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Experimental and Theoretical Study of Air Gap Membrane Distillation for Water-Toluene Removal

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ABSTRACT

In this work, Toluene is removed from a Toluene-water solution using the Air gap membrane distillation method. In the membrane module of the AGMD system, a microporous, hydrophobic Poly-tetra-fluoro-ethylene (PTFE) membrane was used. On permeate flux and membrane selectivity, the effect of air gap width, was investigated. On increasing the air gap width from 3 mm to 11 mm, the permeate flux drastically decreased from 4.65 kg/m^2•h to 1.90 kg/m^2•h at 2 lpm of feed flow rate at a constant feed temperature of 60°C. On increasing the air gap width from 3 mm to 11 mm, it was found that the selectivity of Toluene in the permeate increased, which confirms the separation of Toluene via Air Gap Membrane Distillation. At a wavelength of 260 nm, the UV-Vis spectrophotometer is used to measure the Toluene concentration in permeate. The membrane morphology of a hydrophobic PTFE membrane was investigated using FE-SEM after a 150 hour of experiment.

Keyword: AGMD, Toluene (VOCs), Poly-tetra-fluoro-ethylene (PTFE), Selectivity, Membrane Fouling

1. INTRODUCTION

Volatile organic compounds (VOCs) are one of the most dangerous pollutants, endangering both human and environmental health. In photochemical smog reactions, it has been demonstrated that VOCs are crucial precursors for the formation of fine particulate matter (secondary organic aerosols) and ozone. In addition to posing a significant environmental risk, many VOCs are strictly regulated due to their toxic and carcinogenic properties. Numbers of techniques have been applied to remove VOCs including adsorption, incineration, catalytic combustion, membrane separation, biological treatment, and non-thermal plasma [1][2]. Among all of these methods, Membrane Distillation is the best suited because of some compelling advantages viz. high system compactness, operate at low temperature (30°C-90°C) [3].

Membrane distillation (MD) is a temperature-driven procedure in which a microporous membrane separates two liquids or solutions. Due to the non-isothermal nature of the process, molecules of vapor will migrate through the membrane from the high to low vapor pressure side [4]. The membrane distillation separation mechanism is founded on the vapor-liquid equilibrium. This process necessitates that the membrane be dry and that only vapor be present in the pores [5]. To prevent wettability, the maximal pore size must be small (0.1-0.5 pm), the liquid's surface tension must be high, and the membrane material's surface energy must be low [6]. These specifications are met by hydrophobic microporous membranes manufactured from polypropylene (PP), poly-tetra-fluoro-ethylene (PTFE), polyethylene (PE), and polyvinylidene fluoride (PVDF) [7]. The MD process has prospective applications in desalination, blood water removal, milk and juice concentration and alcohol-water mixture separation [8]. The primary advantages of membrane distillation over conventional distillation processes are the elimination of operating mist at a lower cost and the possibility of overcoming corrosion problems by using plastic apparatus. This process is appealing because it can utilize available energy sources, such as solar energy or refuse energy, in industrial processes. However, the procedure is still hindered by the high cost of membranes and the risk of membrane wetting. Membrane Distillation can be categorized into 4 types (i) Direct Contact membrane Distillation (DCMD), (ii) Air Gap Membrane Distillation (AGMD), (iii) Sweeping Gas Membrane Distillation (SGMD) and (iv) Vacuum Membrane Distillation (VMD).

Air-gap membrane distillation (AGMD) is a configuration of membrane distillation in which a stationary air gap exists between the membrane and a condensation surface on the permeate side. The feed evaporates at the interface between the membrane and the liquid, and the evaporated substance passes through the membrane. The liquid then passes through the air gap and condenses on the coolant plate [9]. The temperature difference between the feed

