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Application of Nanofiltration and Reverse Osmosis in Wastewater Treatment Containing Ethylene Glycol from South Pars Gas Complex Wastewater

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ABSTRACT

Today, industrial wastewater discharge has become one of the serious problems of governments. Industrial wastewater contains harmful and dangerous compounds that can endanger human health and the environment. In addition, in the oil, gas, and petrochemical industries, ethylene glycol is used for various purposes, and its removal from wastewater has become a challenge. Industries located on the shores of the Persian Gulf are one of the sources of production of this type of wastewater. In this study, the performance of Nanofiltration (NF) and Reverse Osmosis (RO) membrane processes for treatment of wastewater containing ethylene glycol (EG) from South Pars Gas Complex (SPGC) are investigated and compared. Various process parameters such as pressure, flowrate, and EG removal percentage as well as performance and characteristics of the membrane such as flux and fouling are discussed. It has been observed RO has better performance and less fouling than NF. However, the NF membrane has higher flux due to less compression. RO process achieved 80-99% and NF achieved 60-80% EG rejection during various pressure and flowrates.

Keywords: Wastewater treatment, Nanofiltration, Reverse Osmosis, Ethylene glycol, membrane separation

1.0 INTRODUCTION

Nowadays, water scarcity is one of the major challenges in the world. Water shortage has been increased due to the population growth and consumption as well as pollution of the sources due to humankind activities. The world's population is estimated to increase by more than two billion by 2050 [1]; however, more than one billion people live in water-scare areas in the world today. On the other hand, water consumption has been increased more than fivefold in the last century [2, 3]. Water scarcity is affected by the supply and demand cycle. It is predicted that the average renewable water in the Persian Gulf region is about 1000 cubic meters per capita per year, while the global average is more than 5000 cubic meters per capita. The availability of common water resources is affected by increasing water demand and declining surface and groundwater quality [4].

Water pollution due to urbanization, population growth, industrialization, food production methods, unsustainable water consumption, and inefficient water and wastewater management strategies has become one of the major challenges of governments

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[5]. Industrial wastewater may contain several harmful and undesired contaminants such as toxic heavy metals, dyes, nutrients, microbial organisms, endocrine disrupting compounds, and chemical components [5]. In many countries, wastewater is discharged into water bodies without any treatment or only after an elementary treatment. Discharging wastewater without treatment may cause several environmental issues: Dissolved oxygen for supplying Biochemical Oxygen Demand (BOD), consumed by organic contaminants. Nutrients in the wastewater can affect the aquatic plants Growth and cause eutrophication. Untreated wastewater may release a large amount of toxic components, pathogens, and harmful microorganisms, which can endanger human health [6]. It has been observed that more than 70% of fatal diseases are caused by water pollution [7].

Oil and gas industry wastewater includes chemical components such as ammonia, phenol, cyanide, phosphorus, heavy metals, light hydrocarbons, and heavy hydrocarbons, pH changes, biological contaminants high, and other contaminants like solids, oil, suspended substances. Water is part of Natural gas and crude oil reservoirs and it can increase the corrosion rate of the pipes and make hydrates at low temperatures and high pressures during oil and gas extraction and transport. Nowadays, chemical inhibitors are used to prevent hydrate formation. The most common method is using thermodynamic inhibitors such as alcohol, glycols, and electrolytes [8, 9]. Glycol injection is the most common method to prevent hydrate formation in Iran's oil and gas industries; therefore, large amounts of glycol are found in the wastewater of these industries.

Ethylene glycol (EG) is a biodegradable solvent that can be dissolved in water in any proportion and cannot be separated by evaporation. EG is considered one of the main pollutants in industrial wastewater and in addition to environmental hazards, it can also pose risks to human health. The main effects of EG release include absorption through skin contact, brain damage, damage to the central nervous system and joints, eye damage, toxicity, etc. [10, 11].

