Desalination and Water Treatment
www.deswater.com

doi: 10.5004/dwt.2018.23062

129 (2018) 94–100
October

Separation of magnesium and lithium from brine with high Mg^{2+}/Li^{+} ratio by a two-stage nanofiltration process

Qiuyan Bi[a,b], Shiai Xu[a,b,]*

[a]*Shanghai Key Laboratory of Advanced Polymeric Materials, Key Laboratory for Ultrafine Materials of Ministry of Education, School of Materials Science and Engineering, East China University of Science and Technology, Shanghai 200237, China, Tel. +86 21 64253353; emails: saxu@ecust.edu.cn (S. Xu), bqy1110@163.com (Q. Bi)*
[b]*School of Chemical Engineering, Qinghai University, Xining 810016, China*

Received 8 February 2018; Accepted 3 September 2018

ABSTRACT

A two-stage nanofiltration (NF) process was developed to separate magnesium (Mg^{2+}) and lithium (Li^{+}) from brine with a high Mg^{2+}/Li^{+} ratio, and the effects of transmembrane pressure, pH and Mg^{2+}/Li^{+} ratio on the volume permeation flux, ion rejection, separation factor (SF) and Mg^{2+}/Li^{+} ratio of permeate were investigated. As the transmembrane pressure increases, the rejection rates of Mg^{2+} ($R(Mg^{2+})$) and Li^{+} ($R(Li^{+})$) vary slightly for the first NF process; whereas $R(Mg^{2+})$ remains almost constant, but $R(Li^{+})$ increases sharply at first and then levels off for the second NF process. pH is a crucial factor for the two-stage NF process, and Mg^{2+} and Li^{+} can be better separated at lower pH value. The SF of Mg^{2+} and Li^{+} increases with the increase of the Mg^{2+}/Li^{+} ratio of the feed solution. The Mg^{2+}/Li^{+} ratios of permeate of West Taijnar salt lake brines can be reduced from the initial 13.25 to 0.17 at pH 3.5. The long-term filtration test confirms that the two-stage NF process is stable. Thus, the two-stage NF process proposed in this study can be a promising approach to separate Mg^{2+} and Li^{+} from brine with a high Mg^{2+}/Li^{+} ratio.

Keywords: Nanofiltration; Two-stage process; Lithium; Mg^{2+}/Li^{+} ratio; Rejection rate

1. Introduction

Lithium (Li) and Li salts have been widely used in various applications including batteries, aerospace, nuclear power, alloys, drugs and lubricants [1]. The global demand for Li ion battery in the field of electric vehicles is expected to reach $221 billion by the year of 2024 [2]. Various methods have been developed to separate Li^{+} from brine with a high Mg^{2+}/Li^{+} ratio, such as precipitation [3], extraction [4], ion exchange and sorption [5], and electrodialysis [6]. However, each method suffers from some limitations that may make it impractical or undesirable in some circumstances. For instance, a large quantity of precipitating agent is required for the precipitation method, and it is difficult to re-extract solvent for the extraction method. A major challenge for the ion exchange method is the development of novel ion exchange resins with highly selective functional groups. More importantly, these methods are too costly for brine with a high Mg^{2+}/Li^{+} ratio, and thus they are more applicable for the brine with a Mg^{2+}/Li^{+} ratio less than 1.

Nanofiltration (NF) is a pressure-driven membrane process with characteristics between ultrafiltration and reverse osmosis (RO), and it is suitable for a wide range of applications such as water softening [7,8], wastewater reuse, concentration of juices [9], and desalination [10]. NF has been shown to be better than some separation methods in extracting monovalent ions from a system containing multivalent ions [11]. Attempts have been made to separate Mg^{2+} and Li^{+} from brine with a high Mg^{2+}/Li^{+} ratio via a single NF process [6,10,11]. Mg^{2+} can be extracted using the reject solution via the membrane crystallization, membrane distillation or other processes; while Li^{+} can be extracted using the permeate solution via the extraction or other processes. However, separation

---

* Corresponding author.

performance of a single NF process is not as good as expected [10–12]. Thus, it is more desirable to use a two- or multi-stage NF process. In order to address this problem, a two-stage NF process was developed in this study to separate Mg^{2+} and Li^+ from brine, and the effects of transmembrane pressure, pH and Mg^{2+}/Li^+ ratio on the volume permeation flux, ion rejection, separation factor (SF) and Mg^{2+}/Li^+ ratio of permeate were investigated. A long-term filtration test was performed to determine the stability of the proposed two-stage process.

2. Experimental setup

2.1. Materials

The two membranes used in this study, 3B01S-DAC8-1812 (NF1, molecular weight cut-off [MWCO]: 250–300 Da) and 3B02S-DAC8-1812 (NF2, MWCO: 150–200 Da), were supplied by Suntar Membrane Technology Co., Ltd. (Xiamen, China), both of which had an active area of 0.24 m^2 and could stably operate in the pH range of 2.0–11.5. Hydrochloric acid (HCl), LiCl and $MgCl_2·6H_2O$ were provided by Sinopharm Chemical Reagent Co., Ltd. (Beijing, China) and used for preparation of simulated brine with pure water obtained by means of a RO membrane system (UPT-1-5T, China). The mass ratios of Mg^{2+} to Li^+ ranged from 3.43 to 24.01, and the salt concentration (C_f) was fixed at 6.0 $g·L^{-1}$. The raw brine obtained from West Taijnar salt lake was also examined in this study, and the ion concentrations were shown in Table 1. The raw brine was diluted by pure water before feeding to ensure a total $MgCl_2$ and LiCl concentration of 6.0 $g·L^{-1}$. The pH of the diluted brine (feeding brine) was adjusted with HCl.

