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Feasibility of sweeping gas membrane distillation on concentrating triethylene glycol from waste streams

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ABSTRACT

High treatment efficiency and low energy consumption are desired factors for any process intensification from an industrial prospective. It can be noted to be the same for membrane-based processes for treating triethylene glycol (TEG) wastewater, which is generated from dehydration step in natural gas processing. In this study, sweeping gas membrane distillation (SGMD) was applied to investigate both the potential of process intensification of triethylene glycol (TEG) from binary solutions (water and TEG) and from real wastewater. A hydrophobic hollow fiber membrane module (0.255 m^2) was operated with both synthetic and real TEG wastewater. The feasibility of SGMD for concentrating TEG was done by evaluating various key factors which where; optimizing operating condition like feed temperature, feed flow rate, and sweeping gas flow rate vs permeate flux, energy consumption evaluation and fouling analysis. Experimental results showed that SGMD had the ability to concentrate TEG in real wastewater till 98%. During concentrating TEG from 9.69 to 50%, the permeate flux was in the range from 1.6 to 2.4 kg/m^2 h with the ratio of energy of 1.4 kWh/kg. In regard to fouling, estimations were made for total resistance, membrane resistance, boundary layer resistance and fouling resistance. Fouling contributed 69.2, 7.6, and 23.2% respectively, whereas irreversible resistance accounted for 2% of the total fouling. The results, indicate that bench scale SGMD was able to concentrate TEG up to 98% and potential for pilot scale studies are in need for further scale up.

1. Introduction

Water vapor in raw natural gas causes freezing in pipelines coupled with lowering the fuel's calorific value. Triethylene glycol (TEG, $C_6H_{14}O_4$) is used at industrial scale as a dehumidifying agent to absorb and remove water content in the process which is called as dehydration [1]. TEG is odorless and viscous at STP though the density is very close to water. In the gas separation industry, there are two source of TEG wastewater generated from dehydration unit: (1) from the condenser of TEG recovery system, it has a TEG concentration of 0.1% by volume and (2) from TEG trap of natural gas after crossing dehydration unit, the concentration of TEG in this wastewater varies from 5 to 20%. During normal operating conditions in the dehydration process, the physical properties of TEG do not change and there is potential to recover it from the wastewater for reuse/concentrate for further processing. High concentration TEG infested wastewaters pose a threat to the normal operation of a wastewater treatment plant. Due to the high viscosity, TEG has high potential causing pipe-blocking, also forming flocs in biological treatment unit, reduced treatment efficiency. The first type of TEG wastewater (0.1% TEG concentration) can be treated by the conventional wastewater treatment process. The second source, containing very high TEG concentration, is currently incinerated by a licensed company [2].

In literature, there are studies on treatment and/or concentrate ethylene glycol (a compound similar to TEG) employing evaporation, reverse osmosis (RO), nanofiltration (NF) and pervaporation [3,4]. Evaporation process had been proven as an effective method and reported to concentrate ethylene glycol up to 70% [5]. It can be noted that currently at industrial scale evaporative distillation is being used for concentrating TEG from wastewater or within the process itself. Unfortunately, this is a slow process and consumes high energy. Using nanofiltration (NF) process, concentrating ethylene glycol was studied by [6]; however, the permeate flux and rejection of NF membrane were negligible. Larpkiattaworn et. al [3] studied TEG removal by using polyethersulfone (PES-NTR 7450) membrane by pervaporation. From this study 99% TEG was

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rejected at the condition of feed temperature 28 °C and applied pressure of 1 kg/cm^2. By using two nanofiltration (NF) and two reverse osmosis (RO) modules, Jacob et al., [4] found out that the membrane's selectivity were lost when the TEG concentration in feed solution was higher than 10%. The result of rejection examination of NF and RO were 80 and 83–95% respectively. At the initial concentration of TEG of 5%, the highest TEG concentration achieved was 89.12 and 95.74% for RO-ACM5 and RO-NTR759 membranes, respectively. The authors concluded that pressure based membrane based treatment were effective only for wastewater that has a low TEG concentration of 0.1-5%, thus limiting its application 0.1% TEG generated from the condenser of the TEG recovery system.

Membrane distillation (MD) process is thermally-driven where the driving force is by the difference in vapor pressure between both sides of membrane. In this process, hydrophobic membrane responses as a barrier that only allows vapor to cross the pores. The specific physico-chemical properties of TEG which makes is suitable for using it with membrane distillation are as follows; boiling and freezing point at 760 mmHg is at 288 and −4.3 °C, heat of vaporization 62.5 kJ/g mol, viscosity at 20 °C is 49 mPs, surface tension 45.5 mN/m, vapor pressure at 20 °C <0.001 kPa and solubility in water is 100 wt% at 20 °C. Implying that during membrane distillation process from a mixture; only TEG will get concentrated in the feed and water vapor will pass through the hydrophobic pores. In literature, ethylene glycol (EG) separations have been reported for two MD configurations. The first study was conducted with direct contact membrane distillation (DCMD) [7], where it could concentrate EG at 70% of concentration by using DCMD operating at moderate temperature and atmospheric pressure. However, this marked the limitation of DCMD. The authors noted that adverse effects of temperature and concentration polarization. EG was also concentrated using vacuum membrane distillation (VMD) by Mohammadi and Akbarabadi [8] with a rejection of 100%. However, VMD consumes high energy and the authors noted that after ethylene glycol concentration reached 60 wt.%, membrane wetting occurred.

