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Flux Decline and Rejection Characteristics During Nanofiltration of Iron and Deionized Water

Oluranti Agboola^{1, *}, Jannie Maree², Richard Mbaya¹, Andrei Kolesnikov¹, Jakob Johannes Schoeman³, and Caliphs Musa Zvinowanda²

¹ Faculty of Engineering and the Built Environment, Department of Chemical and Metallurgical Engineering, Tshwane University of Technology, Pretoria 0001, South Africa

² Department of Environmental Water and Earth Sciences, Tshwane University of Technology, Pretoria 0001, South Africa ³ Department of Chemical Engineering, Water Utilisation Division, University of Pretoria, 0001, South Africa

This work was studied to determine the flux decline during nanofiltration of iron and deionised water. The rejection characteristic of iron was also studied. A stirred-cell was used for the experiment and Inductively Coupled plasma optical emission was used for iron analysis at various pH and pressure. The significant increased in flux declined at pH 3.01 and 3.44 is possibly caused by crystallized solids formed at the surface of the membrane and thus lead to the reduction of iron rejection at pH 3.01 and 3.44. At higher pressure more water passes through the membrane, thereby increasing the iron rejection. Experiment of clean water flux was done using the deionised water after the different pH experiment to see if the membrane is not fouling. The rejection characteristic of iron was also studied.

Keywords: Nanofiltration, Flux Decline, Rejection, pH, Pressure, Iron, Deionised Water.

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1. INTRODUCTION

A membrane is a permselective barrier that is capable to selectively separate components as a function of physical and chemical properties from a solution when a driving force is applied. Nowadays, membrane separation systems have become very important wastewater treatment technology, which facilitate the removal and recovery of pollutants, solvent i.e., water. Nanofiltration is a pressure-driven membrane process that has intermediate characteristics between ultrafiltration and reverse osmosis. Nanofiltration membranes find its applications in waste water and industrial water treatment (e.g., water softening, removal of colorants, heavy metals and organic matter).¹ The application of nanofiltration for environmental problems provides the best opportunity out of all other membrane systems. This is because of the high efficiency of separation achieved by nanofiltration membranes at relatively lower pressure of operation and their ability to separate organic as well as inorganic solutes. Consumption of energy in nanofiltration membrane is much lower than the reverse osmosis, which makes it an economically feasible unit operation.

Usually, nanofiltration membranes are negatively charged with a selective layer of thickness of $\sim 1 \ \mu m$

coated over the ultra-porous membrane layer support that controls all the transport properties by diffusion, convection and Donnan exclusion mechanisms. The charged nature of the membrane surface allows selective separation of certain ions over the other with very high efficiency of operation particularly for ionic species other than the monovalent ones. When dealing with ionic solutions, the existence of fixed charge in the membrane influences the ion distribution inside the membrane.² The operational parameters such as flowrate, pressure, pH, the membrane molecular weight cutoff (MWCO), pure water permeability and surface charge play an important role in the separations of metal compounds have been found to be dependent on the pH of the solution in specific conditions.^{8,9}

The aim of this work is to test the performance of Nano-Pro membrane through the reproducibility of deionised water and to determine the flux decline during nanofiltration of iron and deionised water.

2. MATERIALS AND METHODS

2.1. Experimental Device and Nanofiltration Membrane

The investigation was carried out using Memcom laboratory stirring cell. A polyamide flat sheet of NF membrane (Nano-Pro-3012) with effective surface area of 0.01075 m^2

^{*}Author to whom correspondence should be addressed.

was used with the cell containing one litre volume of sample. The operating pressure was controlled by high-pressure nitrogen gas connecting with gas pressure regulator. The operating pressure ranges between 10 to 30 bar and the speed was constant throughout the experiment (500 rpm). Inductively Coupled plasma optical emission was used for the analysis of iron.