aqueous solution and the cold surface is the driving force for the evaporation of water and the volatile component at the hot feed/membrane interface. Identical to other membrane distillation configurations, mass and heat transfer occur simultaneously through the membrane in AGMD. In this configuration, the air gap reduces the issue of heat loss due to membrane conduction. However, the air gap between the membrane and the condensing surface creates an additional barrier to mass transfer and decreases permeate flux. Several authors also investigated the existing literature on Air Gap Membrane Distillation, Khalifa et.al, (2014) investigated the influence of design and operating variables on the permeate flux of an AGMD unit for desalination of seawater. Their study's modeling of mass and heat transmission was used to predict the flux. Their predicted flux using analytical models correlated well with experimental findings [10]. Geng et.al., (2014) developed a theoretical model of parallel hollow fiber membranes and heat exchange hollow fibers to calculate local water vapor permeate flux distributions and the temperature based on the mass and energy balances of the hot feed side, which demonstrated that the drop in temperature and local water vapor permeate flux were significantly greater at the upper portion of the module in the hot feed side [11]. (B.Ozbey et.al, 2018) investigated the effect of employing a finned coolant plate as opposed to a flat plate for desalination of saline water. An experimental investigation was conducted to determine the effects of various operating conditions on the performance of air gap PTFE membrane distillation in order to provide a more fundamental understanding of the factors that influence the AGMD process[12].

MD is a complex physical process that involves both heat and mass transfers. Water and volatiles evaporate from the liquid–vapor interface on the feed side, diffuse across the membrane, and are then condensed or removed from the membrane module on the permeate side [13]. As depicted in Fig. 1, heat transfer in AGMD is carried out in six steps: (i) heat transfer from the hot feed solution to the liquid–vapor interface on the membrane surface; (ii) heat transfer across the membrane via conduction and latent heat of vaporization; (iii) heat transfer across the air gap; and (iv) heat transfer across the descending film. (vi) heat transfer in the cooling plate [14]. According to the fundamental theory of heat transfer, when a fluid is in direct contact with a solid surface and its temperatures are different, a thermal boundary layer will form. The thermal boundary layer is adjacent to the solid surface, and it is presumed that the fluid's temperature profile is only apparent in this region. The thermal boundary layer adjacent to the heated membrane surface imposes a heat transfer resistance and establishes the temperature difference between the membrane surface and the bulk liquid. This layer's effect is known as the temperature polarization effect [15].

2. MATERIAL AND CHEMICALS REQUIRED

Hydrophobic microporous (PTFE) poly-tetra-fluoro-ethylene membrane (Purchased from Millipore), Certified ACS grade Toluene (purchased from Savita Chemicals, Jaipur) and Distilled water.

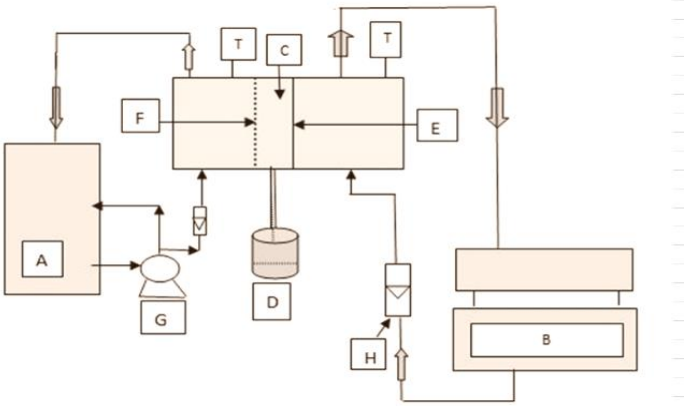

Fig -1: Schematic Representation of Air Gap Membrane Distillation (A= Feed Tank, B= Chiller, C= Air Gap, D =Permeate Receiver, E= Cooling Plate, F= Membrane, G= Pump, H= Rotameter, T= Thermocouples)

2.1 Working of AGMD Experimental Set- Up

The test cell module is divided into three sections: the feed section, the air gap/permeate section, and the refrigeration section. A hydrophobic membrane is positioned between the feed section and the air gap, and a condensing plate is positioned between the air gap and the chilling section. The feed solution was circulated continuously from the feed tank to the feed section and returned to the feed tank using an acid pump (Leakless pumps & Sealings Equipments, Mumbai, India). The feed tank was heated continuously to sustain the desired

temperature. Similarly, the water chiller's cooling water was continuously delivered to the cooling section. By monitoring rotameters, the feed and cooling water flow rates were maintained (Starflow, India). Using digital thermocouples of the J-Type, the feed solution temperature and cooling water temperature were measured at the module entrance. The Toluene and water vapors condensed on the cooling plate after passing through the membrane and air gap. The liquid permeate was captured in the receiver. The concentration of toluene in permeates and retentate was analyzed by using Spectrophotometer (Double beam) at 260 nm wavelength.

