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The Effect of Bulk Feed Temperature for Breaking of Butyric acid – Water Azeotrope using Air Gap Membrane Distillation

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ABSTRACT

Air gap membrane distillation is a thermally operated membrane separation process in which only the vapour phase can pass through a hydrophobic membrane, leaving behind the liquid phase. The driving force in this method is the temperature gradient across the hydrophobic microporous membrane, which creates the difference in vapour pressure and the separation is possible due to differences in their diffusivity rates in the air. Experimentally, I investigated the feasibility of an air gap membrane distillation module with a hydrophobic PTFE membrane for the separation of butyric acid-water azeotrope. Experiments were conducted to determine the effect of bulk feed temperature, feed flow rate, air gap width, cooling water temperature, and cooling water flow rate on permeate flux, selectivity of butyric acid, and concentration of butyric acid in permeate and retentate. The permeate flux increased from 6.71 kg/m^{2}hr to 7.05 kg/m^{2}hr as the bulk feed temperature was increased from 45 °C to 65 °C and the air gap width was increased from 3mm to 7mm. This is due to a rise in total vapour pressure as the temperature rises, which increases permeate flux. The selectivity of butyric acid decreased from 0.49 to 0.38 when the bulk feed temperature was increased from 45 °C to 85 °C and the air gap width was increased from 3mm to 7mm. In addition, as the bulk feed temperature was increased from 45 °C to 65 °C and the air gap width was increased from 3mm to 7mm, the concentration in the permeate decreased while the concentration in the retentate increased. This is primarily due to the membrane's decreased selectivity towards butyric acid. During the experimental investigation, all other parameters, including feed flow rate and cooling water flow rate, were held constant at 2 L/min and 8 °C, respectively. This demonstrates that azeotropes break in both permeate and retentate.

***Keywords-** Air gap membrane distillation, PTFE membrane, Butyric acid-water Azeotrope, effect of bulk feed temperature, Titration method*

1. INTRODUCTION

Air gap membrane distillation is a thermally operated membrane separation process. Only the vapour phase is permitted to pass through a hydrophobic membrane in this method. The difference in vapour pressure is the driving force behind this method. The gradient in temperature is generated across the hydrophobic membrane (Kimura et al. 1987). Four membrane distillation techniques exist: direct contact membrane distillation (DCMD), vacuum membrane distillation (VMD), air gap membrane distillation (AGMD), and swiping gas membrane distillation (SGMD). The configuration of the distillate channel distinguishes each technique from the others. In this investigation, the applicability of the air gap membrane distillation (AGMD) technology for breaking the butyric acid-water azeotrope (18.4% butyric acid) was investigated.

For the breaking of azeotrope, a number of techniques have been used, including azeotropic distillation (Widagdo and Seider 1996, Mortaheb and Kosuge 2004), extractive distillation (Lek-utaiwan et al. 2010, Lei et al. 2005), capillary distillation, adsorptive distillation, pervaporation, and diffusion distillation (D. Fullarton and E. U.

Schlünder, 1986). These techniques have drawbacks such as high energy requirements, limited entrainer possibilities, and recovery of the key component. To overcome these restrictions, the AGMD process is used in place of the conventional method for azeotrope breaking (C. H. Gooding and F. J. Bahouth 1985, Udriot et al. 1994, Banat 1999 a, b, c, Khayet et al. 2011, and Kalla et al. 2019). The diffusivity of various components in air is important in breaking azeotropes. Experiments were conducted to determine the effect of operating parameters, including feed flow rate, bulk feed temperature (45 °C – 65 °C), air gap width (3 mm – 7 mm), cooling water flow rate, and cooling water temperature, on total permeate flux, butyric acid selectivity, and the concentration of butyric acid in permeate and retentate.

2. LITERATURE REVIEW

Propionic acid-H_2O (Udriot et al. 1994), HCl-H_2O (Udriot et al. 1994), formic acid-H_2O (Banat 1999 b, c), and HCl-H_2O (Kalla et al. 2019) azeotropic mixtures have been examined for the breaking of the azeotropic point via air gap membrane distillation. Udriot et al. (1994) were the first to investigate azeotropic mixture separation in air gap membrane distillation for the systems of hydrochloric acid/water and propionic acid/water. Additionally, he determined that the pazeotropic point of the propionic acid/water system has vanished entirely. In their investigation, neither temperature nor concentration polarisation were taken into account.