Various methods have been used to treat industrial wastewater, such as Reverse Osmosis (RO), Nanofiltration (NF), Membrane Bioreactor (MBR), Adsorption, etc. Bayat *et al*. evaluated a MBR system to treat wastewater containing EG from a petrochemical industry wastewater. They found that the performance of COD removal in this process is between 85 – 97.5% [11]. Another study by Zinadini *et al*. investigate application of UF-MBR process to treat milk industry wastewater. Similarly, it has been found this hybrid system increase the COD and organic matter removal efficiency [12]. Pervez *et al*. assessed the application and performance of NMs-MBR membranes. They concluded applying NMs-MBR will decrease fouling rate, and increase removal efficiency [13]. Jacob *et al*. investigated two NF and two RO membranes for TEG removal from two wastewater streams containing TEG in range of 0.1 – 10 volume percent [14]. These membranes achieved 89 – 96% TEG removal, while it has been found 10 – 70% fouling in membranes. This study focused on investigating operational parameters and membrane fouling for NF and RO membranes to treat wastewater containing glycol. NF and RO are able to remove polyvalent ions and salts such as iron, manganese, uranium, and pesticides, organic pollutants, bacteria, viruses, and microorganisms, treat large-scale purified water, and remove water hardness and salinity. NF is more costefficient than RO, but RO is able to remove monovalent ions [3, 15].

In the present study, the performance of NF and RO membrane modules in wastewater treatment and EG recovery at the fourth phase of the South Pars Gas Complex (SPGC) refinery is investigated and analyzed. The main goals of this study are to recover EG to reuse in industry and treat water to return to the industrial cycle or use agricultural lands as well as prevent environmental degradation and pollution of the Persian Gulf. Furthermore, the effects of operational parameters such as pressure and flowrate on the performance of the NF package are investigated and assessed.

2.0 METHODS

2.1 Experimental Device

The experimental device consists of several parts:

A micron filter and a carbon filter for pre-treatment,
A 60l tank with a cooling system,
A high-pressure pump (HPP) manufactured by Ebara (Model: Compact AM 15) to supply the required pressure to input water to the membrane,
A high-pressure membrane package,
Two flowmeters manufactured by MBLD (model: LZT-1002M) to measure the flowrate of purified water and wastewater,
Three relative barometers manufactured by Nuova FIMA (model: EN 837-1) to measure the inlet flow pressure to the HPP, the pressure applied to the membrane, and outlet treated water pressure,
A Feed pump manufactured by RENYUWANG (model: QB-60),
Anti-scalant injection pump manufactured by Injecta (model: Athena 2),
A Solenoid Valve manufactured by Uni-D (model: UW-15), and
A TDS meter manufactured by LUNA water.

The schematic of the applied experimental device and equipment are shown in Figure 1.

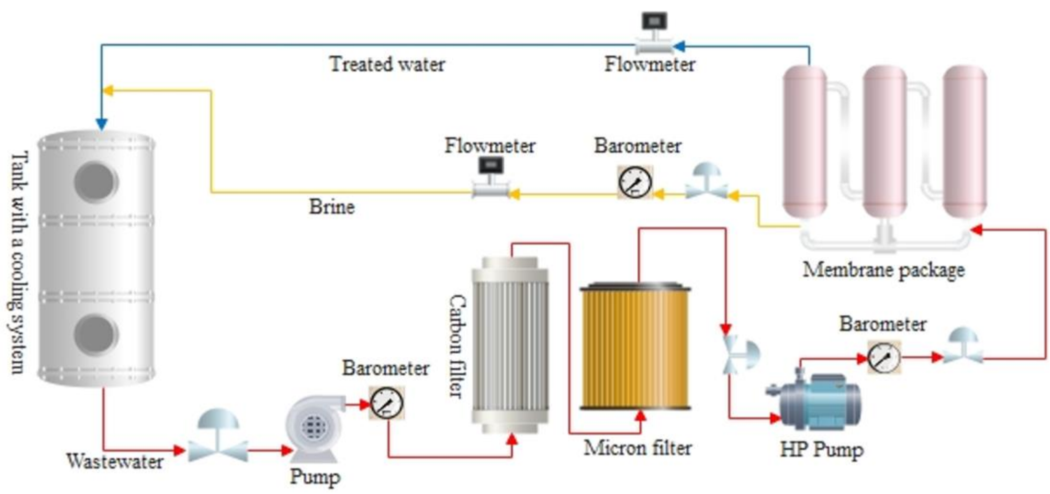

Figure 1 Schematic of experimental device and equipment

2.2 Membrane Modules Properties

In this study, one NF and one RO membrane module have been applied and investigated. NF membrane is a spiral wound membrane NE4040-90 model manufactured by CSM LENNTECH Co., and RO spiral wound membrane model is RE4040-BE that manufactured by CSM LENNTCH Co. The characterization of membranes and modules are listed in Table 1.

