The effect Cooling Water Temperature in Air Gap Membrane Distillation for Breaking of Butyric acid – Water Azeotrope

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Abstract – *Air gap membrane distillation is a thermal membrane separation method. In this method, the vapour phase is let through a hydrophobic membrane. The vapour pressure difference is created due to the temperature gradient across the hydrophobic microporous membrane, which is the driving force. I have investigated experimentally the feasibility of Air Gap Membrane Distillation module with PTFE hydrophobic membrane for the breaking of butyric acid-water azeotrope. Because of differences in diffusivity rates in the air, separation is possible. The influence of bulk feed temperature on the diffusivity of water and butyric acid, as well as the vapour pressure of butyric acid and water, was examined in this paper. Also, the effect of cooling water temperature (5-20 ^0C) on total permeate flux, selectivity of butyric acid, and the concentration of butyric acid in permeate and retentate for various air gap (3-11mm) were experimentally studied. All other parameters were held constant during the experiment, including the Bulk Feed Temperature of 80 ^oC, Feed Flow Rate of 2 L/min, and Cooling Water Flow Rate of 2 L/min.*

Keywords- *AGMD, PTFE polymeric membrane, Butyric acid-water Azeotrope, influence of cooling water temperature, Titration method*

I. INTRODUCTION

Air gap membrane distillation is a membrane separation process operated thermally. In this method, only vapour phase is permitted through a hydrophobic membrane. The vapour pressure difference is the driving force for this method. The temperature gradient is created across the hydrophobic membrane. (Gore 1982, DrioIi and Wu 1985, Kimura et al. 1987).

Direct contact membrane distillation (DCMD), vacuum membrane distillation (VMD), air gap membrane distillation (AGMD), and swiping gas membrane distillation (SGMD) are the four membrane distillation methods. All the technique differs from one another due to the arrangement in the distillate channel. The applicability of the air gap membrane distillation (AGMD) technology for breaking the butyric acid-water azeotrope (18.4 wt% butyric acid) has been experimentally examined in this work. Several methods such as azeotropic distillation (T. Mahdi et. al 1985, 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) has been carried out for the breaking of azeotrope. The disadvantages of these approaches include high energy requirements, restricted entrainer possibilities, and recovery of the essential component. To overcome these limitations, AGMD process is used as a substitute for the conventional method for the breaking of azeotropes (C. H. Gooding and F. J. Bahouth 1985, Udriot et al. 1994, Banat 1999 a, c, d, Khayet et al. 2011, and Kalla et al. 2019). The

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diffusivity of various components in the air is significant in breaking azeotropes. The effect of operating parameters namely feed flow rate (2 – 6 L/min), bulk feed temperature (40 – 80 ^{0}C), air gap width (3 – 11 mm), cooling water flow rate (1 – 5 L/min), and cooling water temperature (5 – 20 ^{0}C) on total permeate flux, butyric acid selectivity, and the concentration of butyric acid in permeate and retentate were investigated experimentally.

II. LITERATURE REVIEW

Air gap membrane distillation has been examined for propionic acid-H$_2$O (Udriot et al. 1994), HCl-H$_2$O (Udriot et al. 1994), formic acid-H$_2$O (Banat 1999 c, d), and, HCl-H$_2$O (Kalla et al. 2018) azeotropic mixtures for the breaking of the azeotropic point. Udriot et al. (1994) was the first to investigate azeotropic mixture separation in air gap membrane distillation for the hydrochloric acid-water and propionic acid-water systems. He also investigated that the azeotropic point of the propionic acid/water system has completely vanished. Temperature and concentration polarisation were not taken into account in their research.