Banat et al. (1999 b, c) examined the performance of air gap membrane distillation in breaking formic acid-water azeotropes using the Stefan-Maxwell mass transfer mathematical model for the multicomponent system and compared Fickian and Stefan-Maxwell based mathematical models. The authors found that the Stefan-Maxwell model matches the experimental data better than the Fickian model based on binary molecular diffusion. Banat et al. (1999 d) investigated the impact of inert gases, namely helium, air, and sulphur hexafluoride, on the azeotropic formic acid/water mixture. The selectivity of formic acid was observed to be 0.96, 0.90, and 0.85 to 0.86 when using helium, air, and sulphur hexafluoride, respectively. Similar to helium, air and sulphur hexafluoride had the highest AGMD permeate flux. It was discovered that sulphur hexafluoride, a heavier inert gas, assists in the removal of the azeotropic point more than lighter gases like air and helium.

Kimura and Nakao (1987) studied the separation of systems such as HNO_3-water, HCl-water, and formic acid-water at various concentrations without taking into account the possible effect of AGMD on the azeotropic point of these systems. It is remarkable that very few experimental studies have been conducted in this fruitful area of study. Kalla et al. (2019) studied the effect of various operating parameters on total permeate flux, selectivity, and HCl concentration in permeate and retentate, including feed concentration, feed temperature, feed flow rate, air gap width, cooling water temperature, and cooling water flow rate. This study investigated the effect of feed temperature, air gap width, and operating duration on the breaking of the HCl-water azeotropic point.

3. METHODOLOGY

Figure 1 depicts a schematic diagram of the AGMD setup that was employed for the experiment. The feed section, the air gap section, and the cooling section make up the three sections of the experimental setup. Under different operating conditions, the separation of butyric acid/water azeotropic mixtures was investigated using the AGMD approach. The cooling plate is fitted between the air gap and cooling sections, and the hydrophobic membrane is installed between the feed and air gap sections. The feed solution with the desired temperature was circulated to the feed portion using the pump. The cooling compartment also received pumping and circulation of the cooling water. Digital thermocouples were used to measure the feed solution temperature and cooling water temperature, while rotameters were employed to manage the feed and cooling water flow rates. After passing through the membrane and air gap and collecting on the cooling plate, butyric acid and water vapours liquefied. The permeate solution was then transferred into the receiver.

At 18.4 weight % of butyric acid, the butyric acid forms an azeotrope with water. To find out whether the azeotrope broke or not, the butyric acid concentration in the permeate and retentate was tested. The butyric acid concentration in permeate and retentate must be either lower or higher than the butyric acid-water azeotropic mixture concentration level, which was determined by the acid-base titration method.

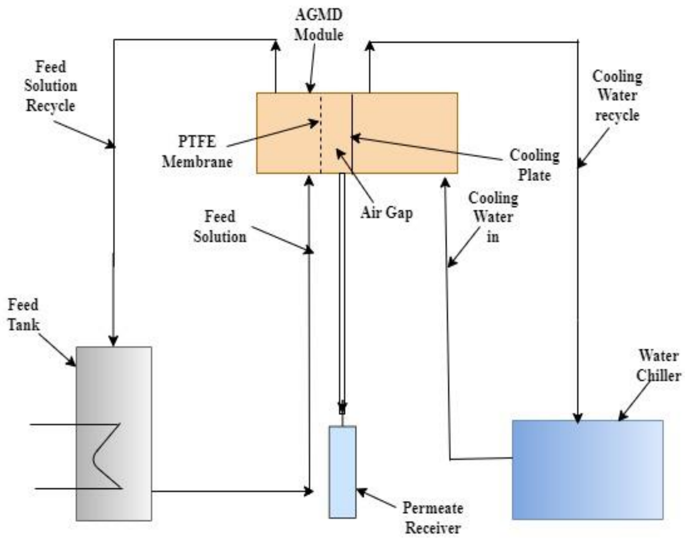

Figure 1: Diagram of Experimental Setup of Air Gap Membrane Distillation

4. RESULT & DISCUSSION

4.1 Diffusivity of Butyric Acid and Water in Air with Temperature

Figure 3 demonstrates that as feed bulk temperature increased from 25 °C to 80 °C, the air diffusivity of butyric acid and water increased linearly. Figure 2 demonstrates that the diffusivity of water in the air is greater than that of butyric acid in the air. The following equation was derived from a combination of kinetic theory and corresponding state arguments (Transport Phenomena, Second Edition, by R. Byron Bird) in order to estimate the diffusivity of water and butyric acid in low-pressure air. Figure 2 indicates the butyric acid calibration cunve/

