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Effect of Solution Chemistry on the Nanofiltration of Nickel from Aqueous Solution

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Abstract

The rejection of nickel ions on water solutions was studied using aromatic polyamide nanofiltration membrane NF 90 by determination of solution chemistry as the concentration of solution, the pH and ionic strength at 27°C. The experimental results showed that the lower flow solution depends on concentration, solution pH and ionic strength. The solution concentrations showed greater decrease in flux and rejection. Flux decline conducted with a solution of nickel dropped for pH of the solution. At high pH, flux solutions showed higher flux decline than those of low solution pH, while the rejection of ions presented higher rejection. Increased ionic strength had a greater increase in flux decline. The rejection of nickel ions was found to be decreased with decreasing solution pH and increasing ionic strength. Flux and rejection decreased further to the higher ionic strength, which reduces the negative charge repulsion on the surface of the membrane, and thus a decrease of rejection. In addition, comparisons on the decline of flows to co-ions have also been studied in experiments filtration.

Keywords: Flux decline; Membrane; Nickel; Nanofiltration; Rejection

Introduction

The occurrences of metal ions in the aquatic environment have been concern because of their toxicity, while the accumulation may pose various hazards for human health and environment. At the present, the most frequency practiced treatment technology for the removal of metal ions from aqueous solution is chemical precipitation, which only relocated metal ions from aqueous phase, and leave further sludge problems to be solved. Therefore, the removal of metal ions from many industrial wastewaters has stimulated vigorous research activities in the development of appropriate treatment technologies. Membrane separation technologies have been determined to be a feasible option for removal of heavy metal from aqueous solution because of its relative ease of construction and control, and the feasible recovery of valuable metals. Nanofiltration processes is capable of removing heavy metal [1]. They are efficient technologies to remove feed source water in terms of natural organic matter (NOM) [4], inorganic scalants [5-7], salt solution [8-10] and heavy metals [11-15]. Nanofiltration (NF), one of membrane technologies, is a relatively new membrane process, which is considered to be intermediate between ultrafiltration (UF) and reverse osmosis (RO) in terms of operating conditions. NF membrane processes operate at pressures between 50 and 150 psi much lower than RO (200 to 1000 psi), but higher than UF (10 to 70 psi). At the present time, NF is increasingly applied in the field of water treatment. However, membrane fouling caused by organic and inorganic substances can be a major factor for limiting more widespread use of membrane technologies, reducing long-term filtration, and increasing costs for membrane operation through higher labor, cleaning and replacement. Inorganic fouling (i.e. negative and positive ions) can be a significant factor that enhances permeate flux decline during filtration. This may cause an increased concentration polarization that exceeds solubility limit, resulting precipitation (i.e. Ca^{2+}, Mg^{2+}, CO_3^{2-}, SO_4^{2-}, and PO_4^{3-}). This has been recently investigated by Jarusutthirak et al. [7]. Molinari et al. [12] investigated the interactions between membranes (RO and NF) and inorganic pollutants (i.e. SiO_2, NO_3^-, Mn^{+2}, and humic acid). They showed that membrane fouling was caused by the interactions between the membranes and other ions. Other factors, which can cause membrane fouling, are solution pH, ionic strength, concentration, solution composition and operating conditions.

The objective of this work was to investigate the effects of solution chemistry during nanofiltration of nickel solution. The effects of solution chemistry (i.e. concentration, solution pH and ionic strength) were determined on nanofiltration fouling. The discussion of this study was further adapted to improve membrane filtration for long-term operation.

Experimental

Nanofiltration characteristics

An aromatic polyamide thin-film composite NF-90 membrane, produced by Dow-FilmTec., was chosen to determine the effect of solution chemistry on nanofiltration performance. According to the manufacturer, the maximum operating pressure is 600 psi (or 4,137.6 kPa), maximum feed flow rate is 16 gpm (3.6 m³/hr), and maximum operating temperature 113°F (45°C) and the operating pH is ranged from 1 to 12.

Analytical method

Nickel ion concentration was measured by using atomic absorption (AA) spectrometry (AAnalyst 200 Version 2, Perkin Elmer Corp.). Measurements of solution pH, conductivity and temperature were made using pH meter (shott), and conductivity meter (shott) respectively. Ionic strengths were calculated using a correlation between conductivity and ionic strength of NaCl standard, I.S.[M]=$0.5\Sigma C_i Z_i^2$ (C_i is the ion concentration and Z_i is the number of ions).

