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Potential of Nanofiltration Membrane in Groundwater Treatment for Drinking Water Resources

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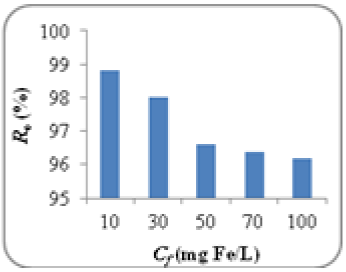
Graphical abstract



Abstract

Groundwater in Malaysia has become an alternative water resouces for daily needs. However, the presence of iron and manganese has been the major problem that caused the water unsuitable for drinking due to reddish colour and bad taste. Therefore, groundwater should be well treated from any hazardous metal before consumption. A dead-end stirred cell was used to investigate the ability of commercial NF membrane in removal of Fe to acceptable level for drinking water. Removal up to 99% for 10 mg/L iron solution at pH9.4 with low pressure of 2 bar was achieved. Further investigation for higher feed concentration is suggested in order to achieve permeate concentration below than 0.3 mg Fe/L. All findings indicated that nanofiltration is a promising technology for groundwater treatment.

Keywords: Ferrous iron; groundwater; nanofiltration

■1.0 INTRODUCTION

Membrane filtration processes in potable water production have increased rapidly over the past decade.[1,2] Microfiltration (MF) and ultrafiltration (UF) are employed to remove microparticles and macromolecules, which generally include inorganic particles, organic colloids such as microorganisms and dissolves organic matter (DOM).[3] Whereas, nanofiltration (NF) membranes have the potential to remove turbidity, microorganisms and hardness, as well as a fraction of the dissolved salts. NF membranes are potentially used for all kinds of water treatment including ground, surface, and wastewater or also used as a pretreatment for desalination.[4] NF offers several advantages such as low operating pressure, high flux, high retention of multivalent anion salts and organic molecular above 300 and low in operation and maintenance costs.

Numerous studies have been reported to investigate the performance of NF membranes in comparison to other removal methods. NF is a suitable method for the removal of a wide range of pollutants from groundwater, in view of drinking water production.[5] The major application of NF membranes is softening but it also applied for the combined removal of non organic matter (NOM), micropollutants, viruses and bacteria, nitrates and arsenic or for partial desalination. NF is a promising technology for arsenic removal from contaminated groundwater as it requires less energy than traditional RO membranes.[6] Polypiperazine membrane and the applied NF processes is successfully utilized for the treatment of hard and extremely hard brackish groundwater as they provided high percentage of hardness removal, high mineral fouling resistance and satisfactory permeate fluxes at low transmembrane pressures.[7] In addition, NF process has overcome technical challenges in comparison to conventional methods in removal of radionuclides that commonly contaminated the drinking groundwater sources in several regions in the Middle East and the Arabian Gulf.[8]

In Malaysia, the major groundwater users are Kelantan and Selangor. High level of iron and manganese that exceeds the allowable value for consumption has become the main problem as reported by Department of Minerals and Geoscience.[9] Presence of excess amount of metal such Fe and Mn may cause coloured water, rusty-brown stains on laundry, odour and bad taste of beverages.[10] In addition, their presence for long period may also cause deposits in pipes, pressure tanks or heaters that may lead to high cost of maintainence either for domestic or industrial usage. Heavy metal contaminations even at low doses and in a long period can cause kidney or liver damage and anemia. Due to these health impact, effort must be taken to purify and minimize iron content in groundwater as investigated by other researchers.[11-14] The World Health Organization, WHO suggested that iron and manganese concentrations in drinking water should be below than 0.3 mg/L and 0.1 mg/L, respectively.[15]

The aim of this research is to develop a filtration system for groundwater treatment that meet the standard of drinking water. In order to approach this aim, study on groundwater quality from selected areas such as Kelantan was conducted as well as investigation on the membrane performance for further improvement. Table 1 shows the parameters of groundwater from selected monitoring wells in North Kelantan that exceed the drinking water quality requirements set by WHO. These data briefly show that rejection of iron and manganese will be the main interests of the treatment process at the selected study area. As for preliminary study, synthetic groundwater with ferrous iron concentrations based on the reported data by Mineral and Geoscience Department, Kelantan were used to investigate the performance of NF membrane.