2.2. Filtration Experiment

Nanofiltration experiments were tested as follows: the operating conditions used for the pH investigation on iron and reproducibility of deionised water are rapid stirrer of 500 rpm, operating pressure of 30 bar with filtration period of 180 minutes. The operating conditions used for pressure investigation of deionised and iron are 500 rpm, filtration period of 180 minutes and operating pressure of 10, 20 and 30 bar at pH 2.

The iron solution at different pH were prepared by adding sodium hydroxide to ferrous sulphate solution to increase the pH to 2.48, 3.01, 3.44 at a target composition of 600 mg/L for iron. 0.62 g, 0.74 g and 0.77 g NaOH was added to the solution to raise the pH of the solution to 2.48, 3.01 and 3.44 respectively. Solution pH was monitored with pH meter. Experiments were conducted at 30, 60, 90, 120, 150 and 180 minutes, while the flux was measured by weighing permeate over time and from this the water recovery can be calculated. Clean water flux experiments were also performed after each pH and pressure to see if the membrane is not fouling.

Ferrous sulphate and sulphuric acid that makes up 1 liter of solution used for the experiment are given below.

$$2\text{FeS}_2 + 7\text{O}_2 + 2\text{H}_2\text{O} \longrightarrow 2\text{FeSO}_4 + 2\text{H}_2\text{SO}_4$$

Composition $\longrightarrow 600 \text{ mg/l} 900 \text{ mg/l}$
Mole ratio $\longrightarrow 1:1$

2.3. Membrane Characteristic

The membrane productivity is expressed as permeate through the membrane. The permeate flux is defined as:

$$J_w = (\ln^{-2} h^{-1}) = \frac{Q}{(A \times t)}$$
 (1)

Where Q is the volume of permeate (l), A is the area of the membrane (m^2) and t is the permeate time (h). And water permeability, L_p (lm⁻² h⁻¹ bar⁻¹) is defined as:

$$L_p = \frac{J_w}{\Delta P} \tag{2}$$

Where, ΔP is the applied pressure (bar).

The solute rejection (R) is defined as:

$$R\% = (1 - (C_p/C_f)) * 100$$
(3)

 C_p and C_f are metal concentration of permeate solution respectively.

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3. DISCUSSION AND RESULT

Figures 1 and 2 show the reproducibility of deionised water. This experiment was done in order to investigate the performance of the membrane materials and to see if the material can be able to reproduce similar results. Excellent reproducibility was found for three different run of deionised water solution. The result of permeate flux as a function of water recovery and time respectively in Figures 1 and 2 show that the membrane materials is reproducible.

Figures 3 and 4 show the effect of pressure on flux and water recovery. The pressure has significant effect on nanofiltration membrane performance. As expected the flux was higher at higher pressure as shown in Figure 3. It could be seen from Figure 3 that the investigation of different pressure for the effect of flux was almost constant because there is no much salinity in deionised water, so the solution will not be concrentrated. Figure 4 shows that the higher the pressure the higher the water recovery.

Many studies have shown that pH level of solutions plays a major role on the performance of the Nanofiltration

Nano-Pro A-3012 Water Recovery with deionised water at 25°C 50.000 45.000 Permeate Flux, L/m²/hr 40.000 35.000 25°C 30.000 Run1-D-H2O Run2-D-H2O 25.000 ARun3-D-H2O 20.000 0 20 40 60 80 100 120 Water Recovery, %

Fig. 1. Permeate flux as a function of water recovery.

Nano-Pro A-3012 Permeate flux with deionized water at 25°C



Fig. 2. Permeate flux as a function of time.

♦ pH = 2

■ pH = 2.48

▲ pH = 3.01

pH = 3.44

100

80

Effect of pH on Flux



Fig. 3. Flux of deionised water for Nano-proA-3012 versus time at different membrane pressure (25 °C).



45

40

35

30

25

20

15

10

5

0 [

××

40

% Water Recovery

60

20

Permeate flux, L/m²/hr

Fig. 4. Flux of deionised water for Nano-proA-3012 versus water recovery different membrane pressure (25 °C).

membrane. The pH of the feed was controlled by addition of NaOH. It can be seen in Figures 5 and 6 that increase in the osmotic pressure as a result of the NaOH added to the solution caused flux to decline at 3.01 and 3.44, therefore the rate and extent of flux decline increased with



Fig. 5. Flux as a function of time at different pH values.

