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Development of Bench-Scale Direct Contact Membrane Distillation System for Treatment of Palm Oil Mill Effluent

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ABSTRACT

The oil palm industries generate large amounts of effluent that can cause pollution and, as a result, pose serious threats to the environment. Recently, membrane distillation (MD) has been identified as a potential candidate for treatment of palm oil mill effluents (POME). This is because the release of POME at high temperature may reduce the need to preheat the effluent prior to operation. The objective of this article is to manufacture a bench-scale MD system and assess the performance of the system in POME treatment. This project started with the design of a process and instrumentation diagram (P&ID) and continued with the fabrication part. Subsequently, the performance of the direct contact membrane distillation (DCMD) system was tested using POME as a feed solution. Three different feed temperatures (55 °C, 65 °C and 75 °C) were analyzed during the testing. The results obtained show that the permeate flux increased with the increase of feed temperature with the highest flux is 10.76 kg/m^2.hr. More than 85% of pollutants including ammonia nitrogen, color, chemical oxygen demand (COD) and turbidity were rejected. This shows that the system is capable of treating POME and producing high quality permeate water.

Keywords: Membrane distillation, oil palm industry, POME, PVDF, wastewater treatment

1.0 INTRODUCTION

Membrane distillation (MD) is a thermally driven separation process in which two phases with varying partial pressures are separated by a hydrophobic microporous membrane [1-3]. Water vapor from the high partial pressure side penetrates the hydrophobic membrane, followed by a condensation process from the cold permeate side [4-5]. This unique process has made MD exclusive for an application where water is the major component of the feed solution. This is due to the fact that MD can reject dissolved and nonvolatile species, which can be eliminated at 100% [6-7]. In addition, MD operates at low temperature and pressure, making it a promising membrane separation technology, especially for industrial effluent treatment [2, 7].

In spite of the numerous advantages of the MD system, this technology also presents challenges in industrial sectors. The significant obstacle lies from the development of the suitable membrane. Researchers are struggling to develop a novel membrane that can achieve delicate balance between hydrophobicity with consistent properties, such as pore size, thickness, and surface roughness. These membrane properties are crucial for optimal performance and to avoid membrane wetting and fouling, which can significantly reduce the system's efficiency and lifespan [8].

Another major challenge in MD development is the technological readiness of the MD system for

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commercial or industrial applications. Up to now, most studies have reported improved permeate flux and reduced membrane fouling through the use of a bench-scale MD system [9]. Despite the relatively high number of research papers on MD, records on patent applications for MD equipment and processes or membrane development have remained relatively constant over the past decade [10]. Although small-scale prototypes have demonstrated MD's feasibility, the upgrade requires overcoming various engineering and design barriers.

Several factors must be taken into account, where the module design and fabrication must be appropriate for larger membrane surfaces, robust module construction and efficient heat transfer mechanisms to ensure efficient mass and energy transfer on an industrial scale. In addition to the compatibility of adhesives, seals, spacers, and feed distributors, certain membrane system components such as housings and connectors, must be carefully selected [11]. Furthermore, the piping system must be durable to treat highly hazardous and corrosive industrial effluents.

In general, MD has four main configurations which are sweeping gas membrane distillation (SGMD), vacuum membrane distillation (VMD), air gap membrane distillation (AGMD) and direct contact membrane distillation (DCMD) [12-14]. The SGMD operates by allowing a cold inert gas stream from the permeate side as a sweeping gas. The gas will then transport the vapor molecules outside the membrane module where condensation takes place [14]. Among these configurations, SGMD is the least used configuration due to the high operational cost of the external condensing system. On the other hand, VMD uses vacuum pump as the driving force at the permeate side. The applied vacuum pressure is lower than the equilibrium vapor pressure and the condensation takes place outside of the membrane module [12]. Meanwhile water vapor in AGMD is condensed on a cold surface that has been separated from the membrane via an air gap. The heat losses are reduced in this configuration by addition of a stagnant air gap between membrane and condensation surface [14]. In DCMD configuration, liquid from the permeate and condensate sides is in direct contact with the hydrophobic membrane, whereas cold distilled/deionized water will be used as a condensation medium. DCMD is the most applicable configuration because of the simplest fabrication configuration and minimal equipment used [15-16].