Table 1 Groundwater characteristic from selected monitoring well in North Kelantan

Parameter	Reported	Benchmark
Iron (mg/L)	0.7 - 94	0.3
Manganese (mg/L)	0.2 – 3.5	0.1
Sodium (mg/L)	0.2 - 1578	200
Chloride (mg/L)	335 - 1485	250
Total Dissolves Solids, TDS (mg/L)	2604 - 3188	1000
Turbidity (NTU)	11 - 2730	5

(*Source*: Minerals and Geoscience Department of Kelantan, Malaysia)

2.0 EXPERIMENTAL

2.1 Materials

A commercial NF membrane denoted as TFC-SR3 was used throughout the experiments. The polyamide flat sheet membrane was supplied by Sterlitech Corp, USA and manufactured by Koch, USA with 200 molecular weight cut off (MWCO). All chemicals, solvents and reagents used were of analytical grade with high purity. Ultra-pure water with conductivity less than 1µS/cm was used to prepare artificial groundwater with ferrous iron at concentration of 10-100 mg/L by using $FeCl_2.4H_2O$ (HmbG® Chemicals). Ferrous iron reagent powder (HACH Permachem USA) was used to determine the content of Fe^{2+} in permeate for each filtration.

2.2 Characterizations

The applied membranes were characterized to identify the characteristics as shown in Table 2. Pure water permeabilty was conducted at operating pressure 1-5 bar using ultrapure water at room temperature and with a stainless steel dead-end stirred cell (Sterlitech Corporation, WA, Model HP4750) that houses a 49 mm diameter flat membrane sheet with an effective area of 14.6 cm^2. Membranes were immersed in ultrapure water and kept for overnight before compacted at 5 bar for 30 min prior to use. Contact angle measurements were done using a static sessile drop method by goniometer (Rame-Hart, Model 290) with three series of measurement at three different locations. As for salt rejection, NaCl and $MgSO_4$ were used at 100 mg/L and applied membrane pressure at 2 bar.

Table 2 Characterization of applied membrane

Characteristic	TFC-SR3
Pure water permeability ($L.m^{-2}.h^{-1}$)	4.2
Contact angle (°)	47
NaCl Rejection, 100 mg/L (%)	46.5
$MgSO_4$Rejection, 100 mg/L (%)	92.7

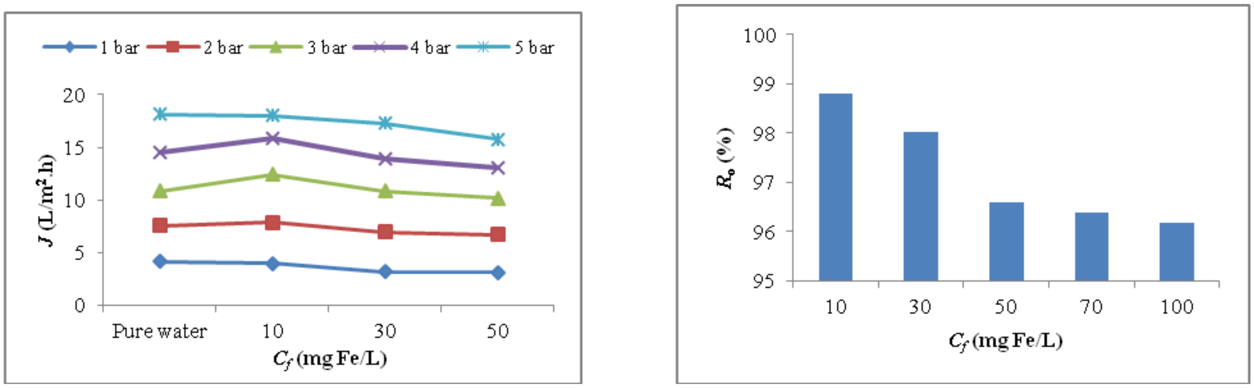
2.3 Filtration

Filtration experiments were performed to investigate the performance of applied membranes in terms of flux and rejection at various transmembrane pressure, feed concentration and feed pH. A bench-scale dead-end NF setup was used and comprises of a nitrogen gas tank, 2000 mL reservoir tank, 300 mL stainless steel stirred cell and a precision balance (Sartorious AG, Germany, Model AX6202) connected to a data acquisition personal computer. Experiments were conducted for 1.5 to 2.5 hr depend on the operating pressure due to collect permeate for immediate analysis of ferrous iron using spectrophotometer (HACH, Model DR3900). Measurement on fouling tendency will be considered for future works.