2.2. Experimental procedure

All experiments were carried out in a laboratory setup on the basis of a spiral wound DK-1812 model (Suntar Membrane Technology Co., Ltd, Xiamen). The filtration system is schematically shown in Fig. 1. Each cell had an active membrane area of 0.24 m^2, and new membranes were thoroughly cleaned with deionized water. The feed temperature was fixed at 20°C ± 0.2°C using a circulating water cooling system. In the filtration tests, a small amount of samples were taken out from the feed and permeate tanks every 30 min for elemental analysis when the system reached a stable state under each operation condition. The flux was measured by collecting permeate, and both permeate and retentate were recycled to feed tanks after flux measurement. In the separation tests, permeate was collected into the permeate tank while the retentate was recycled into the feed tank. The permeate of the first NF stage (in which NF1 was used) was used as the feed for the second NF stage (in which NF2 was used). The separation efficiency of both NF1 and NF2 was characterized by volume flux, rejection rate and SF. All experiments were repeated at least twice depending on the errors, and the averages and deviations were calculated.

Table 1
pH and ion concentrations of the raw brine from West Taijnar salt lake ($g·L^{-1}$)

pH	6.2
Na^+	2.041
K^+	1.043
Li^+	8.666
Ba^{2+}	0.008
Ca^{2+}	0.132
Mg^{2+}	114.800
Mg^{2+}/Li^+	13.25

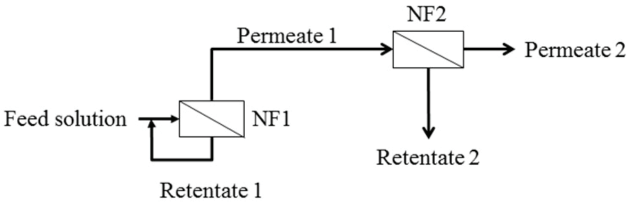

Fig. 1. Schematic of the two-stage filtration system.

The volume permeate flux (J_v, $m^3·m^{-2}$ s^{-1}) was used to describe the rate at which the solution permeated the membrane, which was defined as the permeate solution volume per area per unit of time.

$$J_v = \frac{V}{A \times \Delta t} \quad (1)$$

where V, A, and Δt are the volume of permeate solution (m^3), the active surface area of the membrane (m^2), and the permeation time (s), respectively.

The rejection rate (R, %) was calculated by Eq. (2) as follows:

$$R(i) = (1 - \frac{C_{pi}}{C_{fi}}) \times 100\% \quad (2)$$

where C_{pi} and C_{fi} are the concentration of i in permeate ($g·L^{-1}$) and feed ($g·L^{-1}$), respectively, in which i refers to either Mg^{2+} or Li^+.

The SF (no unit) was used to describe the separation efficiency.

$$\text{SF} = \frac{(C_{\text{Li}^+} / C_{\text{Mg}^{2+}})_{\text{permeate}}}{(C_{\text{Li}^+} / C_{\text{Mg}^{2+}})_{\text{feed}}} \quad (3)$$

where $C_{Mg^{2+}}$ and C_{Li^+} are the Mg^{2+} and Li^+ concentrations in permeate ($g·L^{-1}$) and feed ($g·L^{-1}$), respectively.

2.3. Separation stability

The separation stability of NF1 and NF2 membranes was evaluated by a long-term filtration test using the raw brine with a pH of 6.2 and 3.5, respectively. The membrane was pressurized with DI water at 0.8 MPa and 20°C ± 0.2°C for 0.5 h to achieve a stable stage, and the volume permeate flux (J_{v0}) for pure water was measured. Then, the brine was continuously fed for 3 h, and the steady flux and concentration of Mg^{2+} and Li^+ were measured every 15 min, which were denoted as J_{v1} and C_{pi}, respectively. After that, the fouled membrane was thoroughly washed with DI water at 0.4 MPa and 20°C ± 0.2°C for 20 min, and the volume permeate flux (J_{v2}) for pure water was measured again at 0.8 MPa and 20°C ± 0.2°C. The experiment was stopped after four cycles.

The relative flux (RF, no unit) was calculated by Eq. (4).

$$RF = \frac{J_{v1}}{J_{v0}} \tag{4}$$

2.4. Measurements of ion concentration and pH

Mg^{2+} and Li^+ concentrations were determined using inductively coupled plasma (ICAP6300 spectrometer, USA) as described in our previous study [12]. The pH of the solution was measured using a FE20K pH meter (Mettler Toledo, Shanghai).

