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Assessment of the Pressure Driven Membrane for the Potential Removal of Aniline from Wastewater

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ABSTRACT

The vast utilizing of aniline in diverse industrial applications makes it predominantly recognized in the eco-geological system. This work investigated the feasibility of reverse osmosis (RO) and nanofiltration (NF) membranes for the removing of aniline from wastewater. The performance of the TFC spiral wound membrane was examined with different operating parameters. The effect of feed concentration (10–200 mg/l) and operating pressure (1–4 bar) on flux and aniline rejection were explored. Additionally, the fouling test for the adopted membranes was conducted for 20 h using NaOH as cleaning agent. The results revealed that a high rejection ratio at noticeable low operation pressure was achieved by using TFC membranes for both of the RO and NF technologies. The maximum aniline rejection was 99.8% and 93.25% under a 1 bar pressure and the concentration of feed 10 mg/l for the RO and NF membranes, respectively. These rejection ratios correspond to the permissible concentration of aniline in the wastewater. The water flux obtained was 6.33 and 13.5 LMH for reverse osmosis and nanofiltration membranes, respectively. The augmentation of operation pressure resulted in decreasing of rejection and rising of the flux. The fouling test showed a reduction in flux of about 0.92 and 4.35% for RO and NF membranes, respectively, from its initial value before membrane cleaning. The results also demonstrated that the reverse osmosis membrane is better than the nanofiltration membrane in terms of removal efficiency.

Keywords: nanofiltration, reverse osmosis, aniline, organic fouling, low pressure.

INTRODUCTION

Among many organic components, aniline and its derivatives are well-recognized raw materials for many industrial processes [Tan et al., 2022]. Commonly, It can be found as intermediates in the manufacturing of pharmaceuticals and explosives, herbicides and pesticides, and as a solvent in perfumes, varnish and resins [Yang et al., 2019], dyes and pigments, also as accelerators of rubber production [Li et al., 2016; Benito et al., 2017; Abdel-Rahman et al., 2020]. The high degradation and solubility of aniline makes it one of earnest pollution source, especially in case of direct reject to the water sources. Aniline is a toxic to human, serious health problem might cause by ingestion, inhalation, or contact with the skin have been reported [Dakhil et al., 2021]. Laboratory studies refer that aniline give rise to diet of animals and might may affect the liver, kidneys, and adverse effects in the blood. Also cancer and spleen damage may be diagnosis as a long term effect of exposing to aniline [Zhang et al., 2022]. Moreover, during the traditional water treatment, the benzoic group in aniline components has the ability to react with chlorine which added through the sterilization step to form chlorinated by products. The later has high stability than aniline which may cause serious poisoning threats [Gonsior et al., 2014; Maguire-Boyle and Barron, 2014]. Therefore, aniline levels in industrial wastewater should be carefully controlled, that can be achieved by treating the industrial wastewater before discharge [Chaturvedi, 2022].

Removing aniline and organic compounds from wastewater have been subject of several studies. Biological treatment for aniline removal has been conducted [Li et al., 2010]. In addition, considerable studies were performed to develop a physical or chemical treatment such as adsorption [Abbas and Hussien, 2017; Al-Jubouri et al., 2023], electro-fenton advanced oxidation [Abbas and Abbas, 2022], ligand exchange, and emulsion liquid membrane [Majeed and Mohammed, 2016; Mohammed et al., 2018]. These traditional processes have several drawbacks which promote researcher develop technology taking into consideration the efficiency of separation, complexity of operation and capital cost. The separation efficiency of traditional methods fades with low aniline concertation wastewater that push forwards to find alternative methods for separation [Ren et al., 2014].

