the membrane, and cooling water or gas on the other side [5]. MD is a thermally driven process, in which only vapor molecules are transported through porous hydrophobic

membrane. The liquid feed to be treated by MD must be maintained in direct contact with one side of membrane without penetrating its dry pores unless a transmembrane pressure higher than the membrane liquid entry is applied [4,6]. In theory, the MD has work ability up to 100% salt rejection and can be operated at low temperature that is usually controlled between 40-80°C [7, 8] and at atmosphere pressure, low-grade energy like solar and waste energy can be used for MD water desalination [4]. The MD process consists in a variety of configurations including direct contact membrane distillation (DCMD), vacuum membrane distillation (VMD), air gap membrane distillation (AGMD) and sweeping gas membrane distillation (SGMD) [9]. Of the four configurations, most research focuses on the DCMD, but the limitations of DCMD is vapor moving across the membrane is subject to mass resistance by air trapped in the pores [7]. Substituting DCMD with vacuum membrane distillation (VMD) will reduce this mass transfer resistance and enhance permeate flux [5]. Also, using an inert gas to evacuate the permeate vapor from the membrane module in the SGMD will raise the water production [2] unless it is concerned with the extra-exhausted gas.

In the VMD systems, the feed solution flows through the top of module while the permeate side is under vacuum condition and then subsequently external condensation. In this system, the vacuum pump is used to pull out non-condensable gases and moisture content. If non-condensable gases are not removed, it accumulates in the system and reduces the heat and mass transport. Hence, the vacuum pump is able to reduce the heat losses to obtain higher vapor flow rate in the system [8]. The performance of VMD depends on the differences of transmembrane pressure and transmembrane temperature. Chenggui Sun [10] reported that increase of the water vapor pressure over the liquid interface will increase the driving force for water vapor permeation across the membrane, resulting in an increase in the permeation flux due to vacuum pressure applied induce evaporation of liquid to increase. The rate of evaporation increases as temperature increases. This is so because an increase in temperature corresponds to an increase in the kinetic energy of molecules. At the same time, the rate of condensation increases as the number of particles in the vapor phase increases: more molecules strike the surface of the liquid. When these two processes become equal, the number of particles, and therefore the pressure in the vapor phase, becomes being stabilized. The value of the equilibrium vapor pressure depends on the attractive forces between particles of the liquid and on the temperature of the liquid. Vapor pressure above a liquid increases with increasing temperature [11].

On the other hand, to control and adjust the difference in transmembrane temperature at the surface membrane in the VMD system is quite difficult. Because of no fluid flow on the permeate side, contrasting to the adjustable

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transmembrane temperature differences on the surface membrane in DCMD system that uses the hot water and cold water through the membrane surfaces directly [12]. Likewise, the production of water vapor is much more efficient at higher temperature differences [2]. However, many previous VMD studies have never mentioned about drained-out air after condensation [13-16]. In fact, the VMD system still remains humid air as a byproduct that will be released to atmosphere. So, it will be an advantage if the humid air can be recirculated from the pump back into the module to improve the transmembrane temperature and permeate vapor convection.

The condenser system is an important part of VMD system since it is in charge for the dew formation or water condensation. Muñoz-García [17] expressed that the calculation for dew point temperature would depend not only on the air temperature but also on the air humidity as well. The effective dew formation requires a cold surface temperature below the dew point temperature to entrap drops of water. Moreover, the temperature of air will be lowered after condensation and will be helpful if it can be returned, as a sweep air, to the permeate side of VMD module. Hence, another condenser located next to the vacuum pump will be preferable to decrease the recirculated air temperature and to extend the detention time of dew formation. Consequently, the purpose of this work is to modify the configuration of VMD with humid-air recirculation and double-stage condensation for enhancement of water productivity, as well as to investigate removal efficiency of salt concentration.

## II. EXPERIMENTAL

### A. Membrane material and module

A polyolefin membrane with thickness of 0.30 mm was used as the MD membrane. The membrane had an average pore size of 0.014  $\mu\text{m}$  and porosity of 67.42%, examined with Mercury Porosimetry Analyzer. A contact angle of 124.3° was analyzed with a Ramehart contact angle goniometer of which the value could be quantified as a hydrophobic material. The membrane was configured as a tubular module with an effective surface area of 0.03  $\text{m}^2$ .