$$\frac{PD_{AB}}{(P_{CA}P_{CB})^{\frac{1}{3}}(T_{CA}T_{CB})^{\frac{5}{12}}\left(\frac{1}{M_A}+\frac{1}{M_B}\right)^{\frac{1}{2}}} = a\left(\frac{T}{\sqrt{(T_{CA}T_{CB})}}\right)^{b}$$

(1)

For pair consisting of water and non-polar gas

$a = 3.640\ X(10)^{-4}$

$b = 2.334$

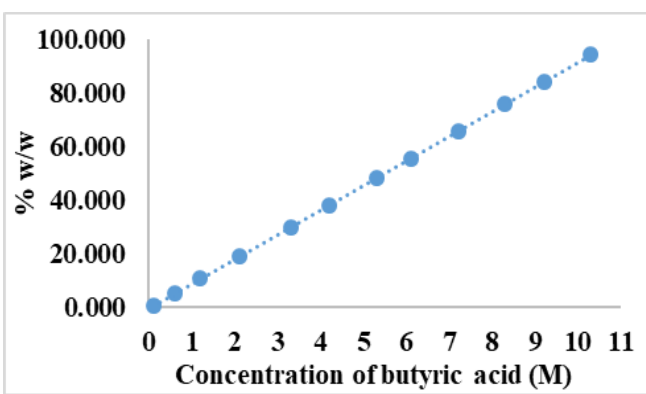

Fig 2: Butyric Acid Calibration Curve

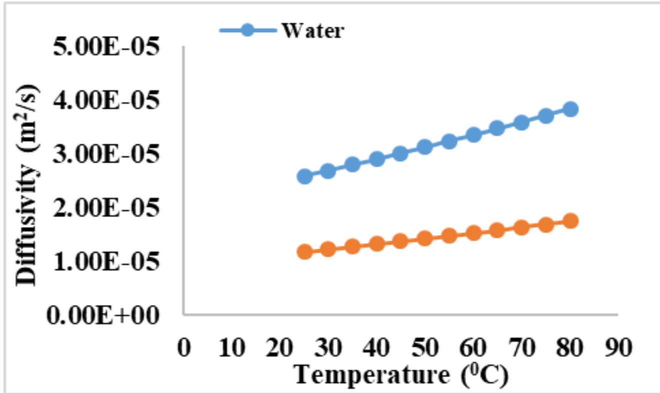

Fig 3: The effect of bulk feed temperature on water and butyric acid diffusivity

4.2 Vapour Pressure of Water and Butyric Acid with Temperature

Figure 4 demonstrates that as the temperature increases from 0 ^{0}C to 80 ^{0}C, the vapour pressure of both water and butyric acid increases exponentially. Figure 6 also demonstrates that the vapour pressure of water is greater than that of butyric acid. This is primarily due to the fact that butyric acid has a lower boiling point than water. The vapour pressures of water and butyric acid were calculated using the Antoine equation.

$$log_{10}(P) = A - \frac{B}{C+T} \tag{2}$$

P = Vapour pressure (mmHg)

T = Temperature (°K)

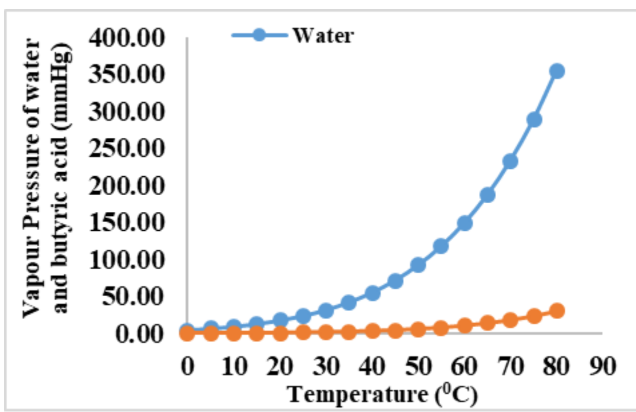

Fig 4: The effect of bulk feed temperature on water and butyric acid vapour pressures