56

Effect of pH on Fe Rejection 120 100 Ъ. **Rejection of** 80 60 • pH2 pH2.48 40 ▲ pH3.01 % 20 $\times pH3.44$ 0 0 20 40 60 100 80 %water Recovery

Fig. 8. Fe rejection as a function of water recovery at different pH values.

Fig. 7. Fe rejection as a function of time at different pH values.

increasing solution pH. The decreased in flux declined at pH 3.01 and 3.44 caused lower rejection. Figures 7 and 8 confirm the expectation that rejection characteristic of Nanofiltration membrane can be controlled by pH. Iron rejection at 3.01 and 3.44 were found to decrease, this was possibly caused by crystallized solids formed at the surface of the membrane as a result of increase in the osmotic pressure and decreased pore volume (size) and thus lead to the reduction in flux and iron rejection. The pH at 3.01 and 3.44 are responsible for changes in the feed solution, causing changes in membrane performance.



Fig. 9. Flux of deionized water as function of time after the investigation of pH at 30 bar.



Fig. 10. Flux of deionized water as function of water recovery after the investigation of pH at 30 bar.



Fig. 11. Flux as function of time at different pressure.

The results of flux as a function of time and water recovery are shown in Figures 9 and 10. This is to investigate if the membrane did not foul after different pH experiment. Clean water flux experiments were performed to see if the membrane did not foul after each experiment. The flux declined was significant after pH 3.44; this is as a result of solute deposited onto the surface of the membrane. The



Fig. 12. Flux as a function of water recovery at different pressure.



Fig. 13. Fe rejection as a function of time at different pressure.



Fig. 14. Fe rejection as a function of water recovery at different pressure.



Fig. 15. Flux of deionized water as function of time after the investigation of pressure.



Flux investigation on deionized water at pH 2

Fig. 16. Flux of deionized water as function of water recovery after the investigation of pressure.

membrane was cleaned and the experiment was repeated to see if the membrane still maintains its performance.

The results of flux and iron rejection as a function of time and water recovery are shown in Figures 11–14 respectively. Pressure difference is the driving force responsible for a Nanofiltration process. At higher pressure, more water passes through the membrane and thus leads to higher flux as shown in Figures 11 and 12. Higher pressure leads to higher flux with increase in water recovery and thus increase the rejection rate: The higher the pressure the higher the rejection of iron as shown in Figures 13 and 14. At pressure of 10 bar, the water recovery was about 40% as shown in Figure 14, that is the lower the pressure the lower the rejection rate of iron and water recovery for the period of filtration.

The results of flux as a function of time and water recovery are shown in Figures 15 and 16. This is to investigate if the membrane did not foul after different pressure experiment. Figures 15 and 16 show that the membrane did not foul much after the experiments of pressure investigation.

4. CONCLUSION

Membrane technologies are accepted widely in portable water production, wastewater treatment and pharmaceutical industries to help stimulate further research and development work on improving the performance of the membrane system in general. Nanofiltration find its applications mostly in industrial water and wasterwater treatment process. It could be seen from the repeatability of deionized water that permeate flux could be maintained under the same condition. Nanofiltration treatment of water containing iron evidenced that transmembrane pressure and pH have great contribution on the flux and the rejection rate. With iron solution, high pH is not too good because it will precipitate thus the reason pH 2-3.44 were used. Flux and rejection decreased significantly at higher pH is due to inceased metal crystallization form on the surface of the membrane. The interaction between the ferrous sulphate solution and sodium hydroxide caused flux reduction due to increased permeate resistance at the membrane surface. The solution of deionised water was not concrentrated in the investigation of prssure on flux thus Technolmake the flux almost constant because there is no much salinity in deionised water. Pure water experiment shows that the membrane did not foul much after the experiments of pressure and pH investigations.

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