Banat et al. (1999 b, c) investigated 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 the Fickian and Stefan-Maxwell based mathematical models. The authors discovered that the Stefan-Maxwell model fits the experimental data better than the binary molecular diffusion-based Fickian model. Banat et al. (1999 d) studied the influence of inert gases, specifically helium, air, and sulphur hexafluoride, on the azeotropic formic acid/water mixture. The formic acid selectivity was observed as 0.96, 0.9, and, 0.85-0.86 when using helium, air, and sulphur hexafluoride, respectively. Similarly, helium had the highest AGMD permeate flows, followed by air and sulphur hexafluoride. It was discovered that the heavier inert gas, sulphur hexafluoride, aids in the removal of the azeotropic point more than the lighter ones, such as air and helium.

Kimura and Nakao (1987) investigated the separation of systems such as HNO$_3$-water, HCl-water, and formic acid-water at various concentrations without considering the possible effect of AGMD diminishing these systems' azeotropic point. It is worth noting that very few experimental investigations have been conducted in this beneficial research area. Kalla et al. (2018) investigated the effect of several 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. The influence of feed temperature, air gap width, and operating time on the breaking of the HCl-water azeotropic point was investigated in this work.

III. METHODOLOGY

The AGMD technique was used to explore the separation of Butyric acid/Water azeotropic mixtures under various operating conditions. The schematic diagram of the AGMD setup used for the investigation is shown in Figure 1. The experimental setup is divided into three sections: the feed portion, the air gap section, and the cooling section. The hydrophobic membrane is installed between the feed and air gap sections, while the cooling plate is installed between the air gap and cooling sections. The pump was used to circulate the feed solution of the desired temperature to the feed section. Also, the cooling water was pumped and circulated to the cooling section. The rotameters were used to maintain feed and cooling water flow rates and digital thermocouples were used to measure feed solution temperature and cooling water temperature. Butyric acid and water vapours got liquified on the cooling plate after forwarding through the membrane and air gap. Finally, the permeate solution was taken in the receiver.

The concentration of butyric acid in the permeate and retentate was measured to determine to verify whether the azeotrope broke or not. The butyric acid forms an azeotrope with water at 18.4 weight% butyric acid concentration. So, butyric acid concentration in permeate and retentate must be either lower or greater than the butyric acid-water azeotropic mixture concentration level, which was measured by the acid-base titration method.
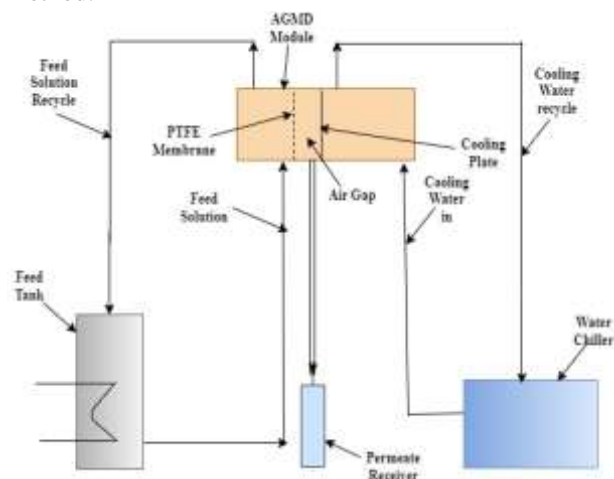

Fig 1: Diagram of Experimental Setup of Air Gap Membrane Distillation

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IV. RESULT & DISCUSSION

Diffusivity of butyric acid and water in air with temperature

Figure 3 shows that on increasing feed bulk temperature from 25 ^{0}C to 80 ^{0}C, the diffusivity of butyric acid and water in the air increased linearly. From figure 2, The diffusivity of water in the air was found to be greater than the diffusivity of butyric acid in the air. The following equation for estimating the diffusivity of water and butyric acid in the air at low pressure has been derived from a combination of kinetic theory and corresponding state arguments (Transport Phenomena by R. Byron Bird, Second Edition)