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Flux decline experiments

The experiments were carried out with three liters of solution containing nickel solution ($NiCl_2$ and $Ni(NO_3)_2$) in varied concentration of 10, 20, 50 and 100 mg/L, while varying solution pH from 4 to 6 and ionic strengths (0.01, 0.05 M as NaCl). Flux decline experiments were tested by using a 400-ml dead-end membrane filtration apparatus (Amicon 8400 USA).

A membrane sheet can be fitted to the cell. The membrane active area is about 41.38 cm^2. The operating pressure was employed via high-pressure regulator of nitrogen cylinder. The permeate flux was kept in a beaker on the electrical balances.

Filtration experiments

Membrane sheets were rinsed with cleaned dionised water, 0.125 M citric acid solution of pH 4, and followed with 10^{-4} M sodium hydroxide solution of pH 10 for 30-min and dried.

Cleaned water flux was determined with a function of transmembrane pressure. Dionised water was subsequently tested for 30-min membrane compaction with velocity rate of 300 rpm. The water flux J_0 was subsequently determined with increased operating
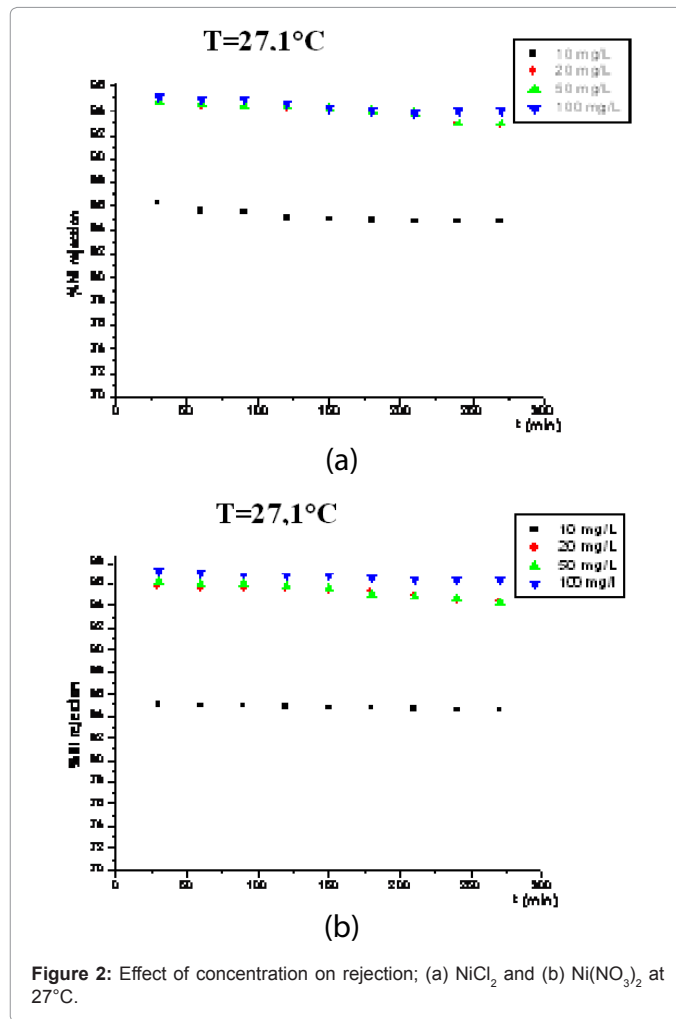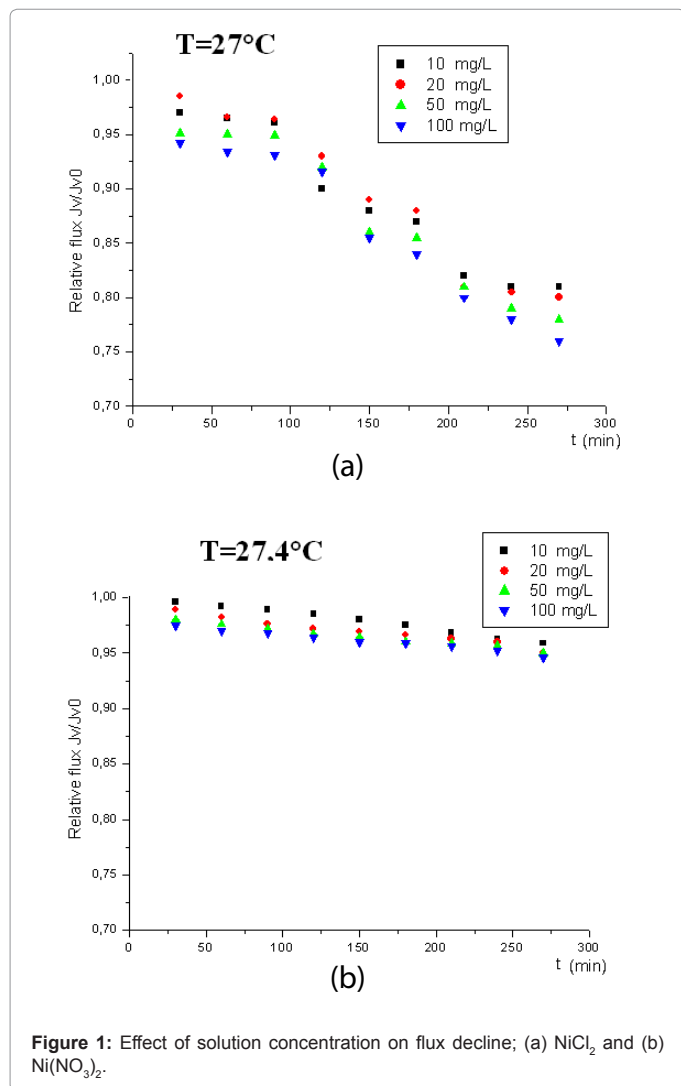