Table 1 The characterization of membranes and modules

Properties	NF	RO
Model	NE4040-90	RE4040-BE
Dimensions	4 * 40 in	4 * 40 in
Effective membrane area	7.9 m^2	7.9 m^2
Monovalent ion rejection (NaCl)	85.0 – 95.0%	99.7%
Divalent ion rejection $(CaCl_2)^{2-}$	90.0 – 95.0%	-
Membrane type	Thin-Film Composite (TFC)	Thin-Film Composite (TFC)
Membrane material	Polyamide (PA)	Polyamide (PA)
Element configuration	Spiral-Wound	Spiral-Wound
Max. Operating Pressure	600 psi (4.14 MPa)	600 psi (4.14 MPa)
Max. Operating Temperature	113 °F (45 °C)	113 °F (45 °C)
Operating pH Range	2.0-11.0	2.0-11.0
Max. SDI (15 min)	5.0	5.0

2.3 Experiment

At first, twice-distilled water is passed through the package for an hour to clean the equipment and membrane modules. After washing, the experimental solution is inputted into the tank, and the device is running for 1.5 hours. In this study, both the treated and the wastewater are returned to the main tank to perform the experiments at a constant concentration.

A pressure valve is used to regulate the pressure applied to the membrane. It takes 90 minutes for the experimental conditions to be stabilized. Then the flowrates, input and output membrane pressures, total dissolved solids (TDS), and temperature are recorded. At the end of the time, the solution in the tank and the filtration stream are sampled simultaneously, to determine the amount of filtration under the applied conditions. It should be noted, before and after the treatment, the device runs for 30 minutes using double distilled water at the desired operating conditions (pressure, flowrate, and membrane pressure), in order to investigate the treatment of the wastewater and outlet wastewater fluxes changes.

This study is carried out in two different sections, each with a single membrane type. Every section consists of two series of general tests on different operating conditions at a temperature range of 30 + 3 ° C.

Step 1. In the first series of both experiments, tests are run at 720 L/h at different pressures of 5, 6, and 7 bar with three inlet feed types. The first type is twice-distilled water, the second type is synthetic wastewater containing 300 ppm EG, and the third type is the wastewater from the Glycol recovery unit of the fourth refinery of SPGC. In order to evaluate membranes performance and determine the optimum pressure out of the three specified pressures, each of EG feeds is treated individually by NF and RO processes.

Step 2. The optimum pressure of 7 bar is determined at step1, in the second series of tests at the constant optimum pressure with three flow rates of 600, 720, and 840 L/h for three feed types, all the steps of the first series are reiterated aim to find the optimum flow rate.

Fouling is one of the most important phenomena in membrane use. Membrane fouling occurs in increasing transmembrane pressure (TMP) or reduction of membrane flux during constant pressure. This phenomena can be reversible or irreversible. If the reversible fouling occurs backwash and chemical cleaning are ways to remove foulants on membrane surface. Fouling can be classified into: Particulate/colloidal fouling, Organic and inorganic fouling, and Biofouling [16, 17]. Organic loading rate (OLR), dissolved hydrogen concentration, hydraulic retention time (HRT), sludge retention time (SRT), salinity, cations in feed, temperature, and nutrients are Important factors related to the fouling [18-24]. The following relation is used to calculate membrane fouling:

$$FR\% = \frac{[\left(\frac{\Delta p}{\mu \times pf_{ww}}\right) - \left(\frac{\Delta p}{\mu \times pf_{wi}}\right)]}{\left(\frac{\Delta p}{\mu \times pf_{wi}}\right) \times 100}$$

Where:

FR% is the Percentage of membrane fouling.

Δp is membrane input and output pressure difference.

μ is the feed viscosity in kg/m.s.

pf_{ww} is permeated flux of twice-distilled water for clear membrane in l/m^2.h.

Another key parameter is membrane flux. It defines the permeate flow divided by the total membrane surface area. The membrane flux can be calculated by following equation [25, 26]:

$$J = \frac{1}{A} . \frac{dv}{dA}$$

Where

J is membrane flux, and

A is membrane surface area.