### B. VMD apparatus

The schematic diagrams of the laboratory scale vacuum membrane distillation systems with double-stage condensers used in this study are shown in Figure 1. Figure 1(a) describes conventional vacuum membrane distillation (CVMD) while Figure 1(b) represents recirculation vacuum membrane distillation (RVMD). Both systems mainly consisted of two thermostatic cycles: a feed cycle and vacuum cycle. The feed cycle compartment was connected to a feed tank together with a thermostatic water bath. The feed water temperatures could be adjusted to 30, 40 and 50°C, corresponding to a vapor pressure of 4.24, 7.37 and 12.33 kPa, respectively. The circulation feed rate was constantly controlled at 1.5 l/min with a water pressure of +10 kPa. Within this condition, consequently, the feed temperature inlet ( $T_{\text{feed in}}$ ) and feed temperature outlet ( $T_{\text{feed out}}$ ) were observed as having the same values as that of the feed tank temperature ( $T_f$ ). On the other hand, the vacuum cycle consisted of condenser I, condenser II, a vacuum pump and two water collectors. The condensers were heat exchangers using adjustable cooling tap water,

whose temperature ranged from 10 to 30 °C. A continuous operating time of 3 hours was set to ensure the steady-state condition. A two-stage diaphragm pump used as the vacuum pump was located between condenser I and II in order to extend the condensation time and reduce the heat transfer from the pump to the recirculated air, especially in the RVMD system. In the CVMD system, vacuum pressures were controlled with a pressure controlling valve [Fig.1(a)] and air from the vacuum pump was released out of the system. However, vacuum pressures of the RVMD system were adjusted with a recirculating valve [Fig.1(b)] and air from the vacuum pump was recirculated into the lumen side of the membrane, as a closed loop. The rate of recirculation was contrastingly affected by the vacuum pressure adjusted [Fig.3]. In both systems, because the feed water was pumped into the outer surface of the tubular membrane (+10 kPa) and the vapor was permeated to the lumen membrane by the vacuum pump in the pressure range of -10 to -50 kPa, consequently the transmembrane pressure ( $P_v - P_f$ ) through the membrane varied from -20 to -60 kPa

The permeate flux,  $J$  ( $\text{L}/\text{m}^2\text{h}$ ) was calculated by:

$$J = \frac{W}{A \times t} \quad (1)$$

where  $W$  was the mass of produced water,  $A$  was the effective membrane area, and  $t$  was the sampling interval.

The dew point temperature,  $T_{\text{dp}}$  (°C), and the effective dew-formation temperature,  $D$  (°C), were calculated according to the following equations [17]:

$$T_{\text{dp}} = \sqrt[3]{\frac{H}{100} \times (112 + 0.9T)} + 0.1T - 112 \quad (2)$$

$$D = T_{\text{dp}} - 4 \quad (3)$$

where the effective dew-formation temperatures consisted of  $D_v$ ,  $D_o$ , and  $D_r$ .  $H$  (%) was the relative humidity in the air, such as  $H_v$ ,  $H_o$ ,  $H_r$ .  $T$  (°C) was the moist (or humid) air temperature, such as  $T_v$ ,  $T_o$ , and  $T_r$ . The efficiency of water condensation depended on the difference between the values of  $D$  and the cooling plate temperature which was equivalent to the cooling water temperature ( $T_c$ ), preferably  $T_c \leq D$ .

$$R = \left( \frac{C_f - C_p}{C_f} \right) \times 100 \quad (4)$$

where  $C_f$  and  $C_p$  were the salt concentrations in the feed and permeate water, respectively.

### C. Feed solution

Ultrapure water was used as the feed solution for comparison of the experimental set-up between CVMD and RVMD. For the salt rejection performance, the ultrapure water was added to NaCl as synthetic solutions for various concentrations such as 1.5, 5, 15, and 30 g/L.