Membrane separation have been proposed as one of the less energetically separations process that can be used to remove aniline from wastewater. Membrane filtration particularly [Al-Alawy and Al-Ameri, 2017], reverse osmosis, and forward osmosis (FO), has been considered for the removal of organic compounds [Osorio et al., 2022], including aniline, from water [Cui et al., 2016]. The pressure-driven desalination technique of reverse osmosis and nanofiltration selectively transports water over salt utilizing a semipermeable membrane [Wang et al., 2021]. The use of NF and RO techniques has several benefits, including continuous operation, the absence of phase changes, high efficiency [Jeong et al., 2021], ease of operation, no need to add chemicals, and good stability [Gherasim and Mikulášek, 2014]. The water flux (J_w) equation in NF and RO membranes is [Hadadian et al., 2021]:

$$J_w = A\ (\Delta P - \Delta\pi) \tag{1}$$

where: A – the pure water permeability coefficient,
ΔP – the operating pressure difference,
$\Delta\pi$ – is the osmotic pressure difference.

The flux of solute (J_s) across the membrane is given by the following equation [Chougradi et al., 2021]:

$$J_s = B\ (\Delta C) \tag{2}$$

where: B – the solute permeability coefficient,
ΔC – the concentration gradient.

However, the accumulation and deposition of undesirable elements inside or on the surface of the membrane are commonly referred to as membrane fouling. This phenomenon considers the key limitation to the efficient use of NF and RO membrane processes. Fouling reduces membrane performance, sustainability, and economic feasibility and, as a result, shortens membrane life. Dissolved particles, partially soluble organic, inorganic macromolecules, and biological microorganisms could be deposited. Among the several possible foulants found in natural and waste streams, dissolved organic matter is the most obstinate [Alsawaftah et al., 2021].

Hidalgo et al., 2014, investigated two flat sheet TFC nanofiltration membranes NF97 and NF99HF from Dow Chemical to remove aniline from wastewater. The effective membrane area was 0.003 m^2. The rejection at feed concentration of 10 ppm and applied pressure ranges from 10 to 30 bar was 60–80 % and 10–20 % for NF97 and NF99HF, respectively. Cui et al., 2016, compared the efficiency of FO and RO systems for aniline removal from wastewater. Five different reverse osmosis membranes were used, and the rejection was 57.3, 52.6, 61.9, 56.6, and 66.6% for Matrimid TFC, PESU TFC, sPPSU TFC, Filmtech BW30-4040, and UTC-70UB, respectively with the concentration of feed 1000 ppm and operating pressure of 10 bar.

The current study highlights the performance of reverse osmosis and nanofiltration membranes processes for the removal of aniline from wastewater utilizing TFC polyamide spiral wound element membranes. The effect of the key parameters specifically operating pressure and feed solution concentration on the removal of aniline were examined. The process efficiency was assessed in terms of water flux and rejection. Furthermore, the membranes performance was evaluated by testing the effect of fouling on the water flux for 20 h.

EXPERIMENTAL WORK

The aniline used in this work was purchased from (Sigma-Aldrich, USA, 99%), sodium hydroxide NaOH (Applichem GmbH, Germany, 98%), and distilled water (1.65 μS/cm conductivity) was used to prepare the aniline solutions. Two types of TFC spiral wound membranes were used, nanofiltration membrane (AXEON NF4-1812, USA) and reverse osmosis membrane (Aventura, Burton$^+$ 1812-75, India). Each membrane with a membrane effective area of 0.39 m^2.

The set up utilized to performed the experiments is shown in Figure 1. The system consists of a feed vessel of 5 L capacity, this vessel provides