3. Results and discussion

3.1. Separation of Mg^{2+} and Li^+ from simulated brine

3.1.1. Effect of transmembrane pressure

Fig. 2 shows that the volume permeate flux of the simulated brines with different Mg^{2+}/Li^+ ratios increases linearly with the increase of transmembrane pressure, indicating that both NF1 and NF2 membranes perform well regardless of the Mg^{2+}/Li^+ ratio. Fig. 3 clearly shows that for NF1 membrane, the rejection rates of Mg^{2+} (R(Mg^{2+})) and Li^+ (R(Li^+)) vary slightly with increasing transmembrane pressure; whereas for NF2 membrane, R(Mg^{2+}) remains largely unchanged, but R(Li^+) increases sharply at first and then slowly with increasing transmembrane pressure. The steric hindrance indicates that Mg^{2+} has a higher retention than Li^+ due to its larger Stokes radius (the sieving effect of NF), and Li^+ has a higher diffusion coefficient than Mg^{2+} [13,14]. As a result, R(Mg^{2+}) is higher than R(Li^+) for both NF1 and NF2 membranes, and thus Mg^{2+} could be preferentially separated from Li^+, which can be explained by Eq. (5) as follows:

$$J_v = B \times \Delta C_S \tag{5}$$

where B and ΔC_S are the permeation coefficient and concentration difference of the salt on the sides of the membrane, respectively.

Clearly, as the transmembrane pressure increases, both the volume permeate flux and ΔC_S increase, resulting in an increase of salt rejection of both Mg^{2+} and Li^+ [15–17]. And the rejection of Li^+ increases quickly at first and then levels off for NF2 membrane, which might be caused by the dense surface or the small pore size of NF2 membrane. Compared with NF1, NF2 has lower volume permeate flux but higher salt rejection due to the small size of NF2. The molecular cut-off of NF1 is 250–300 Da, whereas that of NF2 is 150–200 Da, indicating

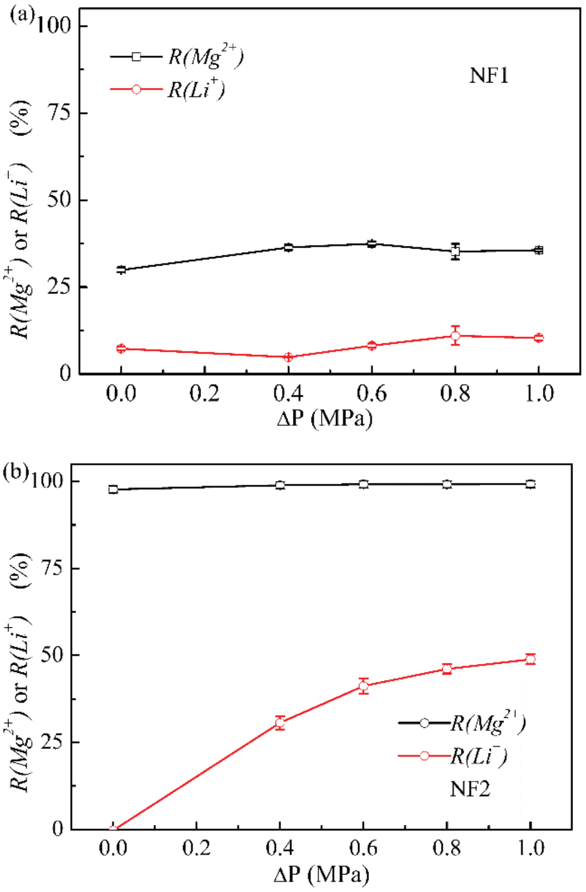

Fig. 3. Effect of transmembrane pressure on rejection rate of Mg^{2+} and Li^+ (Mg^{2+}/Li^+ = 3.43, pH = 6.1), (a) NF1, (b) NF2.

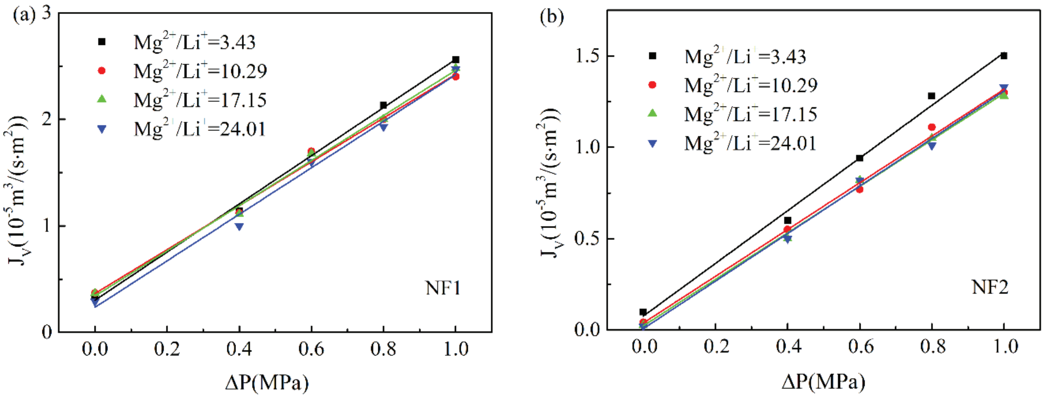

Fig. 2. Effect of transmembrane pressure on the volume permeate flux (pH = 6.1), (a) NF1, (b) NF2.

that NF2 has smaller a pore size than NF1. Taking into account the volume permeate flux and rejection rate, a transmembrane pressure of 0.8 MPa is determined to be suitable.