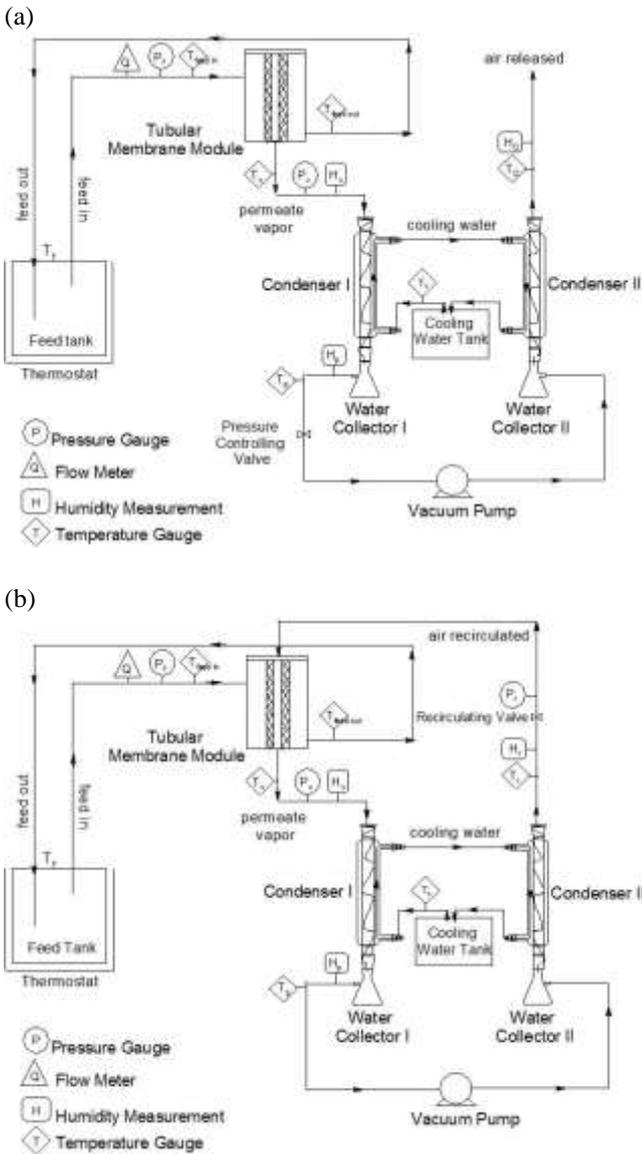


Fig. 1. Schematic diagrams of VMD experimental set-up: (a) CVMD system; (b) RVMD system.

### III. RESULT AND DISCUSSION

#### A. Comparison between CVMD and RVMD

In this case, the condition of the feed water was controlled at a feed temperature ( $T_f$ ) of 50°C and a feed water pressure ( $P_f$ ) of +10 kPa, while the condition of the cooling water temperatures ( $T_c$ ) and the vacuum pressures ( $P_v$ ) were varied in range from 10 to 30°C and -10 to -50 kPa, respectively.

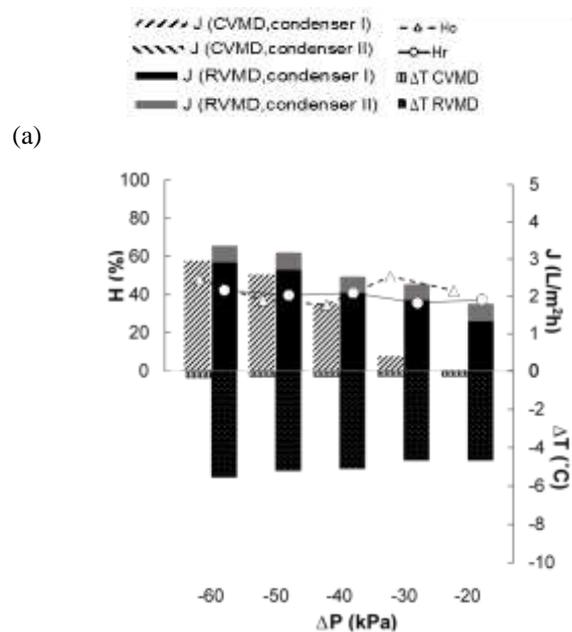
#### B. Humid-air recirculation

Figure 2 illustrates that the values of permeate flux ( $J$ ) obtained by Eq.(1) in RVMD exceeded the values from CVMD for all  $T_c$  conditions. Clearly, increases in permeate fluxes were accompanied by expanding transmembrane pressures ( $\Delta P = P_v - P_f$ ) regarding a fixed vapor pressure for a constant  $T_f$  (50°C). The feed water had evaporated through the hydrophobic membrane pores. The relative humidities of the permeate vapor ( $H_v$ ) in both systems were similarly observed as over 95%, which was the maximum limit of the hygrometer. In the CVMD system, the permeate vapor was condensed and released into the atmosphere even though it