Figure 1: Effect of solution concentration on flux decline; (a) $NiCl_2$ and (b) $Ni(NO_3)_2$.

Figure 2: Effect of concentration on rejection; (a) $NiCl_2$ and (b) $Ni(NO_3)_2$ at 27°C.

pressures before nickel solution was used with the system. Feed nickel solutions were prepared for each tested condition. After filtration was terminated, two steps of cleaning, i.e. hydrodynamic followed by chemical cleaning, were performed. For this, the membrane sheet was cleaned with dionised water, then followed with chemical cleaning, acidic solution (using citric acid) with pH of 4 for 30-min each. After each cleaning, water fluxes at different operating pressures were measured to determine water flux recovery.

Analysis of results

The parameters taken into account were: -The volumetric flux J_v (L/m^2/h or LMH) was determined by measuring the volume of permeate collected in a given time interval divided with membrane area by the relation:

$$J_v = \frac{Q}{A} \qquad (1)$$

Where, Q and A represents flow rate of permeate and the membrane area, respectively. The observed rejection was calculated by the following relation:

$$\% R = (1 - \frac{C_p}{C_i}) \times 100 \qquad (2)$$

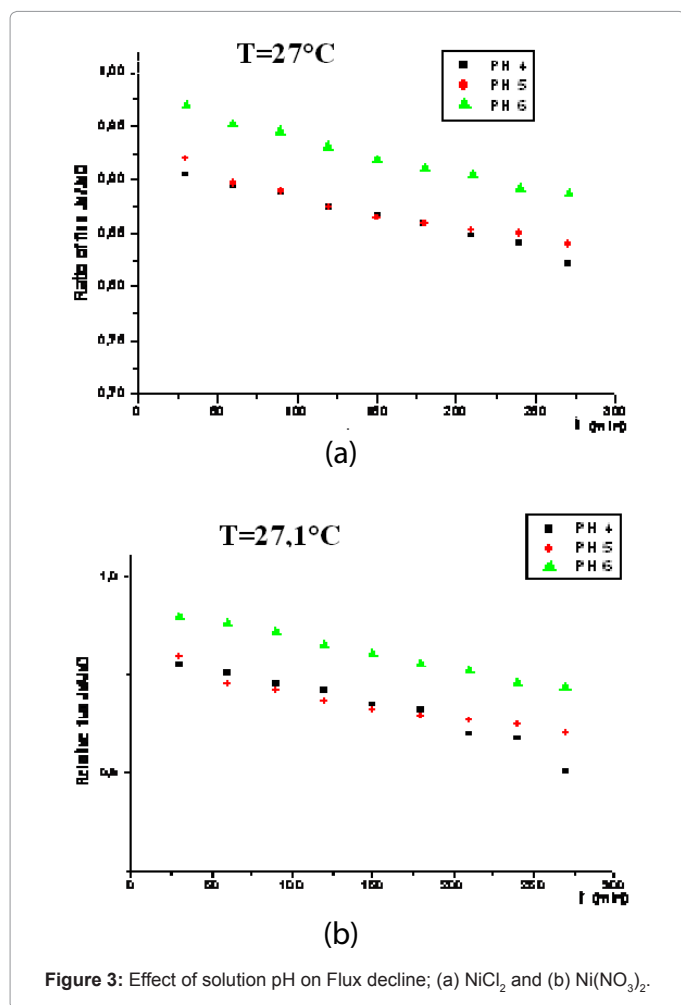Where C_p and C_i are the solution concentrations in the permeate and in the initial feed solution, respectively.