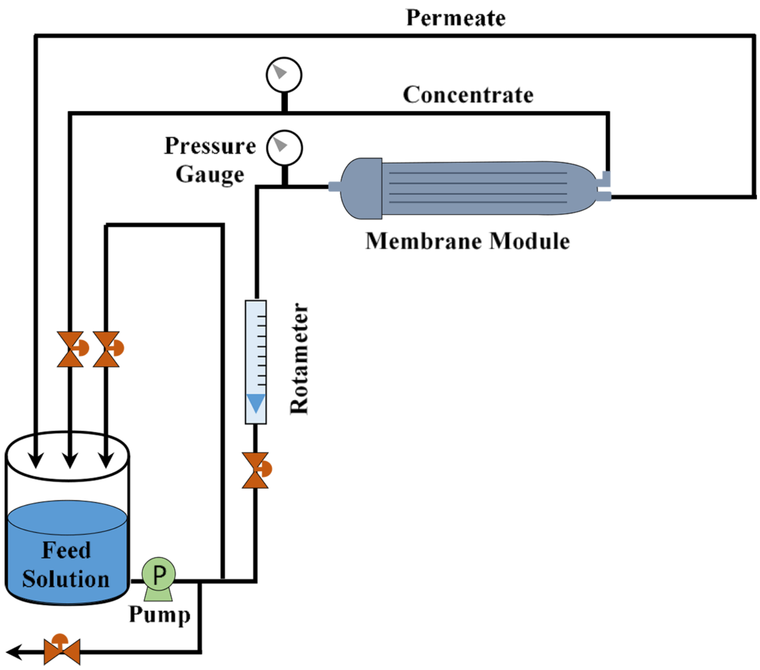

Figure 1. Lab-scale of RO and NF Process

the aniline solution to the system. A diaphragm pump (HEADON, Taiwan, HF-9050) was used to circulate the feed solution. Two pressure gauges were used to control the pressure of the feed and concentrate sides. Rotameter was used to measure the flow rate of the feed solution. To maintain a constant feed concentration, the permeate and concentrate stream was recirculated to the feed tank. The feed solution concentrations were tested in the range of 10–200 ppm, and the operating pressure was investigated in the range of 1–4 bar.

The temperature was kept constant at 25 °C, the flow rate of the feed solution was kept constant at 30 L/h. The fouling experiment was performed at 200 ppm of feed solution concentration with a pressure of 1 bar for 20 h. After 10 h the membrane was washed with NaOH solution of pH = 10 and after that washed with distilled water. Samples were taken periodically to measure Aniline concentration using a UV-spectrometer (Thermo Electron Corporation, GENESYS 10 UV, USA) at a wavelength of 280 nm.

The water flux (J_w) can be calculated from the following equation [Aldahlaki et al., 2020; Salih and Al-Alawy, 2022a]:

$$J_w = \frac{\Delta V}{A_m \, \Delta t} \tag{3}$$

where: ΔV – the volume of water flow from feed to permeate side, A_m – the membrane active area, and Δt – the experiment time.

The rejection (R) of aniline can be calculated from the following equation [Salih and Al-Alawy, 2022b]:

$$R = \left(1 - \frac{C_p}{C_F}\right) * 100\% \tag{4}$$

where: C_p – the aniline concentration in permeate, and C_F – the aniline concentration in the feed.

The solute permeability coefficient (B) was calculated by the following equation [Han and Chung, 2014]:

$$B = \frac{A\,(\Delta P - \Delta\pi)\,(1 - R)}{R} \tag{5}$$

where: A – the water permeability coefficient, R – the solute rejection, ΔP – the applied pressure difference, and $\Delta\pi$ – the osmotic pressure difference.

RESULT AND DISCUSSION

The effect of pressure (1–4 bar) on the water flux and rejection of aniline for different aniline feed concentrations (10–200 mg/l) are shown in Figures 2 to 5 for RO and NF membranes, respectively. Figures 2 and 4 show the impact of pressure on flux for nanofiltration and reverse osmosis membranes, respectively. The experimental results demonstrated that the flux increase with the raise in pressure. Where by increasing the pressure from 1 to 4 bar at C_F = 10 ppm, the water flux augmented from 6.33 to 20.83 LMH for the RO system as in Figure 4. Correspondingly for the NF configuration, the water flux remarkably risen from 13.5 to 49.33 LMH by increasing the system pressure from 1 to 4 bar at C_F = 10 ppm as can be seen in Figure 2. According to Eq. (1) the flux is directly proportional to the system pressure. Also, convective transport plays an important role when the pressure increases. At constant feed concertation, consequently constant osmotic pressure of the feed solution, the increase of the system pressure resulted in augment of the driving force for transfer.