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

For pair consisting of water and non-polar gas
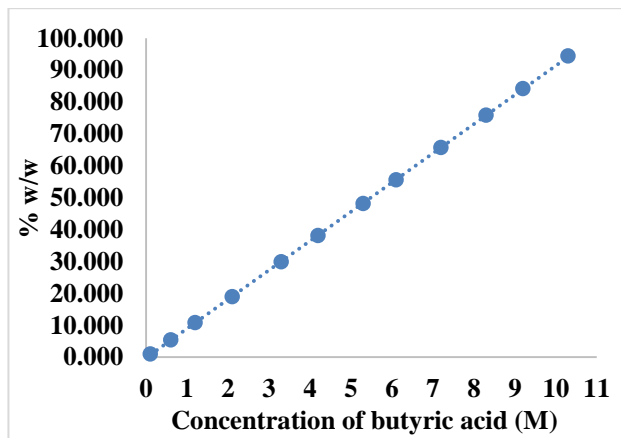$a = 3.640\,X(10)^{-4}$
$b = 2.334$

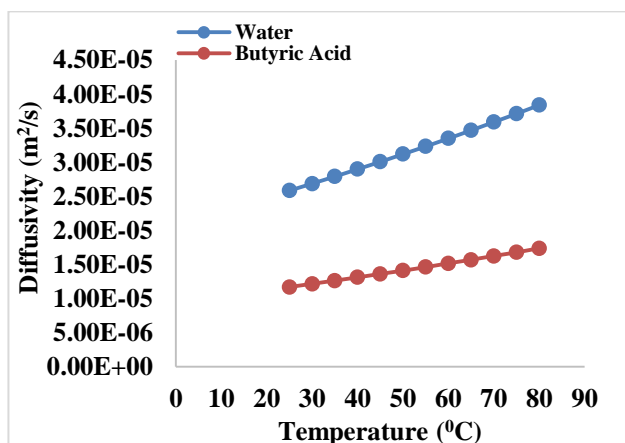*Fig 2: Butyric Acid Calibration Curve*

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

Vapour pressure of water and butyric acid with temperature

Figure 4 indicates that on increasing temperature from 0 ^{0}C to 80 ^{0}C, the vapour pressure of water as well as vapour pressure of butyric acid increases exponentially. Figure 6 also shows that the vapour pressure of water is higher than the vapour pressure of butyric acid. This is mainly due to the fact that the boiling point of butyric acid is less than that of water. The vapour pressure of water as well as butyric acid was calculated by Antoine equation as below.

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

P = Vapour pressure (mmHg)

T = Temperature (^{o}K)
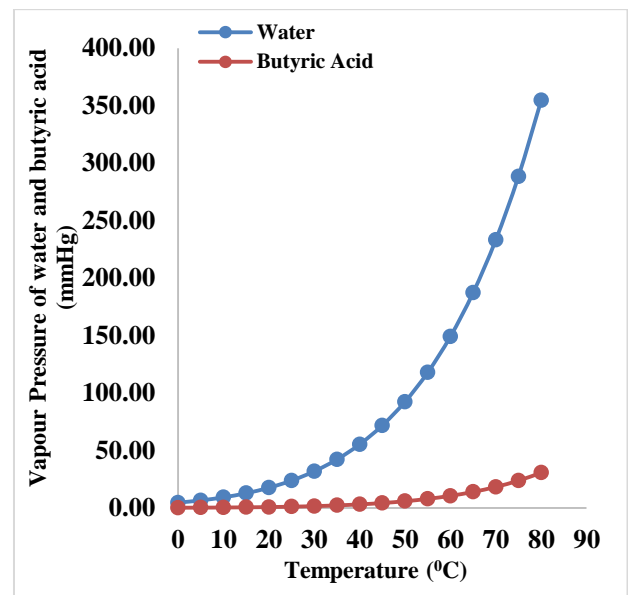
Fig 4: The effect of bulk feed temperature on water and butyric acid vapour pressures