still retained some humidity ( $H_o$ ). On the other hand, the RVMD system recirculated humidity ( $H_r$ ) after condensation back into the membrane module. This recirculated humid air served as an endothermic substance to reduce the temperature ( $T_v$ ) inside the lumen of the tubular membrane, resulting in a larger value of transmembrane temperature ( $\Delta T = T_v - T_f$ ). These  $\Delta T$  values of RVMD were improved further in accordance with higher values of remained  $H_r$  after condensation, with changing  $T_c$  conditions from 10 to 30°C in Fig.2(a) to (c), respectively. Consequently, the values of  $\Delta T$  in RVMD were significantly larger than those of CVMD due to the recirculation of humid air rather than the differences between  $T_f$  and  $T_c$ . The enhancement of  $\Delta T$  eventually provided a higher water productivity, especially in the cases of lower  $\Delta P$ .

#### C. Double stage condensation

Figure 2 also shows that the CVMD system could obtain water production only in condenser I, in contrast to the results from RVMD. The effects of the air flow rate on the permeate flux were then examined. Since the CVMD system was a dead-end mode, the air flow rate ( $A_o$ ) was extremely low regardless of the change in  $\Delta P$  [Fig.3]. The water condensation in CVMD was simply completed within the first condenser due to a sufficiently long detention time, while the RVMD system had much higher air flow rates. The recirculated air flow rates ( $A_r$ ) inversely varied with  $\Delta P$ , as shown in Fig.3. Because the high value of  $A_r$  shortened the detention time within one condenser, the dew formation was unable to be accomplished without an extra condenser. Subsequently, the condenser II guaranteed extended condensation to generate more water. The advantages of the double-stage condenser could be clearly seen in the cases of higher  $T_c$  (20 and 30°C) which remained relatively high  $H_r$ . In addition, changes in the effective dew-formation temperatures before ( $D_v$ ) and after ( $D_o$ ,  $D_r$ ) condensation, shown in Fig.4, confirmed that the double-stage condenser could effectively remove moisture from the permeate vapors as much as possible, since the values of  $D_o$  and  $D_r$  had come close to the values of  $T_c$ .