3.1.2. Effect of feed solution pH

The surface charge of NF membrane is zero at a certain pH (isoelectric point, IEP), below which it is positively charged and above which it is negatively charged [18,19]. Thus, the pH of the solution can have a significant effect on the separation process. The IEPs of NF membranes used in this study range from 4.00 to 4.55 [20]. As shown in Fig. 4, R(Mg^{2+}) decreases for NF1, but changes only slightly for NF2 with increasing pH, whereas R(Li^+) increases with increasing pH for both NF1 and NF2. Similar result was also reported by Sun et al. [10] and Ji et al. [21]. Additionally, the SF of Mg^{2+} and Li^+ decreases sharply with the increase of pH, suggesting that the separation efficiency of the two membranes decreases with the increase of pH. This can be attributed to the following reasons. First, the surfaces and pores are positively charged at a pH lower than IEP but negatively charged at a pH higher than IEP, and the negative charges increase with the increase of pH [22]. According to the Donnan exclusion theory, a positively charged membrane (pH < IEP) is beneficial for the separation of Mg^{2+} and Li^+ from brine, and the Donnan exclusion on rejection decreases as the counter-ion valance increases (pH > IEP). As the pH of the solution is adjusted with HCl, the lower the pH of the solution is, the higher the H^+ concentration will be. Thus, the negative charges on the surface of NF membranes can be screened and then turned to positive charges by H^+, and thus Mg^{2+} and Li^+ will be repelled by the positively charged surface. However, the electric potential at the separation interface resulting from a large amount of Cl^- through the membrane can drive Li^+ to pass through the membrane to maintain electroneutrality [6]. The Donnan exclusion can be reduced as the pH increases, leading to a decrease of R(Mg^{2+}) and an increase of R(Li^+). The dielectric exclusion (DE) also plays a crucial role in the separation of Mg^{2+} and Li^+ from brine, which is partly due to ions with different dielectric constants at the interface of external solution and pore solution of membranes. The exclusion energy is proportional to the square of ion charges including anions and cations, and the concentrations of H^+ and total ions are higher at a lower pH. Thus, DE can contribute to the separation of Mg^{2+} and Li^+ at lower pH, and this effect becomes less pronounced with increasing pH. Finally, according to the sieving effect of NF membranes, Mg^{2+} has a larger size than Li^+, and Li^+ is a monovalent ion which can pass through the NF membrane more easily than the divalent ion (Mg^{2+}). The surface of NF2 membrane is much denser than that of NF1 membrane, resulting in better separation effect.

3.1.3. Effect of the Mg^{2+}/Li^+ ratio

Fig. 5 shows the R(Mg^{2+}), R(Li^+) and SF as a function of the Mg^{2+}/Li^+ ratio of simulated brine. It shows that as the Mg^{2+}/Li^+

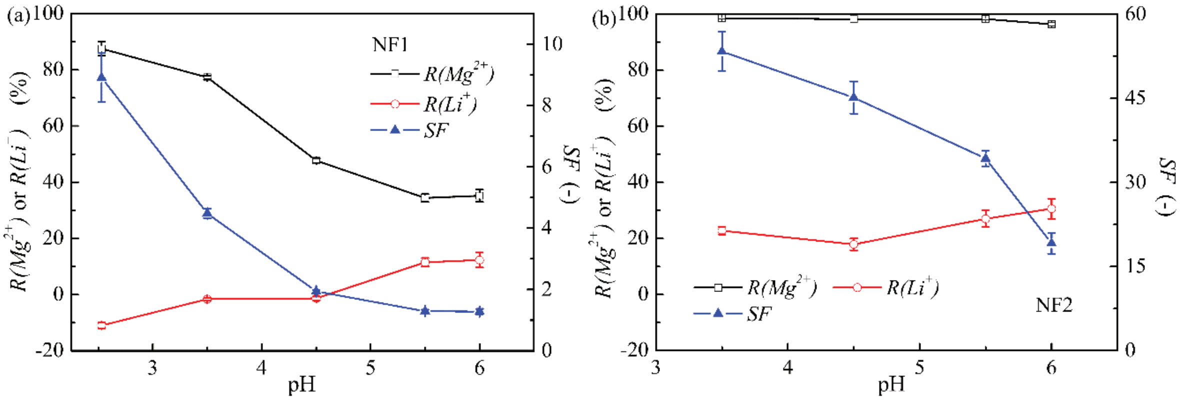

Fig. 4. Effect of simulated brine pH on the SF and rejection rate of Mg^{2+} and Li^+ ($Mg^{2+}/Li^+ = 3.43$, $\Delta P = 0.8$ MPa), (a) NF1, (b) NF2.