The other important parameter that has been investigated in this study is Rejection. The rejection or removal of EG has been calculated by following equation [25]:

$$R\% = \frac{CF - CP}{CF} . 100$$

Where

R% is rejection (removal) percentage,

CF is feed contaminant concentration, and

CP is permeate contaminant concentration.

3.0 RESULTS AND DISCUSSION

The results of experiments on the removal of EG from industrial wastewater of the Fourth Refinery and synthesized wastewater are investigated at different pressures and flow rates using NF and RO membranes. In each series of experiments, the effect of pressure and flow rate on the water flux and EG removal rate are plotted and discussed.

In addition, the membranes' performance at different pressures and flow rates have been investigated.

3.1 The Effect of Pressure and Feed Type on Water Flux

The applied pressure range in step1 is 5 to 7 bar. It has been seen in Figure 2, flowrate increased by increasing in pressure. In the comparison of industrial and synthetic samples, the flow rate of the industrial sample is lower than the synthetic sample, which is due to the high concentration of industrial wastewater. Figure 2 shows the effect of pressure and feed type on water flux at the NF and RO process.

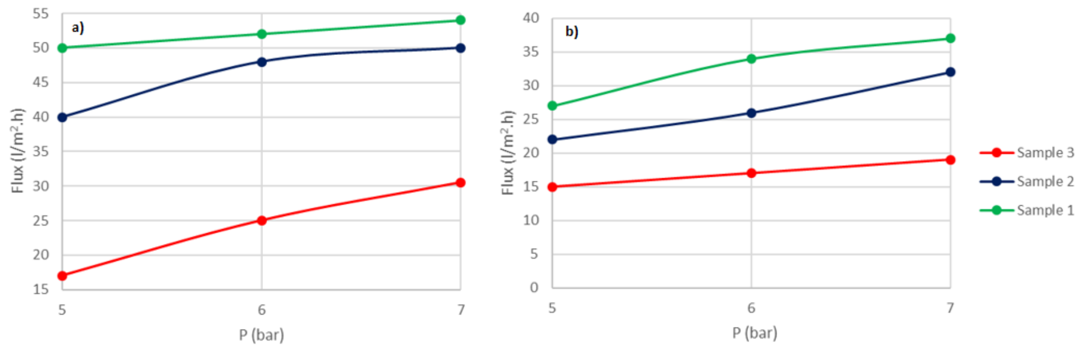

Figure 2. The effect of pressure and feed type on water flux at a) NF and b) RO process. Sample 1: twice-distilled water, Sample 2: synthetic sample, Sample 3: industrial sample

3.2 Effect of Pressure and Feed Type on the Concentration of Treated Water

Figure 3 show the effect of pressure on the concentration of treated water by NF and RO respectively. It has been seen pressure increase, decreases the EG content of treated water, which means increasing the pressure is a suitable solution to increase treatment performance. Due to the low concentration in the synthetic sample, the treatment performance for sample 2 is higher than sample 3.

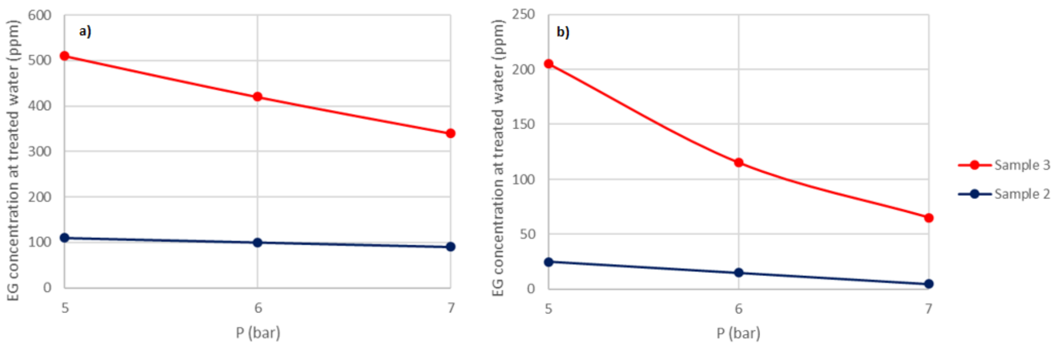

Figure 3. Effect of pressure and feed type on the concentration of treated water by a) NF and b) RO process

3.3 Effect of Pressure on EG Removal Percentage

Results of experiment shows that by Pressure increase, the percentage of EG removal will increased, which can be seen in the Figure 4. The highest treatment rates are observed for both feeds at the pressure of the highest applied pressure (7 bar). Figure 4 shows the effect of pressure on EG removal percentage by a) NF and b) RO process.