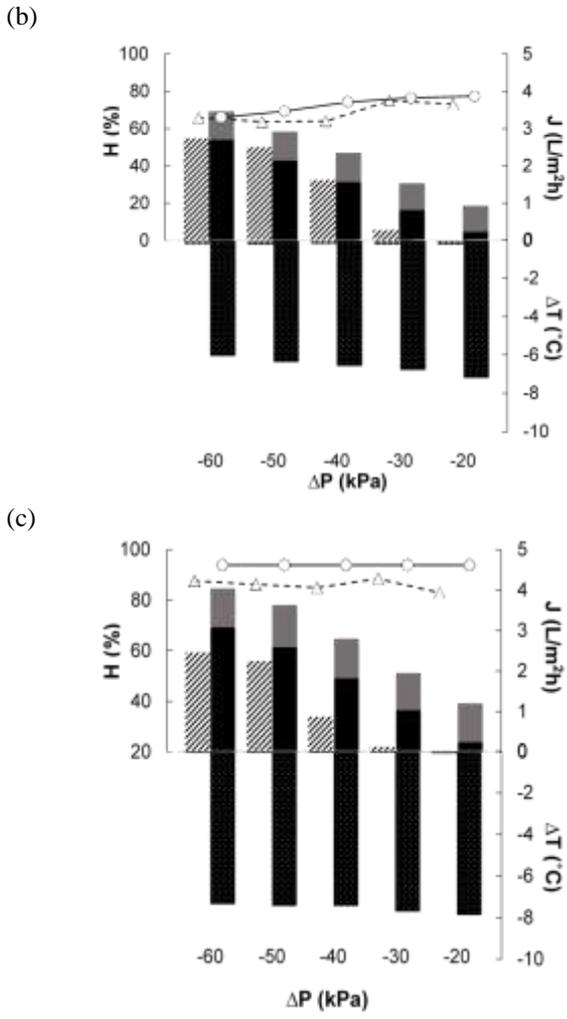


Fig.2. Comparison of CVMD and RVMD systems at feed temperature ( $T_f$ ) 50  $^{\circ}\text{C}$  and feed pressure ( $P_f$ ) +10kPa with different cooling water temperature ( $T_c$ ) at (a) 10 $^{\circ}\text{C}$ , (b) 20 $^{\circ}\text{C}$  and (c) 30 $^{\circ}\text{C}$ .

**D. Dew formation**

Figure 4 demonstrates that the values of  $D_v$  calculated by (2) - (3) were similarly high under all conditions, depending on the relatively high values of  $T_v$  (41~52 $^{\circ}\text{C}$ ) and  $H_v$  ( $\geq 95\%$ ). In Fig.4(c), the values of  $D_{v,RVMD}$  differed markedly from those of  $D_{v,CVMD}$ , due to the effects of humid-air recirculation that significantly caused the reduction of  $T_v$  inside the lumen membrane. After the double-stage condensation, it was found that the values of  $D_o$  were always lower than the  $D_v$  values, because the  $H_o$  moving slowly through the condenser was extremely condensed in the CVMD system, while the RVMD forced the  $H_v$  to pass through the condenser swiftly.

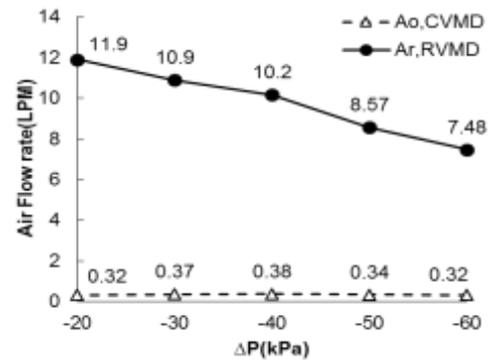


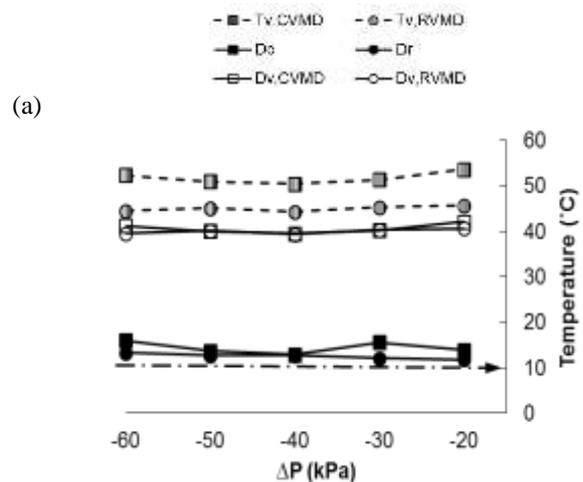
Fig.3. Effect of operating transmembrane pressure ( $\Delta P$ ) on flow rate of released air ( $A_o$ ) in CVMD and recirculated air ( $A_r$ ) in RVMD.