3.0 RESULTS AND DISCUSSION

3.1 Influence of Feed Concentration

In 2012, iron in groundwater for North Kelantan that was reported by Minerals and Geoscience Department are in the range of 0.7 to 94 mg/L. Therefore, synthetic groundwater for this study was prepared at this range by using $FeCl_2.4H_2O$ with objective to study the effect of feed concentration against flux and rejection. Ultra-pure water was used in the preparation of synthetic groundwater in order to investigate the ability of TFC-SR3 membrane to remove ferrous iron to the acceptable value set by WHO. Figure 1(a) presents permeate flux from feed solution at concentration in the range from 10 to 50 mg/L at operating pressure from 1 to 5 bar. It shows that the permeate flux has a linear dependence for all of feed concentrations and were slightly decreased from low to higher concentration. This behavior may be due to increased effect of concentration polarization at higher feed concentration, which may increases intermolecular repulsion and thus reduce the solvent permeability and lead to permeate flux decline. Figure 1(b) shows the observed rejection of Fe according to feed concentrations in the range from 10 to 100 mg/L at low applied pressure. The observed rejection decreased with increasing feed concentration and complied with other studies.[16] Results show that high rejections with more than 96% were obtained for all of the studied concentrations and indicated that the membrane has good separation properties. However, in order to reach acceptable value of Fe in drinking water therefore treatment using lower concentration at 10 and 30 mg/L were preferable. This is due to rejections were more than 97% which resulted that ferrous iron concentrations in permeate were much lower than toxicity level.



(a) (b)

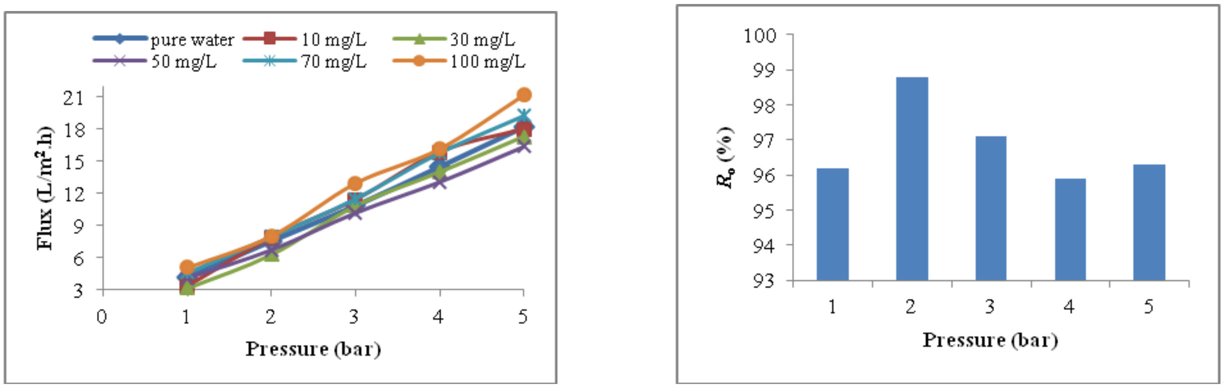
Figure 1 Effect of feed concentration on (a) permeate flux of TFC-SR3 membrane at various operating pressure and (b) rejection of Fe(II) ions by TFC-SR3 membrane at applied pressure of 2.0 bar and pH6.8

3.2 Influence of Applied Pressure

Figure 2(a) shows positive linear relationship between pressure and permate flux. The increase of applied pressure leads to strong increase in permeate flux as reported by other researchers.[6] These results present that TFC-SR3 membrane obeys the Darcy's law which describes the flux (J, L·h^{-1}m^{-2}) as a function of permeability (L_p, L·h^{-1}m^{-2}bar^{-1}) and applied transmembrane pressure (ΔP, bar) taking the osmotic pressure difference between feed and permeate ($\Delta\pi$, bar) into account.