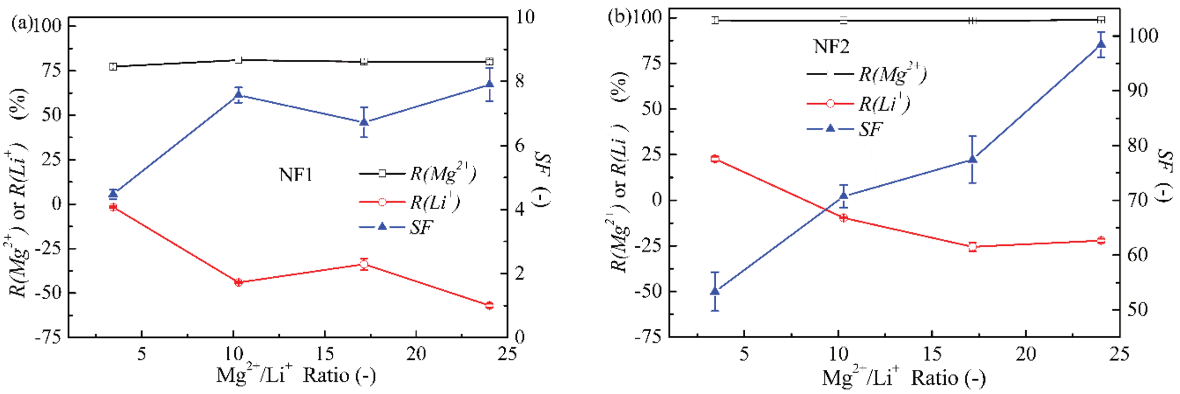

Fig. 5. Effect of the Mg^{2+}/Li^+ ratio of simulated brine on SF and rejection rate of Mg^{2+} and Li^+ (pH = 3.5), (a) NF1, (b) NF2.

ratio increases, R(Mg^{2+}) remains largely unchanged but R(Li^+) decreases continuously. Thus, SF increases with increasing Mg^{2+}/Li^+ ratio of the simulated brine. This is particularly pronounced in the second NF stage, in which R(Mg^{2+}) can reach above 99.0% and R(Li^+) is decreased to –20.0%, and the SF can reach 98 as the Mg^{2+}/Li^+ ratio of the feed solution increases. This is because as the Mg^{2+}/Li^+ ratio increases, the Mg^{2+} concentration and the exclusion energy increase, resulting in a stronger DE and a higher SF [6]. Thus, a high separation efficiency of Mg^{2+} and Li^+ could be expected after the two-stage NF process, which will be discussed below.

3.1.4. Mg^{2+}/Li^+ ratio of permeate

Fig. 6 shows the Mg^{2+}/Li^+ ratio of permeate from simulated brine with different mass concentration ratios of Mg^{2+} to Li^+ (3.43 to 24.01) at pH = 3.5 and 4.5, respectively. Li^+ permeate preferentially to maintain electroneutrality compared with Mg^{2+}, because they have a smaller Stokes radius and a lower valence than Mg^{2+}. As a result, Mg^{2+} and Li^+ can be fractionated by NF1 and NF2 membranes because of ion competition. The Mg^{2+}/Li^+ ratio of permeate decreases and then tends to be stable as the transmembrane pressure increases, which increases with increasing Mg^{2+}/Li^+ ratio and pH of the simulated brine. For NF2 process, the Mg^{2+}/Li^+ ratio of permeate can be reduced to 0.14 at pH = 3.5. This can be explained by the combined effects of Donnan exclusion, DE and steric hindrance that govern the mass transport inside the NF membrane [6]. The lower the pH, the more the number of positively charges on the surface or in the interior of the membrane. As a result, the Donnan exclusion and DE result in a higher separation efficiency. In addition, the R(Mg^{2+}) and R(Li^+) increase with the increase of transmembrane pressure, because increasing volume permeate flux can cause a decrease in permeate concentration (Eq. (5)). Thus, the two-stage NF process may be a promising approach for the separation of Mg^{2+} and Li^+ from brine.

3.2. Separation of Mg^{2+} and Li^+ from salt lake brine

The pH of salt lake brine (originally equal to feed brine) is adjusted to 3.5 with HCl. Fig. 7 shows that the Mg^{2+}/Li^+ ratio of the salt lake brine declines after the two-stage NF process. Especially, the Mg^{2+}/Li^+ ratio of the permeate is decreased from 13.25 to 7.03 in the first NF stage and then to 0.17 in the second NF stage at pH = 3.5, while only a slight decrease is observed at pH = 6.2. This is because the surface of NF1 and NF2 membranes is positively charged at pH = 3.5. The DE