Effect of cooling water temperature

Figures 5 and 6 show the influence of cooling water temperature on permeate flux and selectivity for various air gap widths. The permeate flux was clearly reduced as the cooling water temperature increased from 5 ^{o}C to 20 ^{o}C. This is mostly due to a reduction in the temperature differential across the hydrophobic membrane, which results in a reduction in vapour pressure. Furthermore, an increase in cooling water temperature enhances the selectivity of butyric acid in permeate. Banat et al. 1999 (d) showed a similar result for formic acid-water azeotrope selectivity. Figures 7 and 8 depict the change

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in butyric acid concentration in permeate and retentate with cooling water temperature. It was noted that the butyric acid concentration increases in permeate as well as in the retentate.
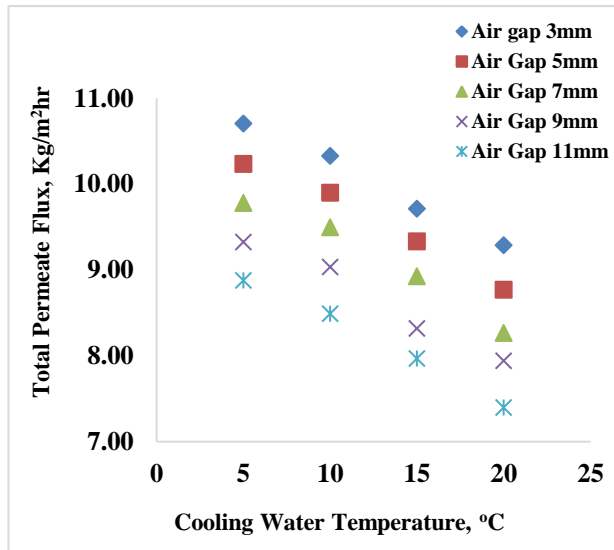

Figure 5: Effect of Cooling Water Temperature on Total Permeate Flux for Various Air Gap Widths

(Feed Temperature = 80 ^{o}C, Feed flow rate = 2L/min, Cooling water flow rate = 1 L/min)
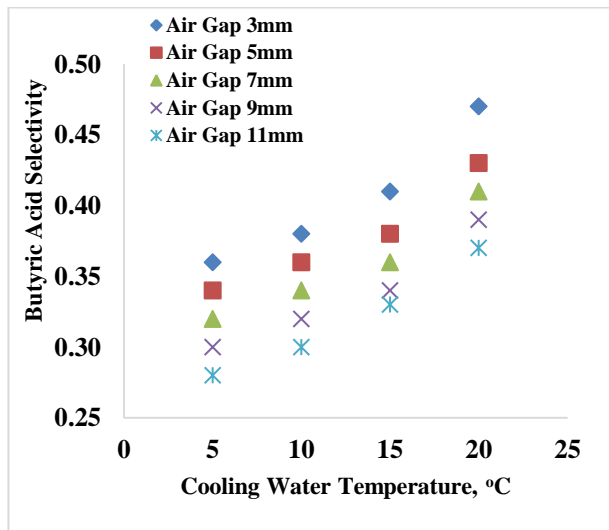

Figure 6: Effect of Cooling Water Temperature on Selectivity for Various Air Gap Widths

(Feed Temperature = 80 ^{o}C, Feed flow rate =2 L/min, Cooling water flow rate = 2 L/min)
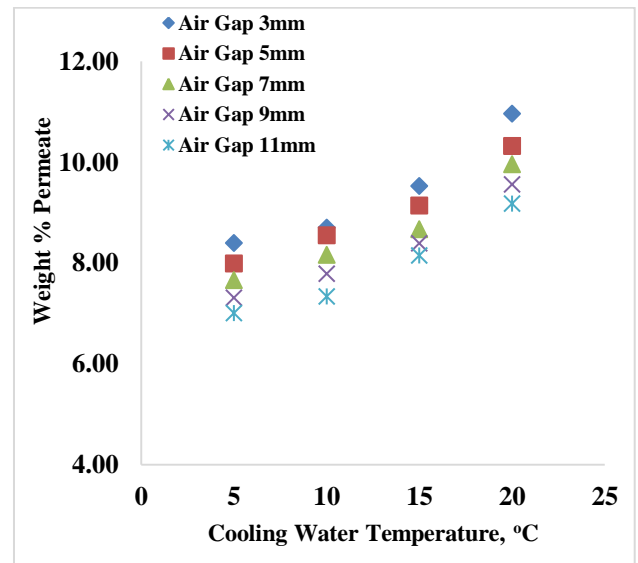