4.3 Effect of Bulk Feed Temperature on Permeate Flux

Figure 5 demonstrates the effect of feed temperature (45 °C to 65 °C) on total permeate flux for various air gap widths (3mm to 7mm), while all other parameters such as cooling water temperature (8 °C), feed flow rate 2 L/min, and cooling water flow rate 2 L/min are held constant. From the figure 5 it was cleared that total permeate flux increased exponentially when feed temperature increased from 45 °C to 65 °C. This is mainly due to the fact that on increasing the feed temperature leads an increase in total vapour pressure, resulting in an increase in permeate flux.

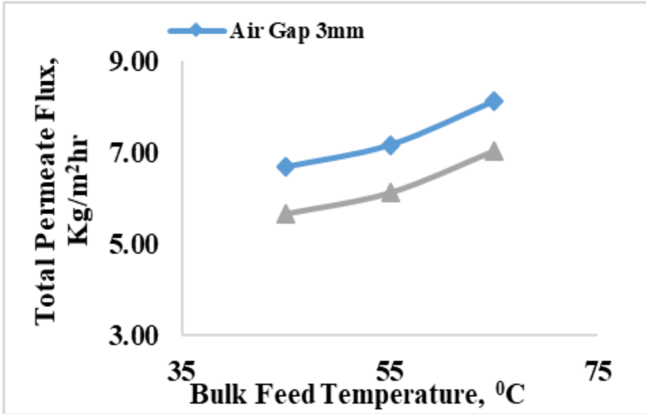

Figure 5: Effect of Bulk Feed Temperature on Total Permeate Flux for Different Air Gap Widths

(Cooling water temperature =8 ºC, Feed flow rate = 2 L/min, Cooling water flow rate = 2 L/min)

4.4 Effect of Bulk Feed Temperature on Selectivity

Figure 6 depicts the change in butyric acid selectivity as a function of feed temperature for various air gap widths. Figure 6 demonstrates that as the feed temperature increases from 45 ºC to 65 ºC, the selectivity of the butyric acid reduces. This is mostly due to the membrane's lower selectivity for butyric acid vapours.

Selectivity determines the degree of separation of any component in membrane distillation. The term "selectivity" define as below:

$$\alpha_{BA} = \left(\frac{y_1}{1-y_1}\right) / \left(\frac{x_1}{1-x_1}\right) \qquad (3)$$

In this equation, y_1 and x_1 represent the mole fractions of butyric acid in permeate and feed, respectively. Figure 6 shows that the membrane selectivity of butyric acid in permeate was determined to be less than unity, which demonstrates that retentate has a higher concentration of butyric acid than permeate.

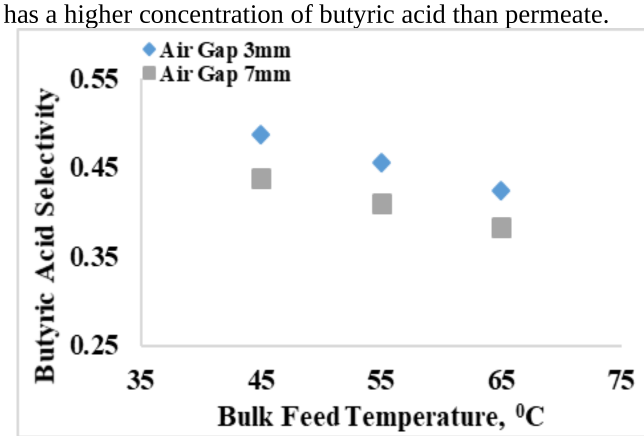

Figure 6: Effect of Temperature on Butyric Acid Selectivity for Different Air Gap Widths

(Cooling water temperature =8 ºC, Feed flow rate = 2 L/min, Cooling water flow rate = 2 L/min)

4.5 Effect of Bulk Feed Temperature on Concentration of Butyric Acid in Permeate and Retentate

Figures 7 and 8 show that the concentration of butyric acid in permeate decreases while the concentration of butyric acid in retentate increases when the bulk feed temperature increases. This is mostly due to the membrane's lower selectivity for butyric acid vapours.