The present work involved the design and manufacturing process of the bench-scale DCMD system for the treatment of palm oil mill effluent (POME). Although the commercial system is easily available in the marketplace, it should be noted that the commercial price of the existing MD system is still considered expensive because it involves heating and cooling processes during the treatment process. The system is patented with limited design specifications that prevent the user/researcher from further exploring the impact of the design system on membrane productivity, such as the design of the membrane module, the types of membrane and the orientation of the membrane module.

The reason for selecting POME as the primary source for the MD processing is because palm oil is the most widely used edible oil in the world, accounting for about 37% of the total vegetable oil production [17]. The process to extract the oil requires significantly large quantities of water for stream sterilizing the palm fruit bunches and clarifying the extracted oil. It is estimated that for 1 ton of crude palm oil produced, about 5 to 7.5 tons of water were used and 50% of this water end up as POME [18]. Discharges of untreated POME into watercourses will pose risks to the environment, particularly to aquatic organisms [19-20]. Another reason for addressing POME as a feed solution for the MD system is due to the thermal properties of the industrial effluent that normally discharges at high temperatures [21-22]. The thermal energy from the effluent can reduce the requirement to preheat the feed solution prior to the testing. In terms of membrane material, Polyvinylidene Fluoride (PVDF) has been employed in the fabricated DCMD system due to its unique advantages such as high mechanical strength, hydrophobic, good processability, chemical stability and heat resistant [12,23]. Prior to the performance testing, several analyses will be conducted to assess the potential of the system to reduce the environmental impact of industrial wastewater.

2.0 EXPERIMENTAL

2.1 Materials

PVDF polymer (Kynar® 760, MW = 440,000 g/mol) was purchased from Arkema Inc., Philadelphia, USA. N-methyl-2-pyrrolidone (NMP) with purity more than 99.5% was purchased from Merck and used as solvent without further purification. The materials used to manufacture the system were obtained from local hardware.

2.2 Membrane Fabrication

The polymer solution was prepared by dissolving 12 wt% PVDF in NMP at a temperature of 60˚C under constant stirring rate (~450 rpm) until a homogeneous solution was obtained. The spinning conditions of each parameter applied in this work are summarized in Table 1, while the detailed description of the spinning process can be found in our previous work [23]. After spinning, the spun fibers were submerged in water for a few days to completely eliminate the residual solvent.

Table 1 Spinning conditions of the PVDF hollow fiber membrane

Spinning conditions	Value
Bore fluid flow rate (ml/min)	2.0
Dope extrusion rate (ml/min)	4.0
Bore fluid composition	Deionized water
Coagulation medium	Tap water
Spinneret OD/ID (mm/mm)	1.15/0.55
Air gap distance (cm)	17.5
Spinning dope temperature (˚C)	25 (±1)
External coagulation temperature (˚C)	25 (±1)

2.3 Design and Fabrication Process of the Direct Contact Membrane Distillation System

Figure 1 shows the Process and Instrument Diagram (P&ID) of the DCMD system, consisting of two thermostatic cycles directly connected to the membrane module. The membrane module is a complete unit composed of hollow fiber membranes, a housing, feed and permeate inlets, as well as the feed and permeate outlets (see Figure 2). In this system, the membrane module will serve as a separation medium for the DCMD process. 20 hollow fibre membranes were inserted into the membrane module and the two ends of the membrane module were sealed with epoxy adhesive to avoid the permeate solution penetration into the shell side.

For the liquid storage system, two stainless steel tanks were prepared.