Figure 3: Effect of solution pH on Flux decline; (a) $NiCl_2$ and (b) $Ni(NO_3)_2$.

Results and Discussions

Effect of solution concentration on flux decline and rejection

In order to determine the effect of metal concentration on flux decline and rejection, Nickel ion concentrations were varied for 10, 20, 50 and 100 mg/L at pH 6 and ionic strength of 0.01 M NaCl. As shown in Figure1 and Figure 2, it can be concluded that nickel ion rejection and solution flux decline increased with increasing solution concentrations. The higher solution concentrations for $NiCl_2$ solution had greater ion rejection, about 94%-98 %, while low solution concentration exhibited lower rejection about 85%-97%. For $Ni(NO_3)_2$ solutions having 100 mg/L concentration had the ion rejection about 98-99%, while those of the lower concentrations were 88-95%.

Effects of solution pH on flux

The effects of solution pH on flux decline of $NiCl_2$ and $Ni(NO_3)_2$ solution were carried out at pH 4,5 and 6 with keeping constant ionic strength 0.01 M as NaCl at 60-psi operating pressure. Nickel concentration was about 20 mg/L. Figure 3 showed relative flux with function of operating period for $NiCl_2$ and $Ni(NO_3)_2$ solution. It can be seen that the rate and extent of flux decline increased with increasing solution pH. For the solution of $NiCl_2$ and $Ni(NO_3)_2$ solution at lower pH, flux solutions showed higher flux decline than those of low solution pH. At low pH, it suggested an increased fixed charge of H^+, decreasing electrical double layer thickness within membrane or both, thus decreased the concentration at the membrane surface. At high pH, the

membrane surface and pores become both more negatively charged due to the presence of anion (inorganic). In addition, the osmotic pressure near the membrane surface increase with high salt rejections, thus decreasing the driving pressure. These mechanisms nickel to a decrease in permeate flux and an increase in salt rejection with pH.

Effects of solution pH on rejection

The effects of the solution pH on rejection of $NiCl_2$ and $Ni(NO_3)_2$ solution was carried out with different solution pH from 4 to 6. Solution was maintained constant with Ionic strength of 0.01 M NaCl, 60 psi operating pressure and solution concentration of 20 mg/L during filtration. The obtained results were presented in Figure 4. Nickel ion rejection was found to be decreased with decreasing solution pH level. It was possibly due to higher solution pH, membrane surface take more negative charges, thus attracting greater lead ion. Consequently, solution pH of 5-6 for $NiCl_2$ had greater ion rejection about 96%-98 %, while low solution pH exhibited lower rejection about 88%-91%. For $Ni(NO_3)_2$ solution, the ion rejection percentages of higher solution pH (5-6) and lower solution pH were 91-94% and 76-81%, respectively.