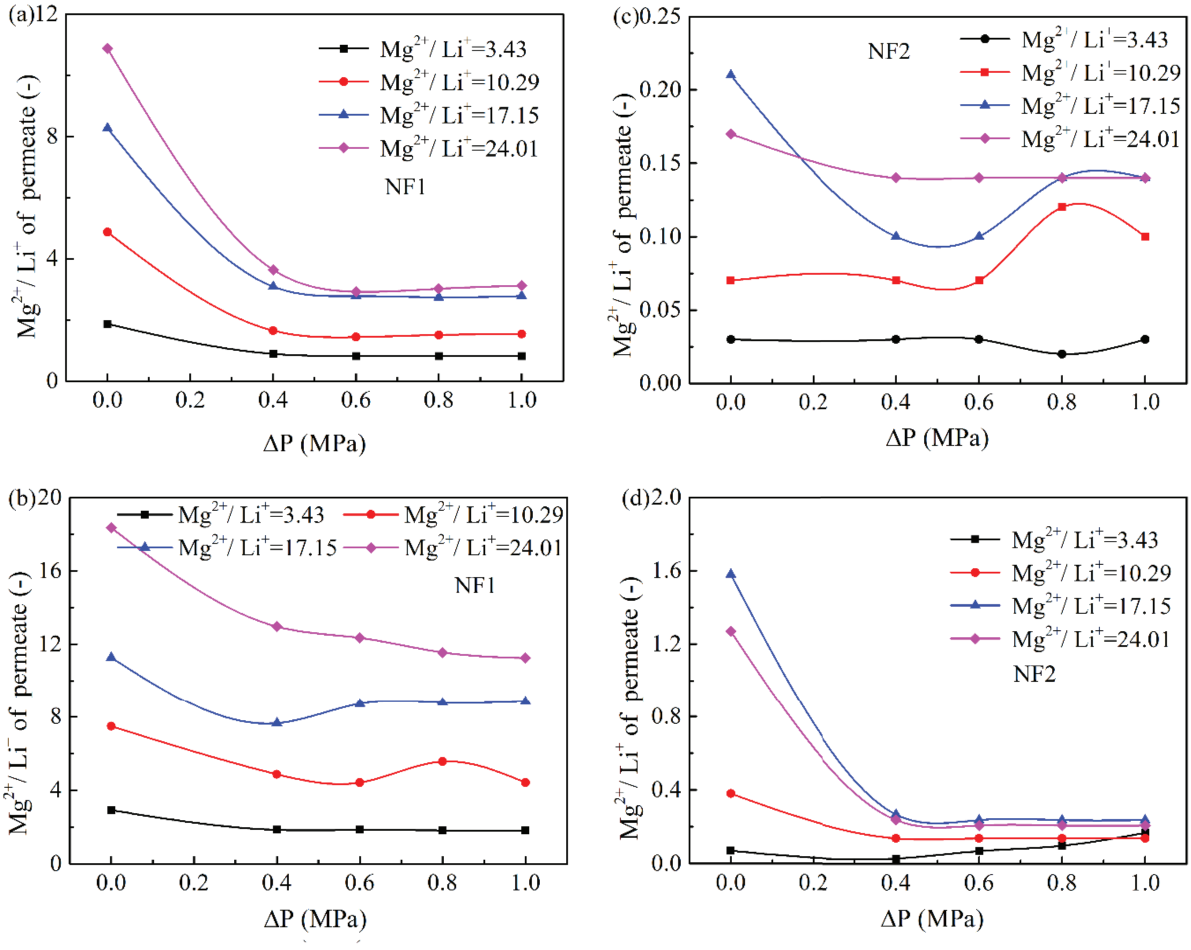

Fig. 6. Effect of transmembrane pressure on the Mg^{2+}/Li^+ ratio of permeate, (a) NF1/pH = 3.5, (b) NF1/pH = 4.5, (c) NF2/pH = 3.5, (d) NF2/pH = 4.5.

and Donnan principle can enhance the separation of Mg^{2+} and Li^+ from brine. The brine with a small Mg^{2+}/Li^+ ratio of 0.17 can be treated directly with extraction or other traditional treatment processes.

3.3. Separation stability

The separation stability of the two-stage NF process was evaluated by RF and Mg^{2+}/Li^+ ratio of the brine permeate. Fig. 8 shows RF and Mg^{2+}/Li^+ ratio of the brine permeate for both NF membranes during the four filtration cycles. In each circle, RF of NF1 and NF2 membranes at pH = 6.2 increases with operating time at the beginning because the membranes are compacted at the beginning of the filtration. After the attainment of a stationary thickness, RF changes slightly in the range of 0.97–1.02, indicating that the two membranes have a stable filtration performance. However, RF of both membranes at pH = 3.5 decreases gradually with operating time. After washing with DI water, NF1 and NF2 membranes show a stable RF value of 0.89 and 0.81, respectively. On the one hand, as the pH of the solution changes from 6.2 to 3.5, the surface of NF membranes becomes positively charged. Thus, the repulsive force of Mg^{2+} and Li^+ increases due to Donnan exclusion, leading to an increase in filtration resistance; and the osmotic pressure of the solution increases as the pH decreases. On the other hand, the membrane matrix would be in a less expanded state at a lower pH because of the lower intramembrane electrostatic repulsion [20,23–25]. Therefore, RF of both membranes at pH = 3.5 is lower than that at pH = 6.2. Fig. 8(b) shows that the Mg^{2+}/Li^+ ratio of NF permeate hardly changes with operating time in each circle and the Mg^{2+}/Li^+ ratio decreases with decreasing pH, which can be attributed to the combined effects of Donnan exclusion and DE [6]. These results confirm that the two-stage NF process has a good separation stability.

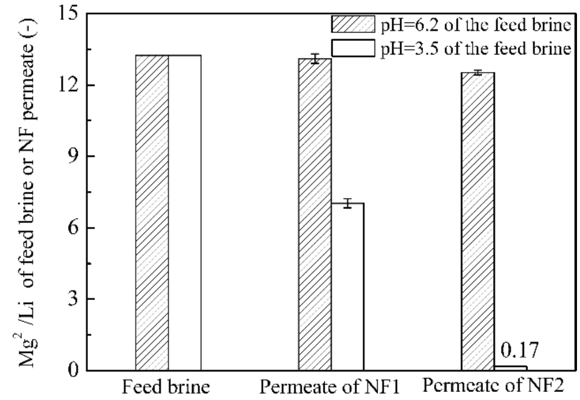

Fig. 7. Mg^{2+}/Li^+ ratio for feed brine and permeate of NF1 and NF2.