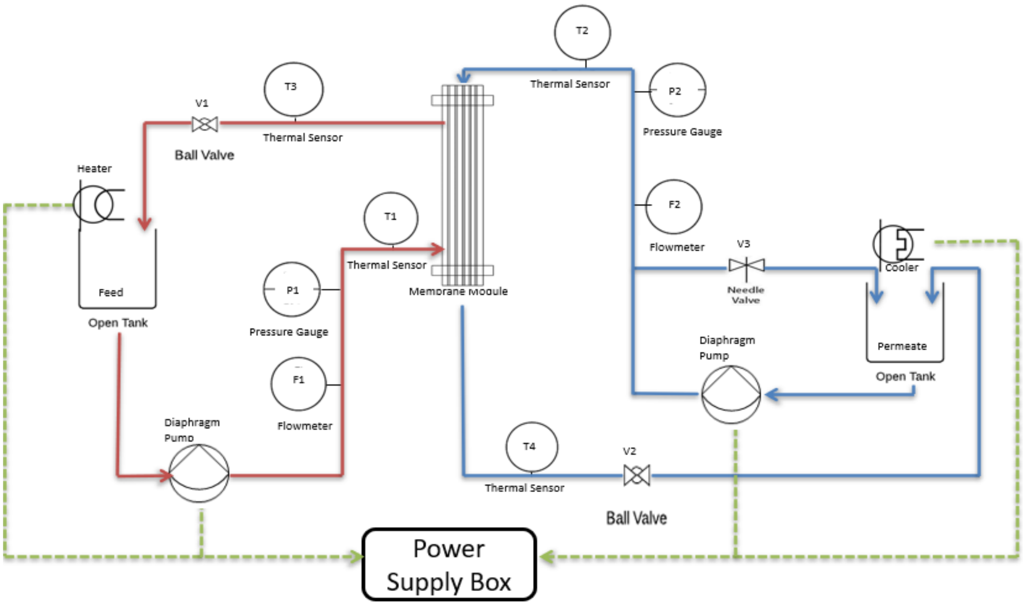

Figure 1 P&ID of DCMD system comprised of hot and cold thermostatic cycles

POME was stored in the feed tank before entering the membrane module on the shell side. On one hand, the condensing medium which is pure water was stored in the permeate tank before entering the membrane module from the lumen side of the hollow fiber membrane. The configuration of the membrane module was adjusted vertically in outside-in mode. Since the separation process can only occur if there is a temperature difference between the two solutions, an electric heater was installed inside the feed tank to heat up the feed solution, while a chiller was used to cool the permeate solution to the desired temperature. The electrical heater (Protech, model 830-S1, Malaysia) and recirculating chiller (Vivo, model RT2, Germany) were used to support the system accordingly. Temperature sensors were placed in several parts of the piping system to detect the actual input and output temperature. Two pumps were installed after the feed and permeate tanks for fluid transmission. To measure the actual pressure of the inlet solutions, two pressure gauges were mounted to the piping system. The ball valve was used as a flow regulator and the needle valve was used as a pressure regulator for the by-pass flow. In order to measure the permeate flux, a top plate balance (Mettler Toledo, model ME3002, Switzerland) was placed under the permeate tank. The final image of the fabricated system is presented in Figure 3.

2.2 Sampling Process

POME samples were collected at Felda Palm Industries Sdn. Bhd., Felda Lepar Hilir 3, Gambang, Pahang, Malaysia with a coordinate of 3°38'37.8"N 103°00'43.0"E. Samples were taken from the anaerobic pond of the wastewater treatment plant. Upon arrival at the laboratory, the samples were properly preserved to avoid chemical and biological changes in the samples. Changes occur rapidly once the waste has been collected. Hence, the samples were kept as cool as possible without freezing in order to minimize the potential for volatilization or biodegradation between sampling and analysis. The samples were refrigerated to 4°C before the analysis, which followed the preserve methods as presented in Table 2.