On the other hand, the results also show that the raise in the feed concertation from 10 to 200 ppm resulted in a flux decline of 31.6% for the RO membrane at P = 1 bar, as shown in Figure 4. Also the flux reduced by 14.82% by increasing the concentration of feed from 10 ppm to 200 ppm for the NF membrane at P = 1 bar, as in Figure 2. This results from an increase in the feed solution's osmotic pressure, which lowers the driving force for transfer, according to Eq. (1) the flux is directly proportional to the driving force. This behavior comes in line with the results obtained by Hidalgo et al., 2014.

Figures 3 and 5 illustrate the effect of pressure on aniline rejection for nanofiltration and reverse osmosis membranes. The experimental results clarify that the rejection decreased as the pressure increased. The rejection of aniline in NF membrane for C_F = 10 mg/l is 93.25% at operation pressure of 1 bar and reduced to 65% when the pressure increased to 4 bar (reduction of 30.29% in rejection). For the RO membrane, the reduction in rejection is 17.58% from 1 to 4 bar of pressure rise. The behavior of aniline rejection may be attributed to the fact that the bonds

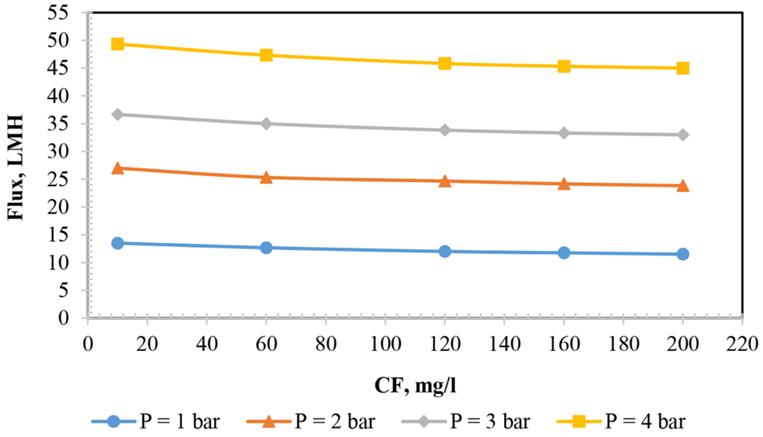

Figure 2. The effect of pressure on water flux for different feed concentration for NF membrane (T = 25 °C, Q_F = 30 L/h)

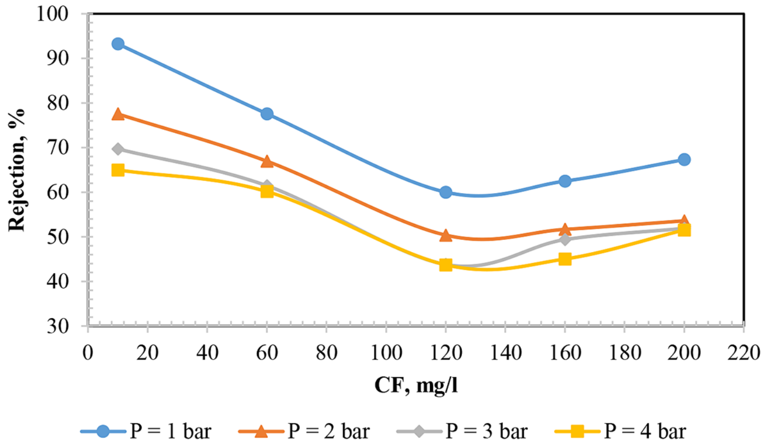

Figure 3. The effect of pressure on rejection for different feed concentration for NF membrane (T = 25 °C, Q_F = 30 L/h)