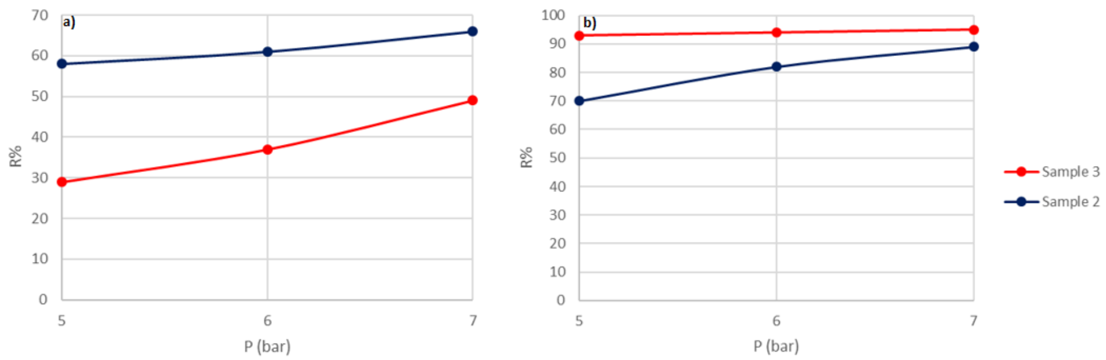

Figure 4 Effect of pressure on EG removal percentage by a) NF and b) RO process

3.4 Effect of Feed Type on EG Removal Percentage at Different Pressures

The amount of EG removal depends on the concentration of EG in the feed. As shown in Figure 5, for the lower concentration of EG in the feed, the separation rate is higher. The separation rate for the synthetic sample is higher than the industrial sample and the separation percentages are 70% and 55%, respectively. In addition, feed concentration plays the main role in membrane fouling. The higher concentration of EG in feed causes greater fouling and it causes less removal percentage. Figure 5 shows the effect of feed type on EG removal percentage by NF and RO process at different pressures.

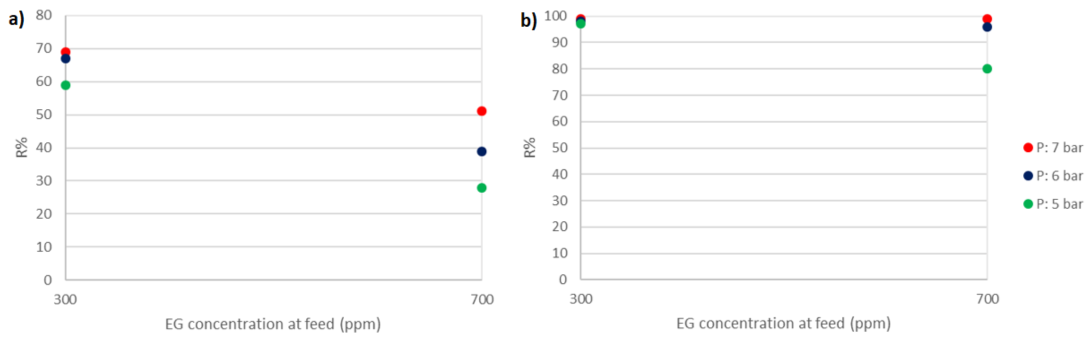

Figure 5 Effect of feed type on EG removal percentage by a) NF and b) RO at different pressures

3.5 Effect of Feed Type and Flowrate on Water Flux

In the second step of the study, the experiments at optimal pressure (7 bar) determined in the first step were performed in different flowrates (600, 720, 840 l/h) to determine the effect of flowrate on separation flux. Experiments were performed as in the first step for three feeds: twice-distilled water, synthetic wastewater with a concentration of 300 ppm EG and wastewater from the fourth refinery of the South Pars Gas Complex (SPGC) with a concentration of 700 ppm. As shown in Figure 6, the flux also increases with increasing flowrate. Figure 6 shows the effect of feed type and flowrate on water flux at NF and RO processes.