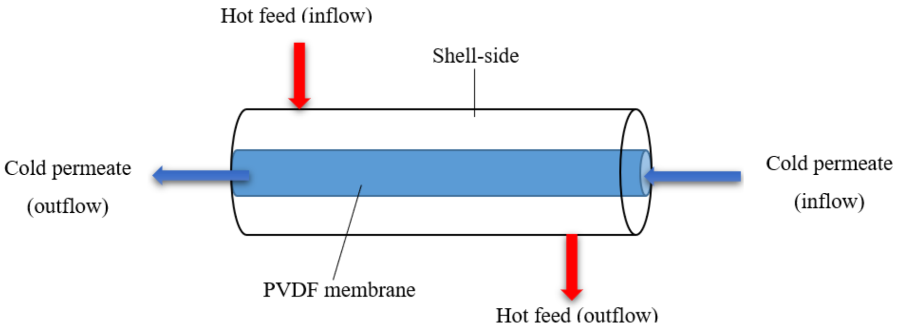

Figure 2 Counter-current flow configuration of the membrane module

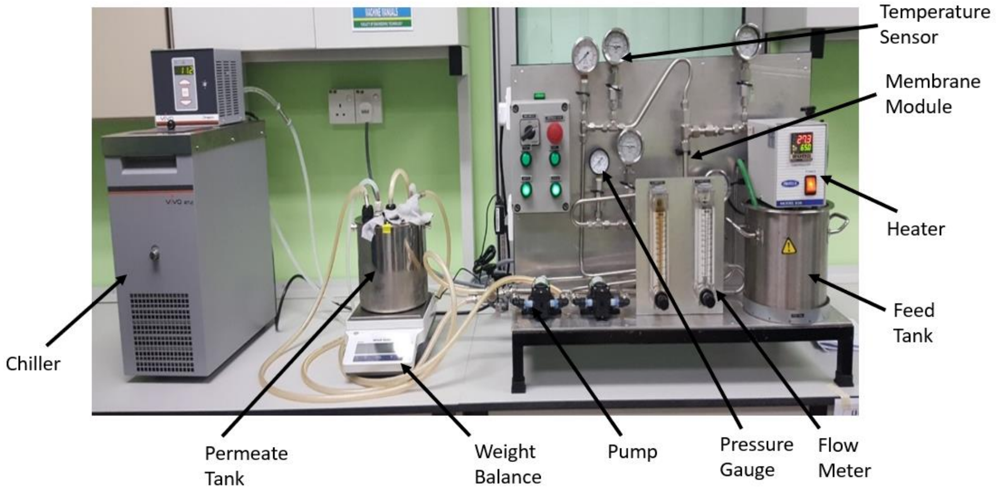

Figure 3 The completed Direct Contact Membrane Distillation (DCMD) system

Table 2 Methods of preserving for selected parameters

Parameter	Preservation Method	Maximum Holding Time
Chemical Oxygen Demand (COD)	Cool 4˚C and add H_2SO_4	28days
Total Suspended Solids (TSS)	Cool 4˚C	7 days
Turbidity	Cool 4˚C	48 hours
Ammonia Nitrogen	Cool 4˚C and add H_2SO_4	28 days
Nitrate Nitrogen	Cool 4˚C and add H_2SO_4	28 days
Color	Cool 4˚C	48hours

* RMB Environmental Laboratories, 2017

Samples before and after testing were analyzed using the Water Analysis Method which referred to the standard water and wastewater examination by American Public Heath Associate (APHA). Samples collected were analyzed within one week in order to avoid excessive deterioration. The POME parameters were analyzed according to COD, TSS, turbidity, ammonia nitrogen, nitrous nitrogen, pH, and color. The Ex-Situ approach was used for the water analysis.

Prior to the MD testing, the POME samples were filtered using cellulose filter paper (diameter: 125 mm; pore size: 20 μm). The filter assembly consists of a vacuum pump, a Buchner flask and a funnel. Pre-treatment of the effluent is necessary to avoid membrane clogging or membrane fouling. Fouling is the deposition of unwanted substances on the surface of the membrane and inside the pores of the membrane. This will result in a reduction of permeate flux and deterioration of membrane conditions.

2.3 Direct Contact Membrane Distillation Performance Testing

In this study, 2 L of pre-treated POME was stored in the feed tank and heated to a certain temperature. On one side, 2 L of distilled water was stored in permeate tank as the condensing medium. DCMD experiments were conducted at various feed temperatures (55, 65 and 75°C) while maintaining the permeate side temperature at 25°C. The working pressure of the feed and permeate solutions was determined to be 8 psi and 2 psi, respectively. Permeate and feed flowrates were controlled at 0.5 LPM and 2.2 LPM. The experiment was carried out for one hour continuously and the weight of the permeate was recorded every 15-minute. The permeate flux, J_v ($kg/m^2.hr$) in the permeate tank was calculated using Eq. 1.

$$J_v = \frac{\Delta W}{A\Delta t} \qquad (1)$$

where ΔW (kg) is the collected permeate weight, over time Δt (hr) and A (m^2) is membrane area. The effective membrane area was calculated based on Eq. 2.

$$A(m^2) = n\pi d_o L \qquad (2)$$

where n refers to the number of hollow fibers, d_o (m) corresponds to the outer diameter of hollow fibers, L (m) indicates the effective fiber length. Meanwhile, Eq. 3 was used to determine the solute rejection, R (%) of the membrane.

$$R(\%) = \left(1 - \frac{C_P}{C_f}\right) \qquad (3)$$

where C_p and C_f stand for permeate and feed concentration (mg/L), respectively. The liquid samples in the permeate tank were collected every 15 minutes to analyze the quality of the treated wastewater.