Figure 7: Change in Permeate Butyric acid Concentration for Various Air Gap Widths

(Feed Temperature= 80 ^{o}C, Feed flow rate =2 L/min, Cooling water flow rate = 2 L/min)
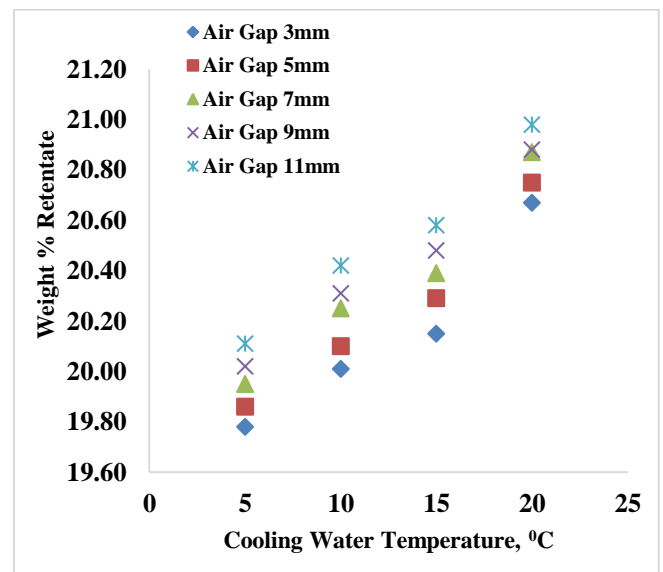

Figure 8: Change in Retentate Butyric acid Concentration for Various Air Gap Widths

(Feed Temperature= 80 ^{o}C, Feed flow rate =2 L/min, Cooling water flow rate = 2 L/min)

V. CONCLUSION

The breaking of the butyric acid/Water azeotrope mixture was studied in an air gap membrane distillation with a PTFE membrane. Based on experimental results, the following conclusions can be drawn.

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- From kinetic theory and corresponding state arguments, it can be cleared that on increasing feed bulk temperature from 25 ^{0}C to 80 ^{0}C, the diffusivity of butyric acid as well as water in the air increases linearly. Also, it was observed that the diffusivity of water in the air is higher than the diffusivity of butyric acid in the air. From Antoine's equation, it was clear that on increasing temperature from 25 ^{0}C to 80 ^{0}C, the vapour pressure of water as well as butyric acid increases exponentially. Also, it was observed that the vapour pressure of water is higher than the vapour pressure of butyric acid.

- The experimental results cleared that at 3mm air gap width, the total permeate flux decreased from 10.71 to 9.29 kg/m^{2}h and the selectivity of butyric acid decreased from 0.36 to 0.41 on increasing cooling water temperature from 5 ^{0}C to 20 ^{0}C. Also, on increasing the cooling water temperature from 5 ^{0}C to 20 ^{0}C at 3 mm air gap thickness, the concentration of butyric acid (wt%) in permeate increases from 8.40 to 10.96 whereas the concentration of butyric acid (wt%) in retentate increases from 19.78 to 20.67 The butyric acid concentration in the permeate and the retentate was analysed by acid-base titration.

- The selectivity of butyric acid to the membrane in permeate was found less than one, which shows that a lower concentration of butyric acid in permeate was obtained as compared to permeate.

- The findings of the experiments show that the butyric acid-Water azeotrope breaks in both penetrate and retentate. Therefore, it is concluded that a strong possibility of using the AGMD technique for the breaking of the azeotrope.

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