Effects of ionic strength on flux

Figure 5 presents the effect of ionic strength on flux that was carried out at pH 6 with different ionic strengths of 0.01 and 0.05 M as NaCl. It was observed that the extent and rate of solution flux decline increased with increasing ionic strength. In the study, increases in ion
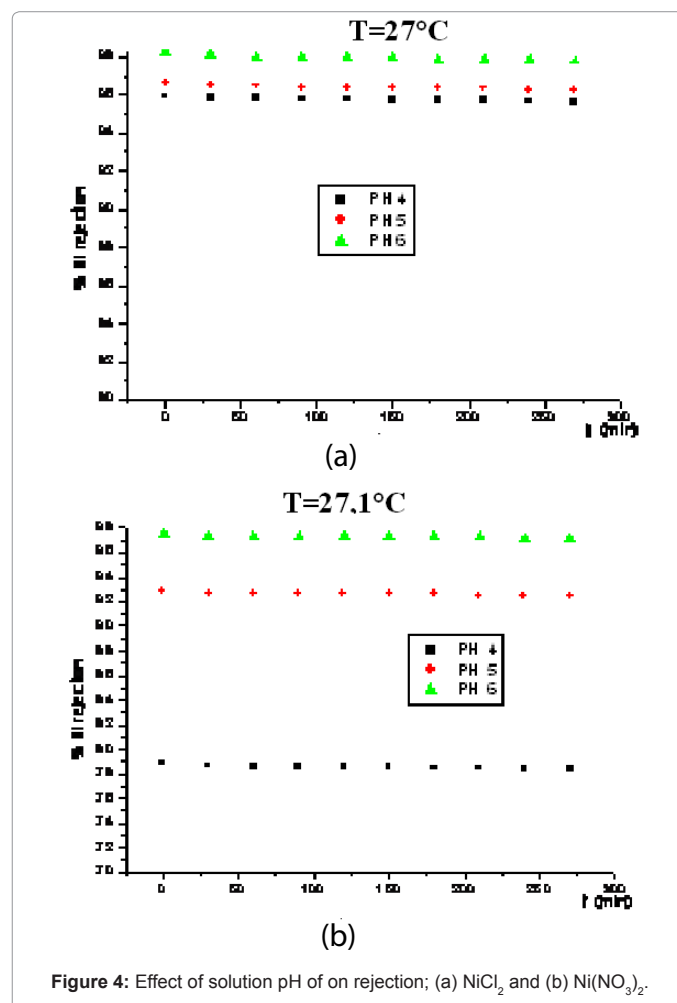

Figure 4: Effect of solution pH of on rejection; (a) $NiCl_2$ and (b) $Ni(NO_3)_2$.

concentration can reduce the permeability of charged membranes [20,21], thus reduced permeate flux. The results showed the similar trend for both of 20 mg/L $NiCl_2$ and $Ni(NO_3)_2$ solution.

Effect of ionic strength on nickel ion rejection

The effect of salt solution on nickel ion rejection were carried out with ionic strengths of 0.01 and 0.05 M as NaCl and the concentration of 20 mg/L, as shown in Figure 6. Solution pH of 6 for $NiCl_2$ and $Ni(NO_3)_2$ solution was kept constant during filtration. It was found that Nickel rejection at ionic strength of 0.05 M showed lower than those at ionic strength of 0.01 M. This was possibly due to increasing salt concentration, reducing membrane permeability, thus allowing nickel ion passage through the membrane surface.