3.0 RESULTS AND DISCUSSION

3.1 Pre-treatment Analysis

The characteristics of POME samples taken from the anaerobic pond and after pre-treatment with filter paper were summarized in Table 3. From the analysis, TSS and turbidity decreased substantially by approximately 76% and 80%, respectively. However, only 10.60% of COD were rejected after pre-treatment. Ammonia nitrogen also declined by only 18.33%. Observations indicate that the color of POME after pre-treatment is still brown, although nearly 45% reduction in color has been achieved. The results showed that the pre-treatment is not sufficient to purify the POME. The brownish color is attributed to the high content of carotene, pectin, tannin, phenolic compounds, and lignin. POME also contains high levels of carbohydrates, amino acids, free organic acids and inorganic minerals [24]. It can be clearly noticed that the water parameters still beyond the standard set by the Malaysian Department of Environment. In this study, it is proven that simple filtration using cellulose filter paper only manage to remove the TSS and turbidity of the wastewater due to the filter size. To further remove the pollutants, the wastewater must be treated using advanced technology like MD.

3.2 DCMD Analysis

Figure 4 depicts the rejection rate obtained during the DCMD test. Turbidity is the cloudiness of a liquid caused by suspended solids that are

Table 3 Analysis of wastewater in before and after the pre-treatment process

Parameters	**Unit**	**Before Pre-treatment**	**After Pre-treatment**	***DOE Standard for POME**
COD	mg/L	698	624	100
TSS	mg/L	646.7	150	400
Turbidity	NTU	781.67	153	-
Ammonia Nitrogen	mg/L	60	49	-
Nitrate Nitrogen	mg/L	29	14	200
Color	Pt/Co	4480	2480	200

*Malaysian Department of Environment (DOE), 1982

usually invisible to the naked eye. Using the DCMD system, turbidity is eliminated up to 80%, demonstrating a high performance of the DCMD system to remove suspended and colloidal matter from the effluent. With respect to COD, the rejection rate was over 90%. This may be due to the effectiveness of the PVDF membrane to retain the oxidizing organic material from penetrating into the membrane pores.

Ammoniacal nitrogen (NH_3-N) is a measurement of the total quantity of ammonia, a toxic pollutant in the sample. POME that rich in ammonia nitrogen would inhibit the natural nitrification and cause water hypoxia. This will result in fish poisoning, reduced water purification capacity and, ultimately, severe damage to the aquatic environment. Hence, the ammonia nitrogen elimination is crucial and necessary. In this experiment, ammoniacal nitrogen achieved a rejection rate above 85% in one hour. At the same time, nitrogen compounds act as nutrients in waterways and rivers [25]. Nitrate reactions (NO_3) in fresh water can cause oxygen depletion that may affect aquatic organisms. Based on the data, it should be noted that nitrous nitrogen was eliminated in less than 1 mg/L with a release rate of more than 95 %. This has proven that the DCMD system is effective for the removal of nitrate nitrogen pollutants.

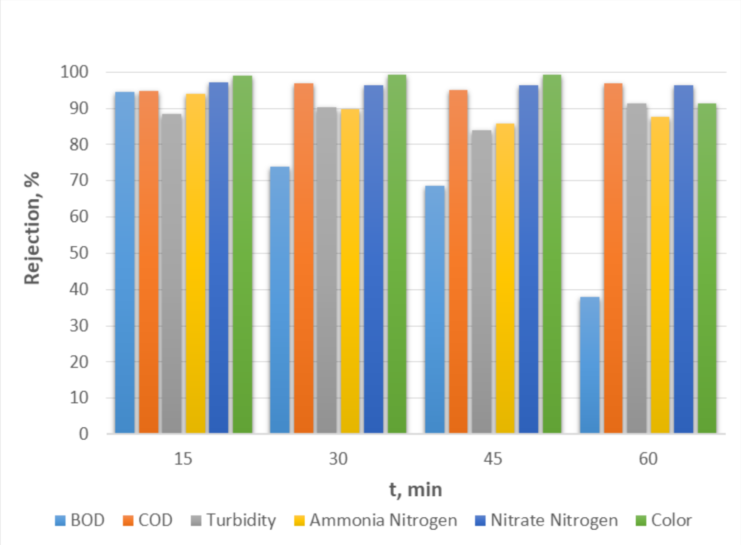

Figure 4 Rejection rate versus time (conditions= hot stream: 55˚C at volume flow rate of 2.7 Lpm, cold stream: 25˚C at volume flow rate of 0.5 Lpm)