**Water productivity**

Figure 5 conclusively describes the variation of  $\Delta P$  on permeate fluxes ( $J$ ) under different  $T_c$  conditions with the stable  $T_f$  at 50 $^{\circ}\text{C}$ . From the results, the higher values of  $\Delta P$  significantly provided more values of  $J$ , since the evaporation rate of the feed water related to the values of  $\Delta P$  in the membrane module. Furthermore, the larger  $\Delta T$  also enhanced the driving force of vapor transport across the membrane. In the CVMD system,  $J$  increased with decreasing  $T_c$ . On the other hand,  $J$  in the RVMD system did not reveal the same tendencies, because the lower  $T_c$  would unfortunately not result in the larger  $\Delta T$ , but the  $\Delta T$  depended mainly on  $H_r$ , as discussed in Fig.2. In these cases, the  $\Delta P$  and  $\Delta T$  simultaneously enhanced the  $J$ , especially in the higher  $T_c$  conditions. The maximum flux of 4.04  $\text{L}/\text{m}^2\text{h}$  was then found in RVMD at  $\Delta P$  -60 kPa and  $T_c$  30 $^{\circ}\text{C}$ , while the CVMD system gave the maximum flux of 2.89  $\text{L}/\text{m}^2\text{h}$  at  $\Delta P$  -60 kPa and  $T_c$  10 $^{\circ}\text{C}$ . Consequently, the modified configuration using humid-aid recirculation coupled with double-stage condensation could significantly enhance the water productivity for vacuum membrane distillation.

**Performance of RVMD**

To obtain the optimum conditions of RVMD for various feed water temperatures ( $T_f$ ), experiments were conducted with different  $T_f$  (30 to 50 $^{\circ}\text{C}$ ),  $T_c$  (10 to 30 $^{\circ}\text{C}$ ) and  $\Delta P$  (-20 to -60 kPa).



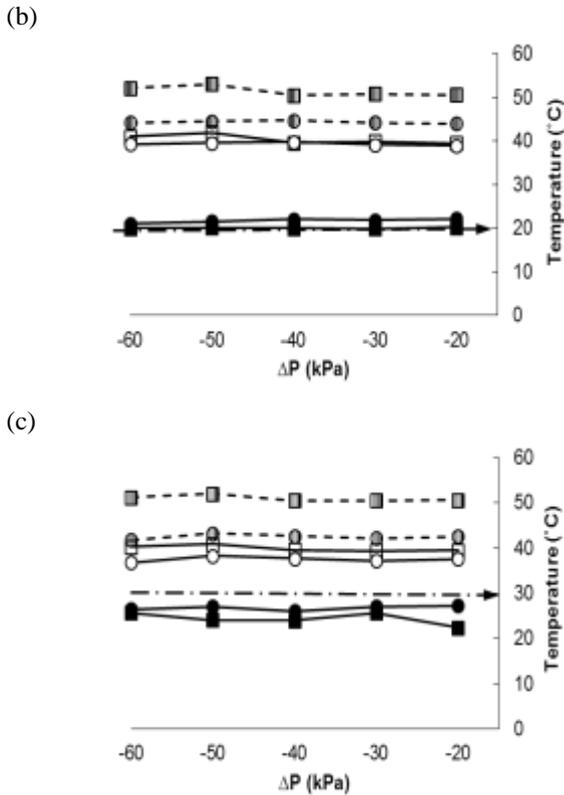


Fig.4. Comparison of temperatures before and after condensation with different cooling temperature ( $T_c$ ) at (a) 10°C, (b) 20°C and (c) 30°C.

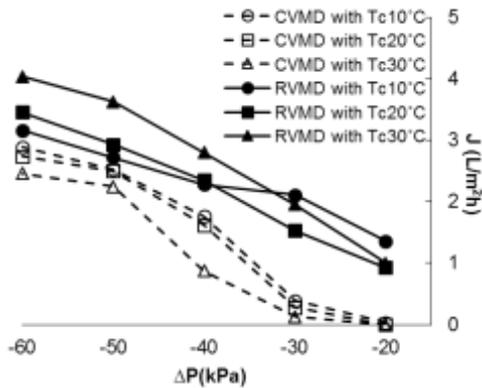


Fig.5. Effects of different transmembrane pressure ( $\Delta P$ ) at constant feed temperature ( $T_f$ ) 50°C on permeate flux ( $J$ ) with various cooling water temperature ( $T_c$ ).

**Effect of vacuum pressure**

Figure 6(a)-(c) demonstrates that the permeate fluxes ( $J$ ) were effectively carried out by influences of  $\Delta P$ . It could be seen that the increases in  $\Delta P$  induced the enhancement of permeate fluxes. This was attributed to the change in the feed water evaporation rate. Furthermore, the increases of  $T_f$  from 30 to 50°C at the same  $\Delta P$  could produce more values of  $J$ , since the water vapor pressures were raised simultaneously with increasing  $T_f$ . In other words, the higher  $T_f$  was preferable for the phase transformation of water molecules from liquid to vapor.