Effect of co-ion on solution flux decline

The effect of the co-ion on Nickel solution flux decline was carried out with two types of Ni^{2+} ($NiCl_2$ and $Ni(NO_3)_2$) solution at the concentration of 20 mg/L. The experiments were performed at pH 6, ionic strength of 0.01 M NaCl and 60 psi operating pressure during filtration. From the experiment, it can be seen that $Ni(NO_3)_2$ solution had a slight lower on flux decline than $NiCl_2$ solution. $NiCl_2$ solution showed higher rejection than $Ni(NO_3)_2$ solution. Since the NF membrane is more negatively charged the monovalent anion of Cl^- is more excluded than NO_3^- resulting in greater rejection. Furthermore, the ion rejection is mainly dependent on its hydration energy in the
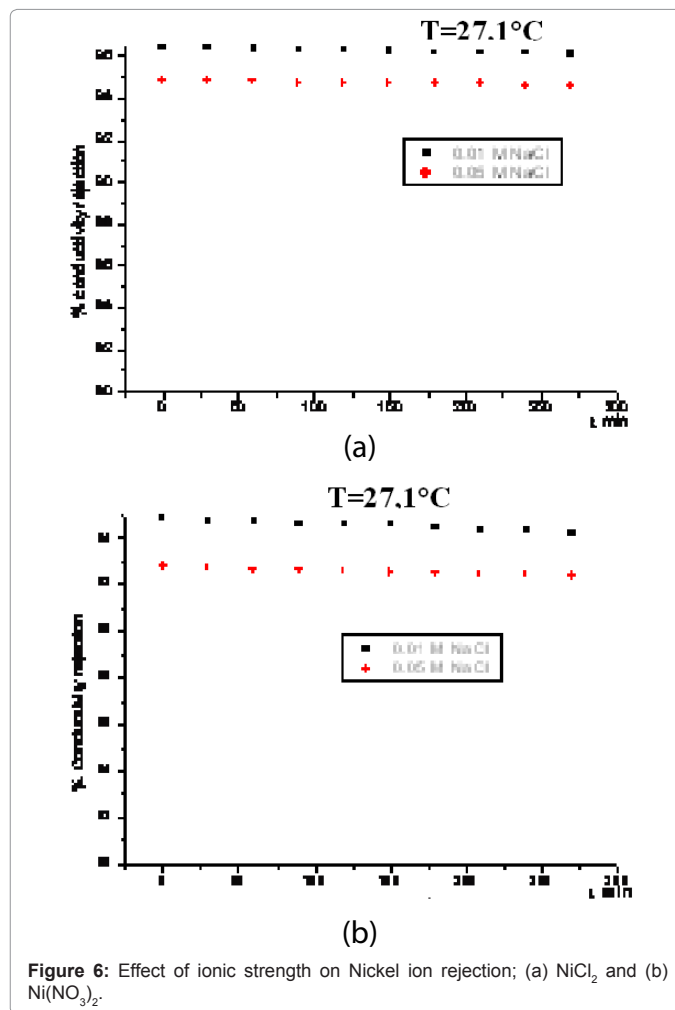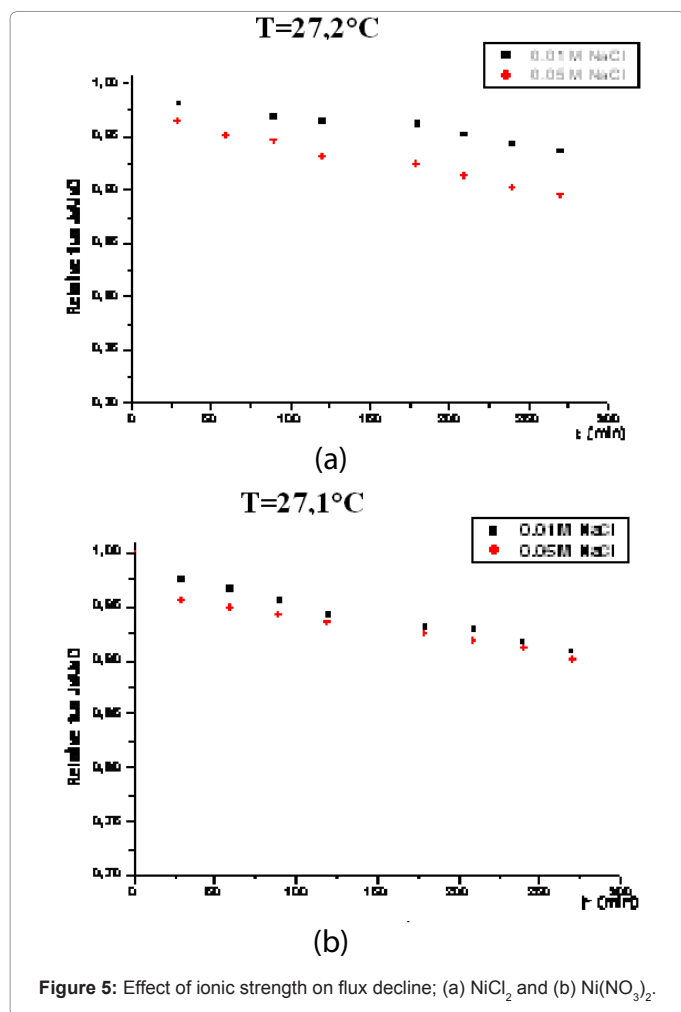

(a)

(b)

Figure 6: Effect of ionic strength on Nickel ion rejection; (a) $NiCl_2$ and (b) $Ni(NO_3)_2$.

solution and it was more retained if it has higher hydration energy, in accordance with the mechanism of solution-diffusion .The hydration energy of Cl^- and NO_3^- are 372 KJ/mol and 328 KJ/mol respectively [21]. However, the higher rejection of chlorides can not be described by the hydration energy owning to the fact that these anions had close hydration energy. It can be explained by the formation of the complexes solution, which involves an electrostatic repulsion with charge membrane and thereafter a strong rejection of the ions.

Conclusion

Nickel ions rejection and flux decline from aqueous solution by nanofiltration was strongly influenced by concentrations, solution pH and ionic strength. Flux decline conducted both in $NiCl_2$ and $Ni(NO_3)_2$ solution decreased for solution pH and concentrations. At higher solution pH, flux solutions showed higher flux decline than those of low solution pH, while le rejection exhibited higher rejection. Increased ionic strength had a greater increase in flux decline. Nickel ion rejection was found to be decreased with decreasing concentrations, solution pH and increasing ionic strength.

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(a)

(b)

Figure 5: Effect of ionic strength on flux decline; (a) $NiCl_2$ and (b) $Ni(NO_3)_2$.

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