Table -1 Membrane Characteristics

Properties	Range
Membrane Diameter (mm)	90
Material	PTFE
Porosity (%)	85
Thickness (µm)	150
Pore Size (µm)	0.22

3. Results and Discussions

The experimental flux and selectivity can be calculated using the formula:

$$J = \frac{\vartheta * d}{S_m * time}$$

Where, J is permeation flux (kg/m^2.h), v is amount of permeate (m^3), S_m is membrane surface area contact with the feed (m^2), d is water density (kg/m^3), t is the time duration.

$$\beta = \frac{(\frac{a_b}{b_b})}{(\frac{1-a_b}{1-b_b})}$$

where, β, a_b and b_b are toluene selectivity, mole fractions of toluene in permeate, and retentate respectively.

3.1 Effect of Air Gap Width on Permeate Flux

It was discovered that the air gap width and feed bulk inlet temperature are the two most influential factors on AGMD performance. From the Figure 2, it can be concluded that on increasing the air gap width from 3 mm to 11 mm, the total permeate flux decreases linearly from 4.65 kg/m^2.h to 1.90 kg/m^2.h, 5.16 kg/m^2.h to 2.72 kg/m^2.h, and 5.46 kg/m^2.h to 3.47 kg/m^2.h at 2, 3, and 4 lpm of feed flow rate respectively under constant feed bulk temperature of 60 °C, 1000 mg/l of toluene concentration, 12°C of cooling water temperature, and 4 lpm of cooling water flow rate.

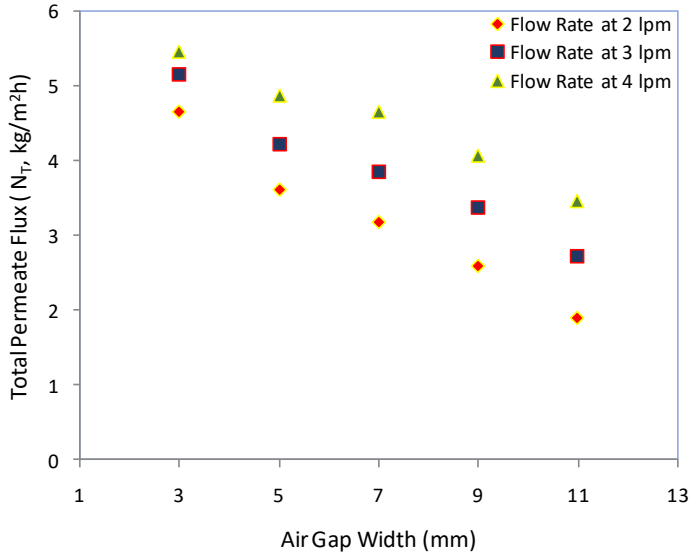

Fig-2 (Effect of air gap width on total permeate flux (Toluene concentration = 1000mg/liter, Feed Inlet temperature = 60°C, Cooling water temperature = 12°C)

This decrease is due to the increased mass transfer resistance at larger air gaps, which decreases the mass transfer coefficient and consequently the transmembrane flux. In addition, upon increasing the air gap width, Knudsen diffusion diminished at a faster rate than molecular diffusion, according to a deeper understanding. Nevertheless, at any given air gap, molecular diffusion predominates Knudsen phenomena. As a consequence, the overall diffusional rate ceases as the air gap increases. From the Figure 3 it can be concluded that on increasing the air gap width from 3 mm to 11 mm, toluene selectivity increased from 62.80 to 54.81, 65.27 to 56.44 and 66.60 to 58.64, respectively which clearly concludes that toluene-water separation taking place using air gap membrane distillation. It is important to note that the air gap width is the most important factor in the separation of toluene from AGMD, as it significantly influences the heat and mass transfer resistance during multicomponent migration across the membrane. Furthermore, (Pedram, Mortaheb, and Arefi-Khonsa, 2019) have reported a positive behavior of air gap width with respect to temperature gradient across the membrane, which ultimately improves permeate permeability, and also confirmed that the air gap width should be minimal for high permeate permeability [16].