between the organic molecules restricts and compressed the solubilized aniline molecules. Unlike the behavior of inorganic salt, aniline which is an organic substance does not depend on the balance of charges on both sides of the membrane. In addition, aniline has high solubility in water in the range of concentrations used in the study, consequently, aniline molecules are carried by water and across the membrane. That could be promoted by the small diameter of the aniline molecule (stokes radius is 2.12 A°) [Ben-David et al., 2006]. Also, another cause that could stand for the aniline permeability through the membrane, is due to the sorption isotherm of aniline in polyamide membrane, which may resembles the behavior of water transfer across the membrane. This is one of the hypotheses to explain the permeability of water through the membrane and the salts do not [Cao et al., 2022]. Figures 6 and 7 show the effect of organic fouling on NF and RO membranes for 10 h and then for a further 10 h after chemical cleaning with NaOH solution. It is notable that the water flux declined with time due to the accumulation of organic fouling. The initial water flux for NF and RO membranes were 11.5 and 4.33 LMH, respectively, and reduced to 8.83 and 3.9 LMH during the first 10 h of operation (23.22 and 9.93% reduction in flux). Thereafter, a chemical cleaning with NaOH solution was conducted to the membranes resulted in advancing of the flux to values of 11 and 4.29 LMH for NF and RO membranes, respectively. The enhancement of the flux could not resuscitation the flux to its initial status with a reduction of about 0.92 and 4.35% for reverse osmosis and nanofiltration membranes, respectively. This reduction after cleaning is due to irreversible fouling. A similar pattern of behavior was reported by Lin, 2017.

In order to emphasize the preferable performance of aniline removal for the adopted types

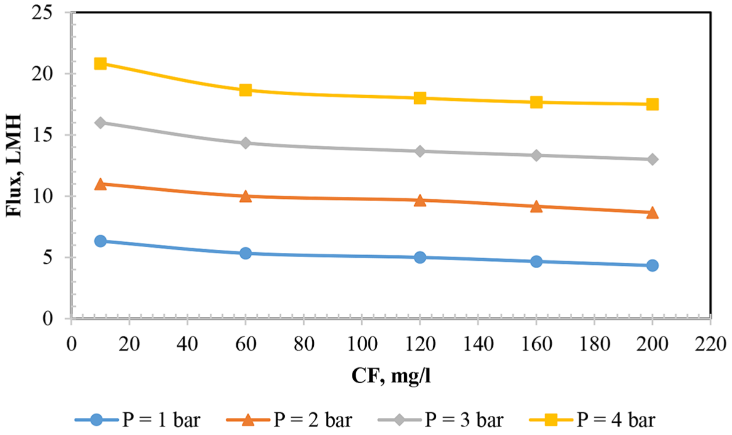

Figure 4. The effect of pressure on water flux for different feed concentration for RO membrane (T = 25 °C, Q_F = 30 L/h)

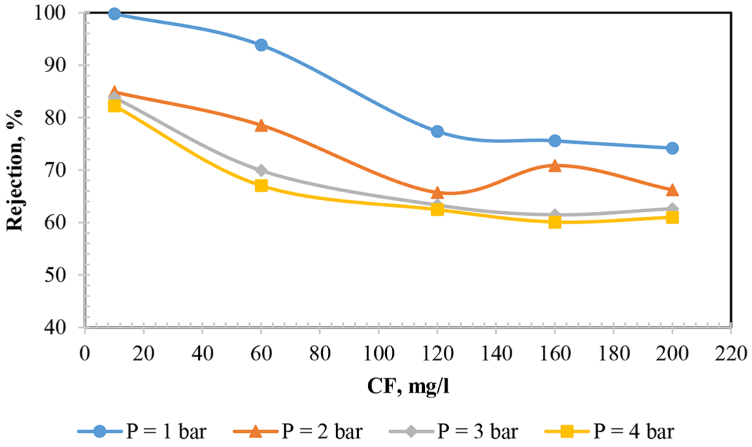

Figure 5. The effect of pressure on rejection for different feed concentration for RO membrane (T = 25 °C, Q_F = 30 L/h)

of membranes, a comparison between NF and RO system was explored as shown in Figures 8 and 9. The results illustrated in the Figure 8 indicate that the flux obtained with RO is less than that of NF. At initial concentration of 10 mg/l and operation pressure of 1 bar, the water flux was 6.33 LMH with RO membrane, and its increased about 113.27% with NF membrane. The amount