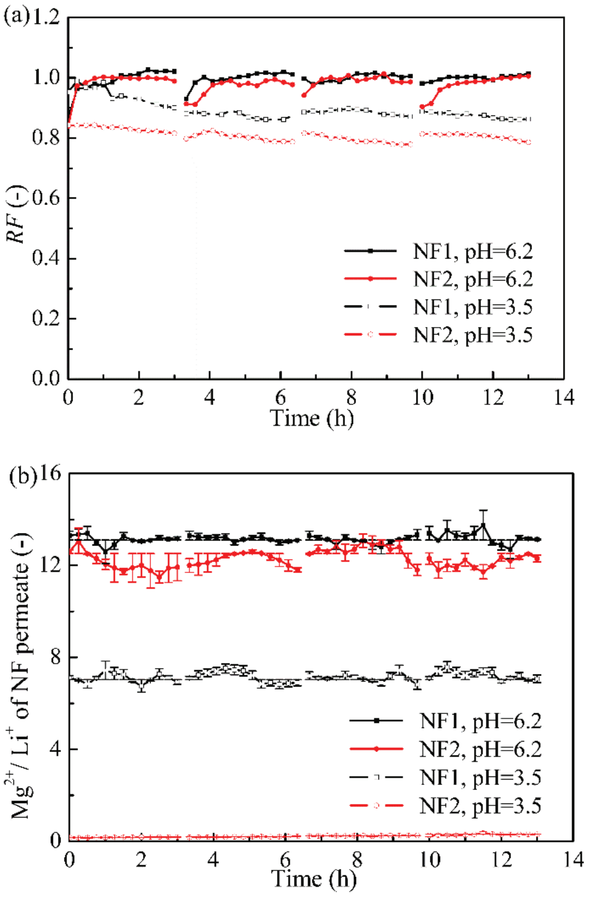

Fig. 8. RF (a) and Mg^{2+}/Li^+ ratio of NF permeate (b) as a function of operating time.

4. Conclusions

A two-stage NF process was proposed in this study for the separation of Mg^{2+} and Li^+ from brine with a high Mg^{2+}/Li^+ ratio. Both NF1 and NF2 membranes have remained well throughout the NF process. As the transmembrane pressure increases, R(Mg^{2+}) and R(Li^+) vary slightly for the NF1 process; whereas R(Mg^{2+}) remains almost constant, but R(Li^+) increases sharply at first and then levels off for the NF2 process. pH plays a critical role in the two-stage NF process. R(Mg^{2+}) decreases with increasing pH, whereas the opposite is observed for that of Li^+. Accordingly, the SF of Mg^{2+} and Li^+ decreases sharply with the increase of pH. The lower the pH is, the better the separation of Mg^{2+} and Li^+ will be. The SF increases as the Mg^{2+}/Li^+ ratio of feed solution increases. The Mg^{2+}/Li^+ ratio of permeate of simulated brine and brine obtained from West Taijnar salt lake can be reduced to 0.14 and 0.17 at pH 3.5, respectively. The long-term filtration test confirms that the two-stage NF process is quite stable. The effects of other cations and anions in brine on the separation of Mg^{2+} and Li^+ as well as the accurate economic analysis should be studied in the future. The Mg^{2+} and Li^+ in salt lake brine may be completely separated with a positively charged NF membrane.

Acknowledgements

This research is financially supported by the National Natural Science Foundation of China (U 1507123), the Foundation from Qinghai Science and Technology Department (2017-HZ-803), Thousand Talents Program of Qinghai Province, the open project of the State Key Laboratory of Chemical Engineering (SKL-ChE-15A05) and the Young and Middle-aged Fund of Qinghai University (2015-QGY-1).

Symbols

A — Active surface area of membrane, m^2
B — Permeation coefficient, $m^4 \cdot kg^{-1}\ s^{-1}$
C_{fi} — Concentration of i in feed, $g \cdot L^{-1}$
C_{pi} — Concentration of i in permeate, $g \cdot L^{-1}$
J_v — Volume permeate flux, $m^3 \cdot m^{-2} \cdot s^{-1}$
J_{v0} and J_{v2} — Volume permeate flux of DI water before and after long-term filtration test, $m^3 \cdot m^{-2} \cdot s^{-1}$
J_{v1} — Volume permeate flux of brine, $m^3 \cdot m^{-2}\ s^{-1}$
R — Rejection rate, %
RF — Relative flux, no unit
SF — Separation factor, no unit
V — Volume of permeate solution, m^3
ΔC_S — Concentration difference of salt on the sides of the membrane, $g \cdot L^{-1}$
Δt — Permeation time, s
i — Mg^{2+} or Li^+

References

[1] B. Ammundsen, J. Paulsen, Novel lithium-ion cathode materials based on layered manganese oxides, Adv. Mater., 13 (2001) 943–956.

[2] Navigant research, The Global Market for Lithium Ion Batteries for Vehicles Is Expected to Total $221 Billion from 2015 to 2024. http://www. navigantresearch.com/ (2016).