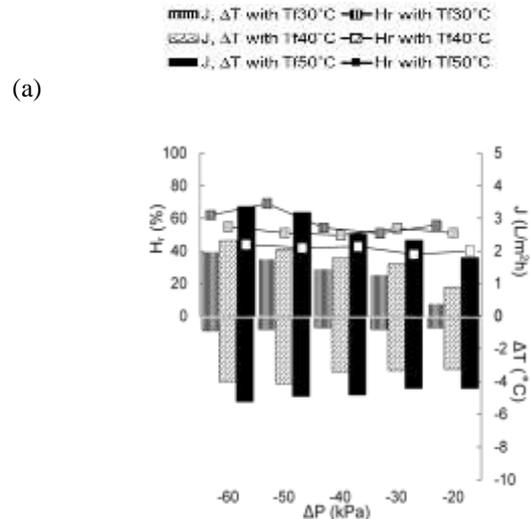
**Effect of transmembrane temperature**

The changes in transmembrane temperature ( $\Delta T = T_v - T_f$ ) were enlarged with increasing  $T_f$  and  $T_c$  due to remaining humidity of recirculated air ( $H_r$ ). The expanded  $\Delta T$  were

more significant in the cases of smaller  $\Delta P$  which related inversely to the recirculation rates [Fig.3], especially when the values of  $H_r$  were over 60%, as shown in Fig.6(b)-(c). The more humid air could perform as the effective endothermic substance to reduce  $T_v$ . After the  $\Delta T$  were gained, penetration of vapor across the membrane could be effectively induced due to a higher transmembrane driving force. Moreover, the higher recirculation rates at lower  $\Delta P$  could also sweep the bulk of the permeate vapor from the lumen of membrane module to the condensers. Consequently, the water productivity of RVMD was successfully improved due to humid-air recirculation instead of  $T_c$  reduction.

**Effect of salt concentration**

A synthetic salt solution was prepared by adding NaCl to the ultrapure water. The feed concentration ( $C_f$ ) varying from 0 to 30 g/L were operated with the RVMD system [Fig.1(b)] in the most preferable conditions of feed temperature ( $T_f$ ) 50°C, cooling temperature ( $T_c$ ) 30°C and transmembrane pressure ( $\Delta P$ ) -60 kPa. At the steady state, the effects of the feed concentrations on the permeate fluxes and salt rejections are examined and described in Fig.7. The permeate fluxes gradually decreased against increases of feed concentration. These declining phenomena could be explained by vapor pressure reduction in the salt-concentrated solution in accordance with a low evaporation rate [18-19]. For example in case of permeate flux of 30 g/L NaCl solution was 2.33 L/m<sup>2</sup>h that lower than ultrapure water (4.04L/m<sup>2</sup>h) due to the decrease of vapor pressure from 12.33kPa to 12.20kPa and other reason of concentration polarization (CP). CP is a cause of flux reduction in other membrane process [20] by the particles of solutes cannot pass through the membrane that caused to accumulate on surface membrane. Thus, the formation of a reversible fouling layer was actually prevented by a cross-flow velocity [21]. In this study the cross-flow velocity as  $5 \times 10^{-3}$  m/s which not enough to decay the surface membrane particle. However, the efficiency of salt rejections ( $R$ ) calculated by (4) remained at around 99.4 to 99.5% consistently, regardless of  $C_f$  changes. Consequently, the RVMD could guarantee an excellent performance with a wide range of  $C_f$  up to 30 g/L.