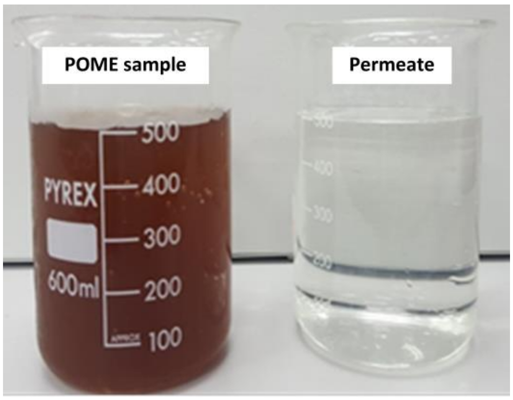

Figure 5 Samples of POME in the feed tank (after pre-treated with filter paper) and final treated water at the permeate tank

The color's rejection rate is nearly 100% after running through the system. The wastewater changed from brownish color to transparent clean water as shown in Figure 5. High retention of organic and inorganic contaminants from POME has resulted in high removal of color as well as turbidity. Since only water vapor is allowed to pass through the pores of the membrane, it is therefore not impossible for the final treated water to be colorless and transparent. The results from the DCMD test at a different feed temperature are shown in Figure 6. The results indicate that the permeate flow is highly influenced by the feed temperature [12]. The maximum permeate flux (10.76 kg/m^2.hr) was achieved when the POME solution was operating at 75°C while the lowest permeate flux was obtained when the feed temperature was 55°C (3.90 kg/m^2.hr). The results are in agreement with the other studies [7,21]. As expected, the increase in feed temperature induces the vapour pressure difference between the two solutions and significantly increases the permeate flux. This is due to the fact that the vapour pressure of the gas-liquid interface on the liquid feed side increases as the feed temperature increases [21]. The supply temperature has a significant impact on the permeate flow as mass and heat transfer occur simultaneously in the DCMD process.

4.0 CONCLUSION

The bench-scale DCMD system was successfully fabricated in this study. The PVDF membrane was used as a separation agent for the treatment of effluents from the palm oil industry.

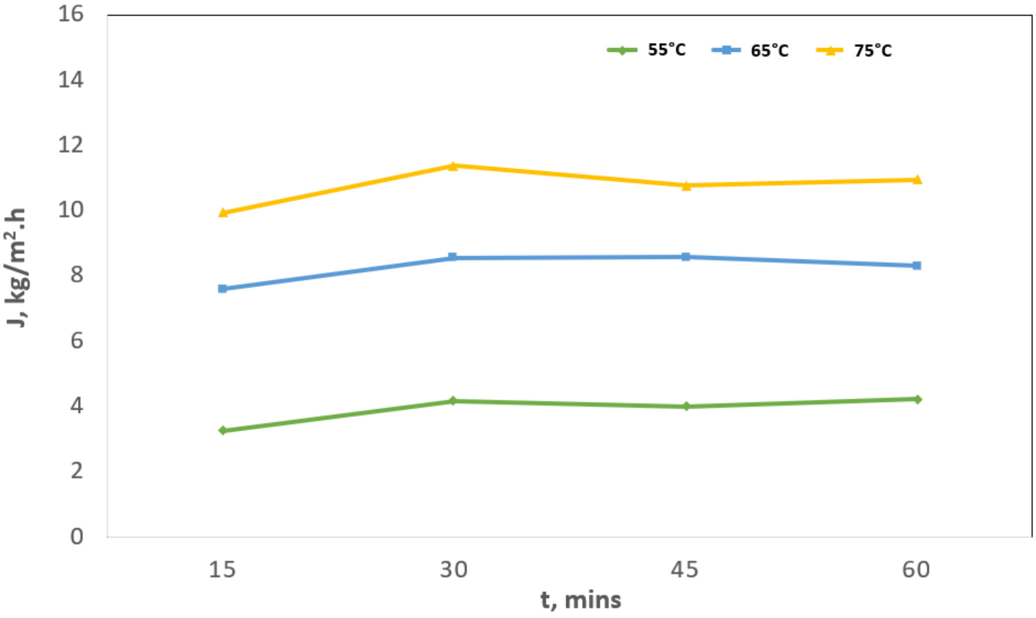

Figure 6 Permeate flux as a function of time and feed temperature

Based on the performance analysis, the results show that the permeate flux was highly dependent on feed temperature. The higher the feed temperature, the higher the permeate flux. With respect to the quality of the treated water, it shows that all parameters tested, including ammonia nitrogen, color, COD and turbidity, were released at over 85%. In conclusion, the self-manufactured DCMD system in this study proved capable of transforming brownish POME into clean, high-quality water using the hydrophobic PVDF hollow fiber membrane.

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