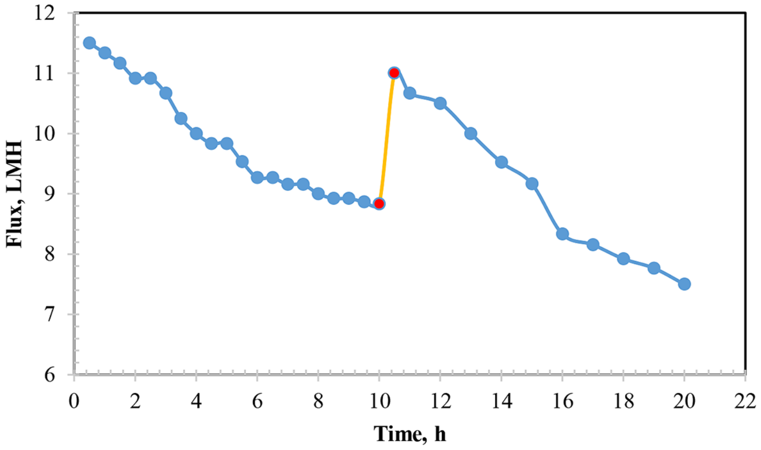

Figure 6. The effect of fouling on flux for NF membrane (P = 1 bar, C_F = 200 mg/l, T = 25 °C, Q_F = 30 L/h)

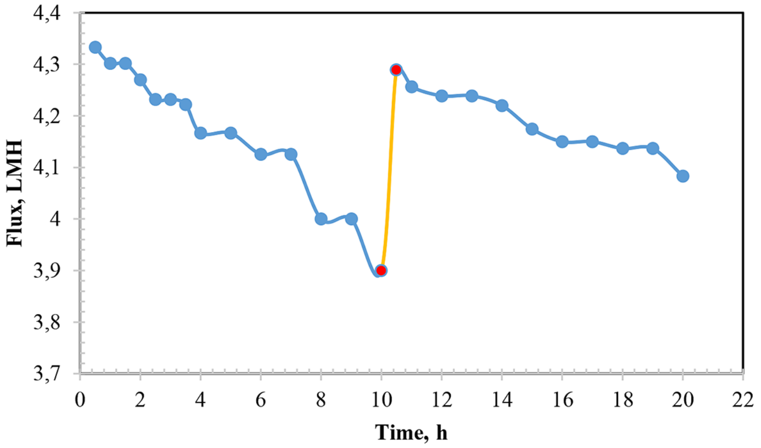

Figure 7. The effect of fouling on flux for RO membrane (P = 1 bar, C_F = 200 mg/l, T = 25 °C, Q_F = 30 L/h)

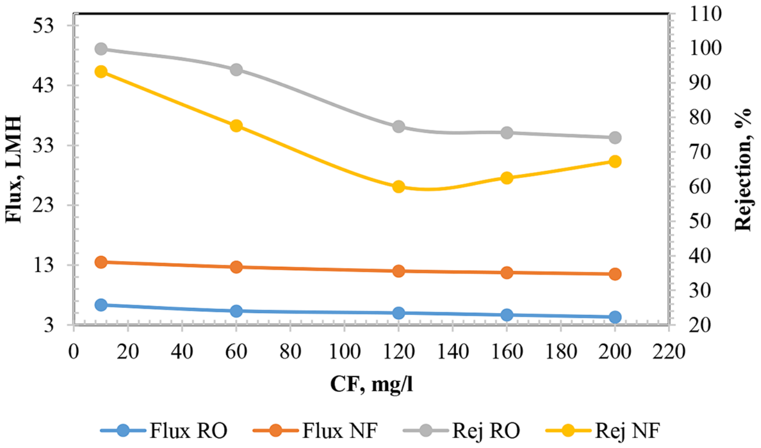

Figure 8. Comparison between NF and RO membranes for different feed concentration (P = 1 bar, T = 25 °C, Q_F = 30 L/h)