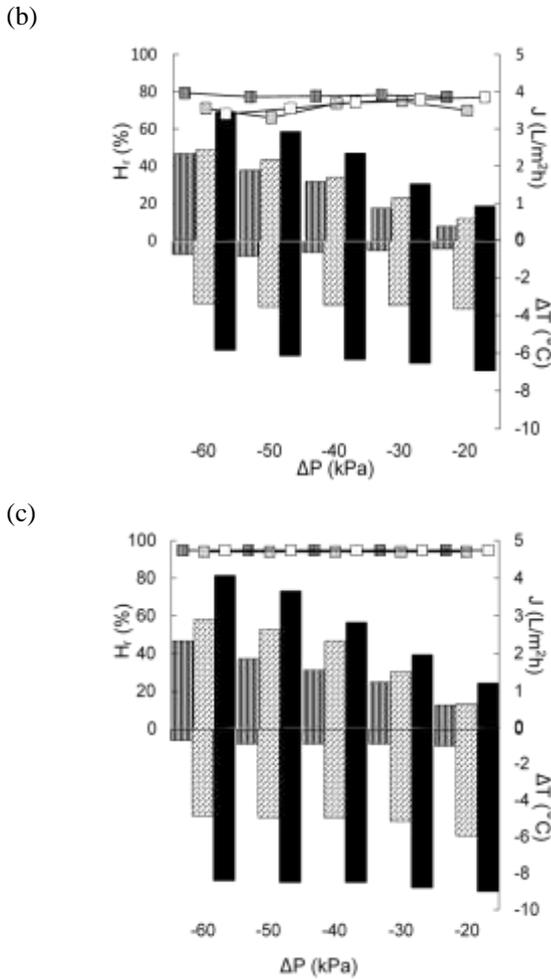


Fig.6. Performances of permeate flux ( $J$ ) in RVMD with different cooling temperature ( $T_c$ ) at (a)  $10^\circ\text{C}$ ; (b)  $20^\circ\text{C}$ ; (c)  $30^\circ\text{C}$ .

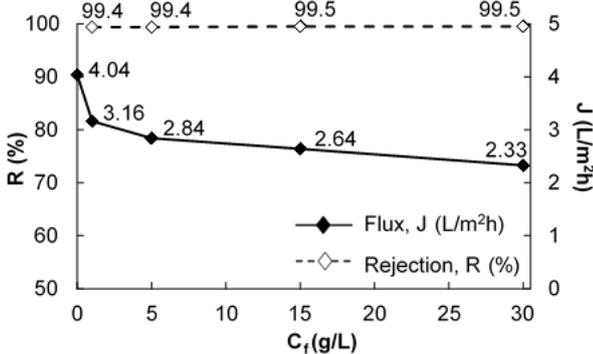


Fig.7. Effects of feed concentration ( $C_f$ ) on permeate flux ( $J$ ) and salt rejection ( $R$ ).

#### IV. CONCLUSION

This work proposed the modified configuration of vacuum membrane distillation comprising humid-air recirculation and double-stage condensation. The recirculation vacuum membrane distillation (RVMD) indicated that water productivity could be significantly improved from the conventional vacuum membrane distillation (CVMD). The humid-air recirculation was able to raise transmembrane temperature ( $\Delta T$ ) across the membrane without applying low temperature in the condenser. The maximum pure water flux of RVMD was reached at  $4.04 \text{ L}/\text{m}^2/\text{h}$  in the condition of feed water temperature ( $T_f$ )  $50^\circ\text{C}$ , cooling water temperature ( $T_c$ )  $30^\circ\text{C}$ , and transmembrane pressure ( $\Delta P$ )  $-60 \text{ kPa}$ , while the

maximum value of CVMD was found at  $2.89 \text{ L}/\text{m}^2/\text{h}$  with  $T_f$   $50^\circ\text{C}$ ,  $T_c$   $10^\circ\text{C}$ , and  $\Delta P$   $-60 \text{ kPa}$ . The double-stage condensers had assured the completion for dew formation in CVMD as well as the extension for water condensation in RVMD. However, the remaining humidity after condensation was impossible to be emptied from the condensed air since the effective dew-formation temperatures ( $D_o$ ,  $D_r$ ) had already come close to the  $T_c$ . In addition, the permeate fluxes ( $J$ ) were diminished from  $4.04$  to  $2.33 \text{ L}/\text{m}^2/\text{h}$  with increasing the feed concentrations ( $C_f$ ) from  $0$  to  $30 \text{ g}/\text{L}$  while the change in salt removal efficiencies ( $\sim 99.4$  to  $99.5\%$ ) could not be observed.

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