[3] J.W. An, D.J. Kang, K.T. Tran, M.J. Kim, T. Lim, T. Tran, Recovery of lithium from Uyuni salar brine, Hydrometallurgy, 117 (2012) 64–70.

[4] G.P. Zhang, W. Qin, Y.Z. Tan, Y.Y. Dai, Separation of magnesium and lithium by solvent extraction using di-(2-ethylhexyl) phosphoric acid (D2EHPA), J. Tsinghua Univ. Sci. Technol., 50 (2010) 430–433.

[5] G.P. Xiao, K.F. Tong, L.S. Zhou, J.L. Xiao, S.Y. Sun, P. Li, J.G. Yu, Adsorption and desorption behavior of lithium ion in spherical PVC-MnO_2 ion sieve, Ind. Eng. Chem. Res., 51 (2012) 10921–10929.

[6] X.M. Wen, P.H. Ma, C.L. Zhu, Q. He, X.C. Deng, Preliminary study on recovering lithium chloride from lithium-containing waters by nanofiltration, Sep. Purif. Technol., 49 (2006) 230–236.

[7] B. Van der Bruggen, C. Vandecasteele, Removal of pollutants from surface water and groundwater by nanofiltration: overview of possible applications in the drinking water industry, Environ. Pollut., 122 (2003) 435–445.

[8] Y.F. Song, T.M. Li, J.G. Zhou, Z.Y. Li, C.J. Gao, Analysis of nanofiltration membrane performance during softening process of simulated brackish groundwater, Desalination, 399 (2016) 159–164.

[9] H. Nabetani, Development of a membrane system for highly concentrated fruit juice, Membrane, 21 (1996) 102–108.

[10] S.Y. Sun, L.J. Cai, X.Y. Nie, X.F. Song, J.G. Yu, Separation of magnesium and lithium from brine using a Desal nanofiltration membrane, J. Water Process Eng., 7 (2015) 210–217.

[11] G. Yang, H. Shi, W.Q. Liu, W.H. Xing, N.P. Xu, Investigation of Mg^{2+}/Li^+ separation by nanofiltration, Chin. J. Chem. Eng., 19 (2011) 586–591.

[12] Q.Y. Bi, Z.Q. Zhang, C.Y. Zhao, Z.Q. Tao, Study on the recovery of lithium from high Mg^{2+}/Li^+ ratio brine by nanofiltration, Water Sci. Technol., 70 (2014) 1690–1694.

[13] W.R. Bowen, J.S. Welfoot, Modeling the performance of membrane nanofiltration-critical assessment and model development, Chem. Eng. Sci., 57 (2002) 1121–1137.

[14] J. Sabate, J. Labanda, J. Llorens, Influence of coion and counterion size on multi-ionic solution nanofiltration, J. Membr. Sci., 345 (2009) 298–304.

[15] X.L. Wang, W.N. Wang, D.X. Wang, Experimental investigation on separation performance of nanofiltration membranes for inorganic electrolyte solutions, Desalination, 145 (2002) 115–122.

[16] R. Levenstein, D. Hasson, R. Semiat, Utilization of the Donnan effect for improving electrolyte separation with nanofiltration membrane, J. Membr. Sci., 116 (1996) 11–92.

[17] L.C. Li, B.G. Wang, H.M. Tan, T.L. Chen, J.P. Xu, A novel nanofiltration membrane prepared with PAMAM and TMC by in situ interfacial polymerization on PEK-C ultrafiltration membrane, J. Membr. Sci., 269 (2006) 84–93.

[18] Y.Z. Xu, R.E. Lebrun, Investigation of the solute separation by charged nanofiltration membrane: effect of pH, ionic strength and solute type, J. Membr. Sci., 158 (1999) 93–104.

[19] M. Nilsson, G. Trägårdh, K. Östergren, The influence of pH, salt and temperature on nanofiltration performance, J. Membr. Sci., 312 (2008) 97–106.

[20] J.J. Qin, M.H. Oo, H. Lee, B. Coniglio, Effect of feed pH on permeate pH and ion rejection under acidic conditions in NF process, J. Membr. Sci., 1–2 (2004) 153–159.

[21] C. Ji, J. Zhang, Z.J. Zhang, S.Y. Sun, P. Li, J.G. Yu, Separation properties of magnesium and lithium from brine with high Mg^{2+}/Li^+ ratio by DK nanofiltration membrane, Membr. Sci. Technol., 34 (2014) 79–85 (in Chinese).

[22] C. Mazzoni, L. Bruni, S. Bandini, Nanofiltration: Role of the Electrolyte and pH on Desal DK Performances, Ind. Eng. Chem. Res., 46 (2007) 2254–2262.

[23] A.E. Childress, M. Elimelech, Relating nanofiltration membrane performance to membrane charge (electrokinetic) characteristics, Environ. Sci. Technol., 34 (2000) 3710–3716.

[24] M. Mänttäri, A. Pihlajamäki, M. Nyström, Comparison of nanofiltration and tight ultrafiltration membranes in the filtration of paper mill process water, Desalination, 149 (2002) 131–136.

[25] M. Mänttäri, A. Pihlajamäki, M. Nyström, Effect of pH on hydrophilicity and charge and their effect on the filtration efficiency of NF membranes at different pH, J. Membr. Sci., 280 (2006) 311–320.