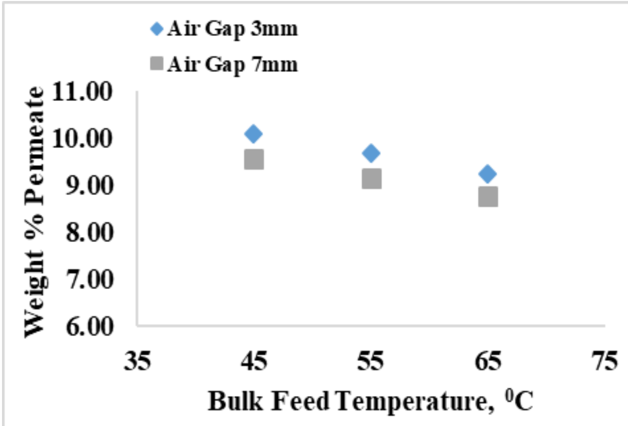

Figure 7: Effect of feed temperature on permeate Butyric acid concentration for different air gap widths (Cooling water temperature =8 ºC, Feed flow rate = 2 L/min, Cooling water flow rate = 2 L/min)

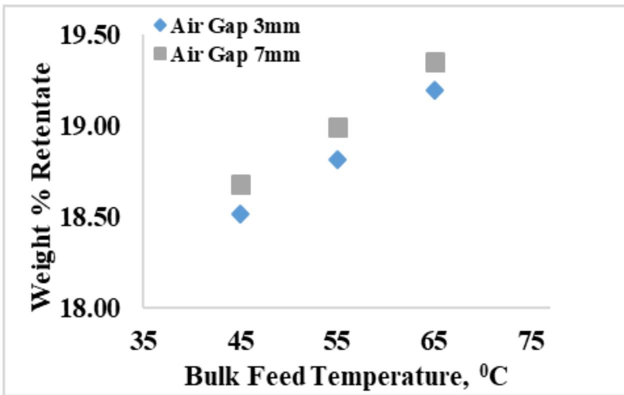

Figure 8: Effect of feed temperature on retentate Butyric acid concentration for different air gap widths (Cooling water temperature =8 ºC, Feed flow rate = 2 L/min, Cooling water flow rate = 2 L/min)

5. CONCLUSION

In an air gap membrane distillation with a PTFE membrane, the breaking of the butyric acid/Water azeotrope mixture was investigated. The following conclusions can be taken from experimental results.

- It is cleared from kinetic theory and corresponding state arguments that increasing the feed bulk temperature from 45 ^{0}C to 80 ^{0}C increases the diffusivity of butyric acid and water in the air linearly. It was also found that the diffusivity of water in the air is greater than that of butyric acid in the air. According to Antoine's equation, the vapour pressure of water and butyric acid increases exponentially as the temperature rises from 45 ^{0}C to 80 ^{0}C. It was also observed that the vapour pressure of water is greater than that of butyric acid.
- The experimental results show that at 3mm air gap width, increasing bulk feed temperature from 45 ^{0}C to 65 ^{0}C increased total permeate flux from 6.71 to 8.13 kg/m^{2}h, concentration of butyric acid in the permeate decreased from 10.10 weight% to 9.25 weight%, while in the retentate it increased from 18.52 weight% to 19.19 weight%, and butyric acid selectivity decreased from 0.49 to 0.42.
- Also, it was cleared that at 7mm air gap width, increasing bulk feed temperature from 45 ^{0}C to 65 ^{0}C increased total permeate flux from 5.66 to 7.05 kg/m^{2}h, concentration of butyric acid in the permeate decreased from 9.56 weight % to 8.76 weight %, while in the retentate it increased from 18.68 weight % to 19.35 weight %, and butyric acid selectivity decreased from 0.44 to 0.38.

- The selectivity experimental results show that the membrane selectivity of butyric acid in permeate was determined to be less than unity, which demonstrates that retentate has a higher concentration of butyric acid than permeate.
- Experiment results indicate that the butyric acid-Water azeotrope breaks in both penetrate and retentate. As a result, it is stated that there is a strong chance of adopting the AGMD approach for azeotrope breaking.

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