$$J = L_p\,(\Delta P - \Delta\pi) \tag{1}$$

However, relationship between rejection and the applied pressure shows unreliable behavior. Rejections of metals were expected to decrease as applied pressures were increased from 1 to 5 bar. At higher pressure, water flux could be increased due to an increase of the preferential sorption of water and thus, the solvent permeability increases rather than solute permeability. Figure 2(b) presents the rejection of ferrous iron for ferrous chloride solution with concentration of 10 mg/L at pH6.8. Results show that iron removal by using TFC-SR3 membrane at this operating condition have reached more than 95% for applied pressure from 1 to 5 bar. In order to reach the allowable limits for Fe set by WHO for drinking water, therefore rejection should be more than 97%. Thus, the applied pressure at 2 and 3 bar are preferable for this operating condition due to the measured permeate concentrations were well below than the allowable value.



(a) (b)

Figure 2 Effect of applied pressure on (a) TFC-SR3 membrane for various feed concentrations and (b) rejection of Fe(II) ions by TFC-SR3 membrane for feed solution with 10 mg Fe/L at pH6.8

3.3 Influence of pH

Based on the idea that negatively charged membranes generally have higher rejection for charged solutes as reported in literatures, therefore the influence of feed pH on iron removal was a part of investigation. The iron rejection by TFC-SR3 membrane increased with increasing feed pH as presented in Figure 3. Results show that rejection of Fe(II) from neutral synthetic groundwater was satisfied with greater than 98%. The performance of this membrane due to Fe(II) rejection was increased to almost 100% as pH of feed solution was adjusted to 9.4 by adding NaOH. Whereas, the rejection was decreased to 95% as pH of feed solution was reduced to 4.4 by adding HCl. These results, briefly present that rejection for 10 mg Fe/L at applied pressure of 2 bar is prefereable in basic and neutral form. This is due to the fact that for this operating condition, permeate concentrations were below than WHO standard limit for drinking water. Whereas in acidic form, ferrous iron removal was below than 97% as required to reach acceptable value for consumption. TFC-SR3 membrane is amphoteric as reported by De Munari *et al.* [17] and it is positively charged at acidic pH while negatively charged at basic pH. Since this membrane is negatively charged at higher pH, it is expected that the rejection towards $FeCl_2$ is increased as increasing of feed pH. Thus, results proved that rejections were dependent on pH at this operating conditions.

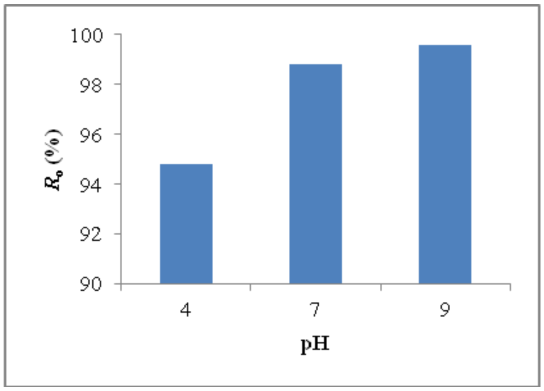


Figure 3 Effect of feed pH on the rejection of Fe(II) ions by TFC-SR3 membrane for feed solution with 10 mg/L at applied pressure of 2.0 bar

■4.0 CONCLUSION

In this study, results showed that removal of ferrous iron by selected commercial NF membrane was very encouraging. Rejections were significantly dependent on feed concentrations, applied pressures and feed pH. These studies indicate that NF membrane technology has successfully treated synthetic groundwater for drinking water resources and has high potential to treat real groundwater from selected monitoring wells in North Kelantan. Rejection of ferrous iron above 97% at pressure lower than 5 bar proved that TFC-SR3 membrane has the potential to reduce the value well below the maximum contaminant level that is set by WHO standards for drinking water. Further studies using other type of membranes are suggested to identify the potential of combination of treatment method for optimization of membranes performance.

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