of reduction in water flux between the NF and RO configuration remained almost for all range of feed concentration. However, the rejection for RO were higher than that obtained with NF for all range of concentration. Where with RO configuration, the rejection scores a high value of 99.8%, and its minimized to 93.25% when NF is used, that values obtained at concentration of 10 ppm.

Furthermore, Figure 9 pointed that the rising in operation pressure priority of NF over the RO in terms of water flux. When operating at 4 bar, with a 10 mg/l initial feed concentration the reduction in water flux was about 57.77% when the configuration switch from NF to RO. Obviously, that reduction in flux minimized when the operation pressure decreased. Again, the rejection for RO obtained at different operation pressure was analogous to that obtained with different feed concentration, which is remarkably higher than that of NF. The disparity between the flux and removal performance of RO and NF could be attributed to the water and solute permeability coefficient for the two membranes. The membrane permeability coefficient of pure water was determined for reverse osmosis and nanofiltration membrane from Eq. 1 by plotting pressure versus water flux for deionized water feed solution ($\Delta\pi = 0$, i.e., $J_w = A\Delta P$) and the slope of the line represents the permeability coefficient of the membrane. It can be noticed that the permeability coefficient of pure water for RO and NF membranes are 5.48 and 12.13 LMH/bar, respectively (i.e. RO permeability coefficient is lower than that of NF by 54.82%). The solute permeability coefficient has been calculated from Eq. 5 and it is found to be 3.056×10^{-9} and 2.453×10^{-7} m/s for RO and NF, respectively. In other words, the RO membrane solute permeability is lower than that of the NF membrane by 98.75%, which leads to high rejection rates for aniline in the RO membrane.

COMPARATIVE ANALYSIS

Aniline is a common pollutant identified in effluent during textile and dyeing wastewater treatment operations that has gained attention due to its high environmental concern. For instance, in Guangdong Province, aniline (<10 mg/l) was discovered in the secondary biological effluent of a significant woolen textile printing and dyeing factory [Zhang et al., 2021]. The ultimate goal for divers unites of water treatment is to frustrate the impact of the pollutant by reducing its concentration to a certain limit. According to Zhang et al., 2022, the aniline in wastewater should not exceed 1ppm. The current research approved that the aniline concentration can be reduced to about 0.67 ppm at low pressure of 1 bar for a feed concentration of 10 ppm, the rejection was 93.25% by using nanofiltration technology. A lower concentration can be reached by using reverse osmosis technology, where the permit concentration hit a concentration of 0.02 ppm at a low operation pressure of 1 bar, with a high rejection of 99.8%.

The previous work of Hidalgo et al., 2014 demonstrated that nanofiltration technology can be used for the treatment of aniline where two types of NF membrane were used (NF97 and NF79HF). The rejection obtained at a relatively high pressure of 10-30 bar was 60-80% for NF97 membrane and 10-20% for the NF79HF