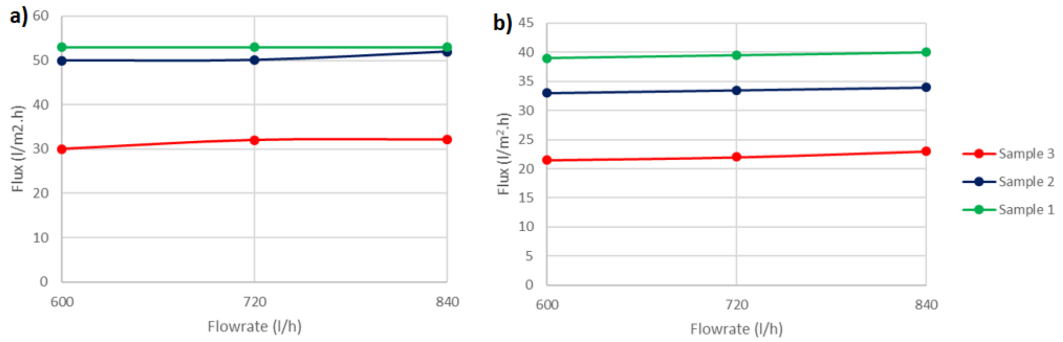

Figure 6 Effect of feed type and flowrate on water flux at a) NF and b) RO process

3.6 The Effect of Feed Type and Flowrate on the Concentration of Treated Water

To find the optimal amount of flowrate, the removal of EG from the feed in different flowrates was compared for synthetic and industrial samples. In this study, it is observed that 720 l/h of flowrate is the optimal amount for the separation of EG from the wastewater. Figure 7 shows the effect of flowrate on amount final EG concentration at NF and RO process.

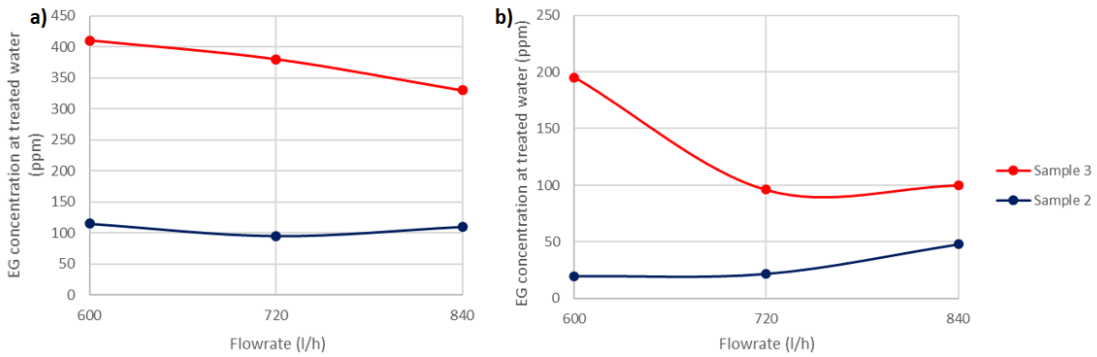

Figure 7 The Effect of feed type and flowrate on the concentration of treated water by a) NF and b) RO process

3.7 The Effect of Feed Type and Flowrate on the EG Concentration of Treated Water

Figure 8 shows the effect of flow rate on EC removal percentage by RO, Figure 9 shows the effect of feed type and flowrate on the EG concentration of the feed at NF and RO process. It can be seen from these graphs that increasing flowrate does not always increase the separation performance. As shown in the figures, 720 l/h flowrate is the optimal flowrate for both synthetic wastewater and industrial wastewater.