In literature, Sweeping gas membrane distillation (SGMD, another configuration of membrane distillation) is mostly used to remove volatile compound than producing water [9,10]. The operating principle for SGMD is, when a feed solution is heated up until desired temperature and transferred to the membrane surface. Here the volatile compounds pass across the membrane pores and to the permeate side, where a carrier gas collects and transfers the vapor for further processing/extraction. Meanwhile, the non-volatile compounds get concentrated and would re-circulated to the feed tank. Ideally the hydrophobic nature of porous membrane distillation prevents liquid from passing through the pores, assuming the liquid-vapor interface takes place at the entrance of each pore. SGMD, is generally better for concentrating feed than recovering permeate, thus making it ideal to concentrate TEG than an another configuration. This configuration has an added advantage that flux generated is not generally influenced by the inlet gas temperature [9], thus ambient air can be used to carry the water vapor to the atmosphere without any further treatment. But in smaller module, it has shown some effect, indicated by small reduction in flux. In DCMD configurations, permeate fluid temperature plays an important role in flux, whereas in SGMD, the role is insignificant. Thus the objective of this study was to investigate the feasibility of concentrating TEG with energy evaluation from both synthetic and real wastewaters by utilizing a bench SGMD system. This objective was achieved by evaluating various key factors which where; optimizing operating condition like feed temperature, feed flow rate, and sweeping gas flow rate vs permeate flux, energy consumption evaluation and fouling analysis.

2. Materials and methods

The hydrophobic hollow fiber MD module which was used in this study was manufactured by Sumitomo Electric Industries, Ltd. POREFLON® (Model number TB-21-02, type No. 130529-1). As reported by the manufacturer, the hollow fiber membrane with a surface area of 0.255 m^2 was made of polytetrafluoethylene (PTFE) with a contact angle 112° and a nominal pore size of 0.45 μm. At first, the membrane module was verified by three experiments, which were: liquid entry pressure (LEP), salt rejection test and with pure water (deionized water) test. The variables in pure water test included: feed temperature, feed flow rate, and sweeping gas flow rate. The best-performing operating conditions were chosen to conduct the next set of experiment. Secondly, synthetic wastewater was used as feed, which was a mixture of pure TEG (obtained from the gas separation plant) and distilled water. This was done to evaluate the performance of MD process on concentrating TEG without any interference from other impurities. Lastly, the membrane systems were operated with real wastewater which was provided by the gas separation plant. In real wastewater, the concentration of TEG varied from 5 to 20%. Moreover, real wastewater contained impurities and were removed using a pretreatment system as described by [4]. This pretreatment system used a microfiltration and ultrafiltration unit together with aeration, with a purpose of removing, suspended solids, oil and grease, BTEX and iron oxide. The intention for operating the membrane system with real wastewater was to identify the performance of hollow fiber membrane distillation in the condition which was close to the real condition at gas separation plant. Energy consumption and membrane fouling phenomena were also evaluated.

2.1. Liquid entry pressure

The liquid entry pressure (LEP) experiment was conducted as follows: one side of permeate-outlet of the hollow fiber membrane module and the feed-outlet were blocked. Deionized water was pushed into the feed-inlet (by a pressure tank with nitrogen gas). Pressure was increased in steps, water passed through the fiber by the outside-in direction. A digital pressure gauge (model: PG-30-102R-N, Copal Electronic Inc.) connected to the data logger (model: EL-USB-4, Lascar Electronics Ltd) was used to record all values at 1 Hz. The theoretical LEP was also calculated by using Laplace equation $(LEP = (-2B.\gamma_l.\cos\theta)/r_{max})$ to be 130.5 kPA (B is pore geometric coefficient (assumed B = 1, cylindrical pore), γ_L is liquid surface tension (γ_L = 0.0644 N/m as surface tension of water at 70 °C), θ is contact angle of the membrane surface with liquid (θ = 112°) and r_{max} is the maximum pore size (r_{max} = 0.45 μm).

2.2. Sweeping gas membrane distillation system

In this study, the system was operated outside-in configuration (feed on the outside and sweeping air on the inside) and in cross flow mode. Feed tank was temperature controlled (30–100 °C) and insulated. The level indicator on the feed tank (with the total volume of 15 L) was used to observe flux. Feed flow rate was 2.4 L/min using a magnetic pump. A bypass from the feed pipe after the pump was provided if the pressure generated was too high or to control the flow rate. But in the present case it was not used, as neither the pressure nor the flowrate was changed during the entire duration of the experiments. Temperature probes and pressure probes were located on the inlet and exit of the feed pipe to the membrane module. Calibrated volume of air was sent to the membrane from an air compressor which was also monitored with pressure and temperature sensors. Heat and mass transfer