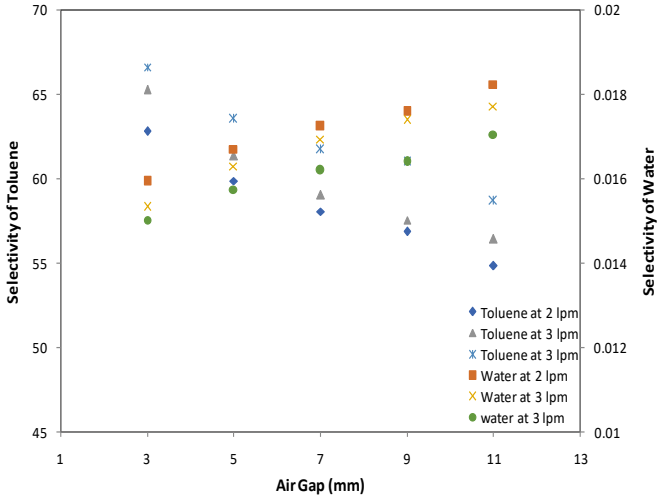

Fig- 3: Selectivity of Toluene and Water at different air gap width

3.2 Fouling Study

Through AGMD setup and continuous run, the efficacy of PTFE membranes was analyzed in this work. The system is operated continuously for 150 hours at a feed flow rate of 2 lpm, a feed bulk inlet temperature of 60°C, a toluene feed concentration of 1000 mg/l, an air gap width of 3 mm, and a cooling water temperature of 12°C at a cooling water flow rate of 4 lpm. Figure 4, depicts FE-SEM and EDS micrographs of a membrane at the magnification of 25,000 x that has been used for 150 hours. SEM and EDS images after experimental use suggests that negligible organic fouling may have occurred on the surface of the membrane, as carbon content increased slightly on the membrane surface after a lengthy run, as observed by the EDS results. Consequently, these SEM-EDS image validate the time-dependent occurrence of a flux-decreasing phenomenon on a micro scale.

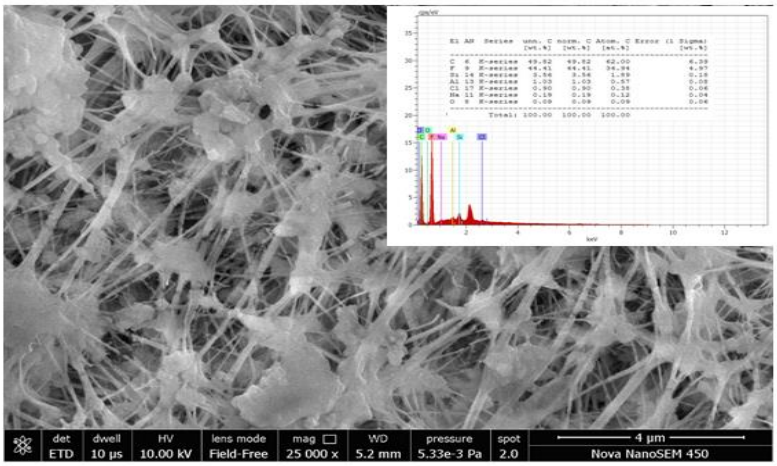

Fig- 4: FE-SEM and EDS image after 150 hours of continuous experimental run.

4. CONCLUSIONS

Experimentally, the performance of Air Gap Membrane Distillation (AGMD) for the separation of toluene from water using a hydrophobic flat sheet PTFE membrane was examined. The effects of air gap width on permeate flux and selectivity was investigated. Moreover, the permeate flux is found to be greater for 3 mm air gap widths that are less than 3 mm. Experiments further showed that increasing the air gap width from 3 to 11 mm decreases the permeate flux from 4.65 kg/m^{2}h to 1.9 kg/m^{2}h. In order to achieve a high permeate flux, it is important to note that the apparatus must be operated at a higher feed bulk temperature and a lower air gap width. It has been determined that the selectivity of toluene in permeate is significantly greater than one, indicating that AGMD is capable of separating toluene effectively. The performance of a membrane under long-term, continuous of 150 hour was investigated, and it was discovered that only negligible organic fouling occurs on the membrane surface. Field Emission Scanning Electron Microscope (FE-SEM) and Electronic Dispersive X-ray Spectroscopy (EDS) were used to investigate the morphology of the PTFE hydrophobic membrane after AGMD experiments.

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