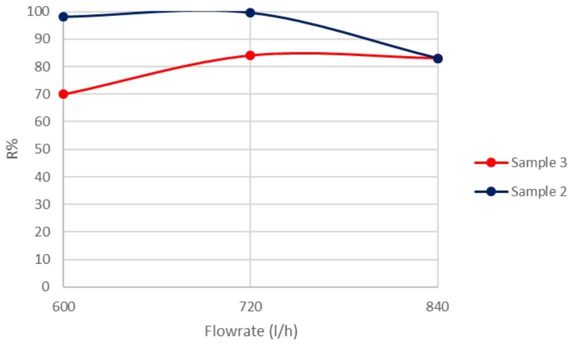

Figure 8 Effect of flow rate on EG removal percentage by RO

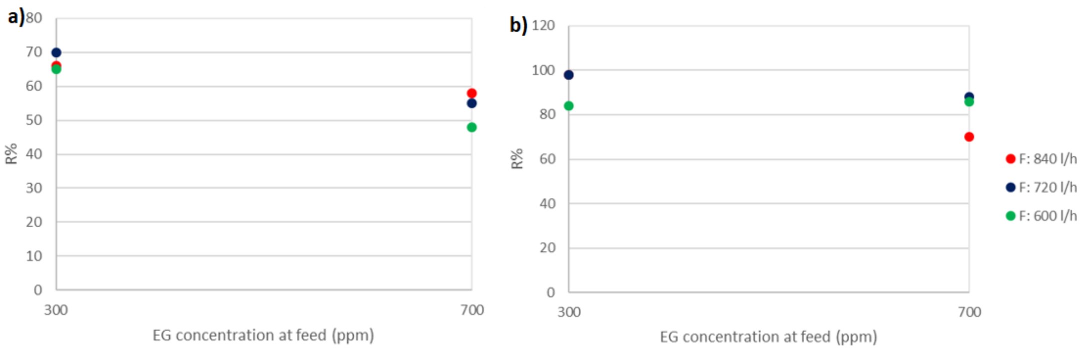

Figure 9 The effect of feed type and flowrate on the EG concentration of the feed at a) NF and b) RO process

3.8 Comparison of Membrane Fouling in Different Pressures

One of the important parameters in the membranes science study is membrane fouling. The following relation is used to calculate membrane fouling:

$$FR\% = \frac{[\left(\frac{\Delta p}{\mu \times pf_{ww}}\right) - \left(\frac{\Delta p}{\mu \times pf_{wi}}\right)]}{\left(\frac{\Delta p}{\mu \times pf_{wi}}\right) \times 100}$$

Where:

FR% is the Percentage of membrane fouling.

Δp is membrane input and output pressure difference.

μ is the feed viscosity in kg/m.s.

pf_{ww} is permeated flux of twice-distilled water for clear membrane in $l/m^2.h$.

pf_{wi} is permeated flux of twice-distilled water for unclear membrane in $l/m^2.h$.

Membrane fouling is an important factor in membrane maintenance. If the fouling rate is reduced, the separation quality gets better and the membrane lifetime becomes longer, therefore, the costs will be decreased. In the present study, the percentage of fouling has been calculated by the above relation in each experiment. Figure 10a shows the comparison of membrane fouling in different pressures. It has been seen that membrane fouling at the RO process is less than the NF process in all pressures. It is due to RO membrane is able to remove very small particulate such as monovalent ions, which is not able in NF, and these components can sweep in the rejection stream before scaling and fouling membrane surface. In similar study, Jacob has been find fouling in NF is stronger than RO [14]. In addition, has been found from the figure by increasing pressure, membrane fouling will increase. This increase is stronger for the NF process. This is because of increasing the TMP. As mentioned at introduction membrane fouling is related to the TMP directly [25, 26].

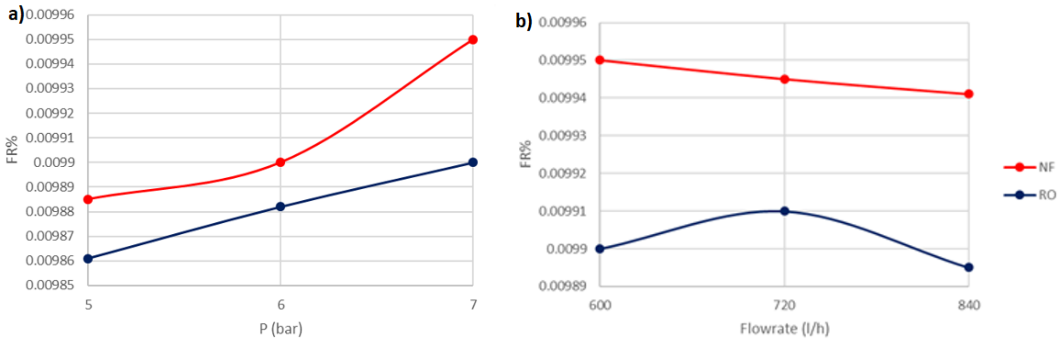

Figure 10 The comparison of membrane fouling in a) different pressures and b) different flowrates at NF and RO membranes