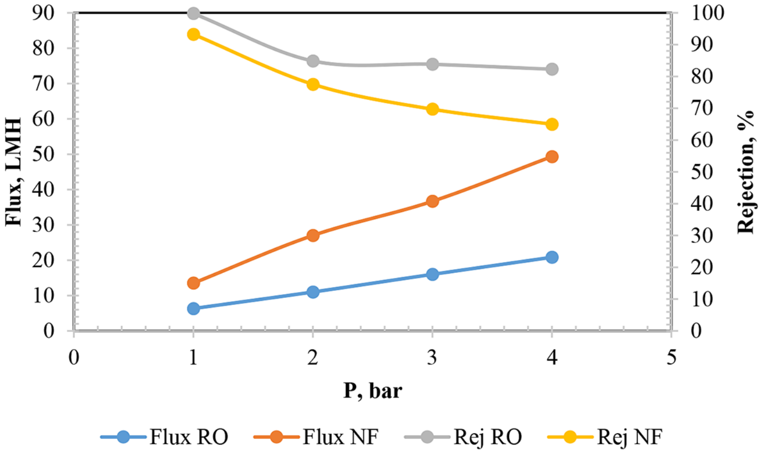

Figure 9. Comparison between NF and RO membranes for different pressures (C_F = 10 mg/l, T = 25 °C, Q_F = 30 L/h)

Table 1. Results of NF and RO membranes for different feed concentration with operating pressure of 1 bar

Feed concentration, ppm	Permeate concentration for NF membrane, ppm	Permeate concentration for RO membrane, ppm
10	0.67	0.02
60	13.44	3.71
160	60	39.1
200	65.4	51.64

membrane. Consequently, for the feed concentration of 10 ppm and this extreme operation pressure, the permit concentration was about 4-2 ppm for NF97 membrane and 9–8 ppm for the NF79HF membrane, which is higher than the acceptable limit for both adopted membranes.

Moreover, a higher feed concentration of aniline (100 ppm) has been treated by using nanofiltration and reverse osmosis technologies [Ben-David et al., 2006]. When the NF-200 membrane was used at a pressure of 20 bar, the rejection achieved was very low of 20%, which gave a permit concentration for aniline of 80 ppm. However, by utilizing the RO technology for the treatment of the same feed concentration, the rejection has been improved. The process has enable of achieving a 63% rejection and the permit concentration reduced to 37 ppm. On the other hand, in the current study, a higher concentration of 120 ppm was treated with NF technology, and the rejection was remarkable higher (60.2%) at very lower operation pressure of 1 bar in comparison to that of NF-200 membrane [Ben-David et al., 2006]. In addition, the rejection dramatically boosted to about 77.37% when the RO technology at same operation pressure of 1 bar.

It can be observed that a distinguish divergence in rejection established between the NF and RO membrane that have been used in the previous work and that adopted in the current work. That diversity in the performance may be contributed to the different in active membrane layer thickness and the synthesis materials, porous membrane layer thickness and structural properties, pore size of the membrane, water permeability, solute permeability. Also the low-pressure RO or NF membranes may include void spaces large enough to allow liquid water to flow convectively through the membrane [Crittenden et al., 2012]. Table 1 lists the results of NF and RO membranes for different feed concentrations with an operating pressure of 1 bar. It can be noticed that the results in Table 1 represent the permeate concentration of aniline from different feed concentrations for NF and RO membranes. According to these results, when the feed concentration is 200 and 160 ppm, it requires 3 stages of the membrane process to reach the permeate to the standard limit. For the feed concentration of 60 ppm, it requires 2 stages only.

CONCLUSIONS

The RO and NF membranes are effective processes for the removal of aniline from wastewater. The aniline concentration in the permeate has correspond with the standard limits at a 1 bar pressure and a concentration of feed of 10 ppm. The maximum removal efficiency for aniline is 93.25 and 99.8% at an operating pressure of 1 bar and a concentration of feed of 10 ppm for NF and RO membranes, respectively. The highest flux was obtained for the same concentration at high pressures, while the highest rejection was obtained for the same concentration at low pressures. RO membranes give better performance in the removal of aniline than NF, while for flux, vice versa. The fouling experiment indicates that there is reversible and irreversible fouling occurs, thus after membrane cleaning, the reduction in flux from its initial state of about 4.35 and 0.92% for NF and RO membranes, respectively.

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