3.9 Comparison of Membrane Fouling at Different Flowrates

As mentioned earlier, the percentage of membrane fouling is one of the most important parameters in membrane science study. Figure 10b shows the comparison of membrane fouling in different flowmeters. It has been seen that membrane fouling at the RO process is less than the NF process in all flowrates. Furthermore, it has been found by increasing flowrate, membrane fouling will decrease.

3.10 Comparison of Flux Diagrams at Different Pressures at RO and NF

Figure 11a compares wastewater treatment flux for all three samples by NF and RO at different pressures. It is observed NF process has more flux than RO for each sample. It is because the RO membrane is more compressible than the NF membrane.

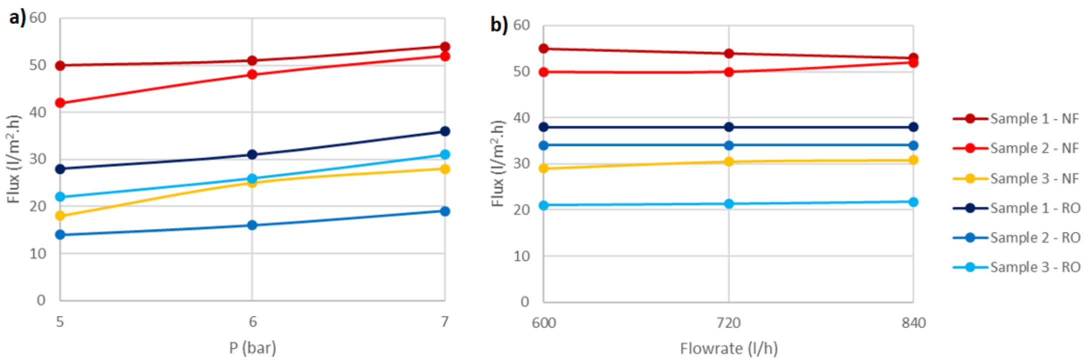

Figure 11 Comparison of flux diagrams at a) different pressures and b) different flowrates at RO and NF

3.11 Comparison of Flux Diagrams at Different Flowrates at RO and NF

Figure 11b compares wastewater treatment flux for all three samples by NF and RO at different flowrates. It has been found NF process has more flux than RO for each sample. As discussed above, it is because the RO membrane has more compression than the NF membrane.

4.0 CONCLUSION

In this study, the experiments were performed in two steps. Each step consists of 18 series of tests. In Step 1 three samples were treated at three different pressures. Sample 1 is twice-distilled water, sample 2 is synthetic wastewater containing 300 ppm EG, and sample 3 is industrial wastewater containing 700 ppm EG from the fourth refinery of SPGC. In step 1, the effects of pressure on treatment parameters such as flux, and EG removal, and membrane fouling have been analyzed. In addition, the optimum pressure for the process has been found in step 1. 7 bar is the optimum pressure. In Step 2 three samples were treated at three different flowrates. The pressure in this step is the optimum pressure, which has been found in step 1. In step 2, the effects of flowrate on treatment parameters and membrane performance like fouling have been analyzed. In addition, the optimum flowrate for the process is found in step 2. The optimum flowrate is 720 l/h.

As observed, for the RO process, the EG removal from synthesized and industrial wastewaters at different pressures are 80% and 99%, respectively, and for the NF process, these are 60% and 80%, respectively, which indicates membrane technologies are suitable choices to treat industrial wastewater containing EG. In addition, it has been observed RO has better performance than NF. Furthermore, it has been observed RO membrane has less fouling than the NF membrane, which means the RO process has more lifetime and fewer operational costs. However, it should be noticed the flux of the NF membrane is higher than RO due to the high compression of the RO membrane. In the similar study Khachonbun investigates NF and RO process to remove glycol from wastewater [27]. She founds these processes are able to remove 90 to 98% of organic materials and due to the material of membranes the rejection percentage are different. It has been found RO has more rejection rate, however NF achieved more membrane flux.

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