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Metal Rejection by Nanofiltration from Diluted Solutions in the Presence of Complexing Agents

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ABSTRACT

Nanofiltration (NF) of solutions containing low levels of metal ions, in the absence and presence of complexing agents, using three polyamide membranes was investigated. For a given membrane, metal rejection increased with decreasing ion concentration and valence of metal ion in the absence of complexing agents. The rejections of 20 mg/L of Co^{2+}, Ni^{2+}, Mn^{2+}, Sr^{2+} (pH 4) were above 90% at an applied pressure of 65 psi with DK (MWCO, 150) and DL (MWCO, 300) membranes and near 15 to 20% with a GE (MWCO, 1000) membrane. In the presence of complexing agents, the rejection of metals was generally higher than those

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obtained in their absence. The rejection of metals increased with increasing solution pH and finally, reached a plateau beyond each certain pH value. Such pH-dependent rejections could be satisfactorily related to the overall formation constants between metal ions and complexing agents.

Key Words: Rejection; Metal ions; Nanofiltration; Complexing agents.

INTRODUCTION

The operation of nuclear fuel power plant steam generators often results in the formation of corrosion products on the walls of all boiler tubes. These corrosion products consist primarily of radioactive iron oxides and metallic copper. The components present on the surfaces also include nickel, zinc, chromium, manganese, and other types of impurities. The encrustation on the surfaces can reduce the performance of the generators. One of the methods dissolving the encrustation is to add strong complexing agents, usually ethylenediaminetetraacetic acid (EDTA), nitrilotriacetic acid (NTA), and citric acid.[1] Therefore, the boiler chemical cleaning water contains radioactive iron, nickel, manganese, and a certain amount of the complexing agents. The volume of low-level radioactive wastewater (LLRW) must be largely reduced, because it eliminates far-reaching "cradle to grave" liabilities and preserves space for the disposal of materials that require long-term isolation.[2]

A number of treatment methods have been applied to reduce the volume of liquid radioactive waste, particularly for LLRW, and to remove radionuclides from the waste. They include thermal treatment,[3] solvent extraction,[4] adsorption/ion exchange,[5] and membrane separation.[6,7] Thermal process could concentrate all species that are not evaporable including nonradioactive components with the radioactive ones and, however, need lots of energy for operation. Although solvent extraction methods are applied to recover and concentrate highly radioactive contaminants allowing disposal of process effluents as the LLRW, they might be uneconomical for the treatment of such solutions with a low level of metal ions. Some ion exchangers, such as clinoptilolite, have been studied for the decontamination of liquid wastes and the concentration of radioactive species. They reveal high removal selectivity for some ions, such as Cs^+ and Sr^{2+}; however, they are more expensive. The concentration of liquid wastes with low-level radioactivity by reverse osmosis (RO) and ion exchange membrane is possible, but the limitations, such as fouling of the membranes and the need for pretreatment, should be considered. In addition, RO needs high-operating pressure and is not selective in separating radioactive and nonradioactive species.

To overcome the above-mentioned drawbacks, the low-pressure membrane processes such as ultrafiltration (UF) and nanofiltration (NF) in combination with other physicochemical methods have been applied.[8–12] For example, Gaubert et al.[10,11] studied selective removal of Cs^+ and Sr^{2+} from a sodium nitrate solution by NF after the ions are complexed with water-soluble ligands resorcinarene and poly(acrylic) acid, respectively. Chihani et al.[12] also examined NF behavior of single Cu^{2+} and Ag^+ from the solutions containing NTA and sodium thiosulfate. On the other hand, Choo et al.[2] recently used NF membrane alone to selectively remove Co(II) species from simulated nuclear liquid wastewater. In this work, the NF of simulated nuclear cleaning solutions containing low levels of multimetal ions were performed. The effects of metal ions concentration, solution pH, and applied pressure on metal rejection, in the absence and presence of complexing agents (EDTA, NTA, citric acid), were investigated.

EXPERIMENTAL

Apparatus, Membranes, and Solutions

All NF experiments were carried out in batch stirred cell (Millipore, USA, Model 8400) with a capacity of 400 cm^3. Three NF filters with a disk type, DK, DL and GE (Osmonics Desal, Vista, California), were tested, all of which were thin-film composite polyamide membranes. The characteristics of the NF membranes are listed in Table 1. Only the membrane with a deviation of pure water flux, measured before and after NF, smaller than 5%, was repeatedly used in this work.

The chloride salts of metals ($ZnCl_2$, $NiCl_2$, $CoCl_2$, $SrCl_2$, $MnCl_2$, $FeCl_2$, $CrCl_3$), complexing agents (EDTA, NTA, citric acid), and other inorganic chemicals were all supplied by Merck Co., as analytical reagent grade. Two types of aqueous solutions were prepared; one contained each metal chloride

Table 1. Characteristics of the NF membranes used.

Properties	DK membrane	DL membrane	GE membrane
MWCO	150	300	1000
Diameter (cm)	7.6	7.6	7.6
Pure water flux[a] ($mL/m^2 \cdot min$)	50	70	90
Maximum pressure (psi)	65	65	65
pH range	2–11	2–11	2–11

[a]Measured after 5 min of operation at an applied pressure of 65 psi.

in deionized water (Millipore Mill-Q), the other consisted of metal chloride and complexing agent in water. The solution pH value was adjusted by adding a small amount of 0.1 mol/dm^3 HCl or NaOH.

Experimental Procedures

In NF experiments, the cell pressure was accurately monitored with N_2 gas by means of a transducer. The temperature was controlled at around 25°C by air conditioner. The feed volume was 250 cm^3 and the cell was stirred at 500 rpm using a magnetic motor. This speed was selected because it can lead to effective agitation but prevents formation of a serious vortex in the cell. The first 10 cm^3 of the permeate was discarded and the rejections of metals were obtained by analyzing the next permeate increment of 20 cm^3. Because the permeate composition slightly varied with filtration time, the rejections were integrally averaged in nature.

The solution pH values were measured using a pH meter (Horiba, Japan, Model F-23). The concentrations of metal ions in the feed, permeate, and retentate were determined by inductively coupled plasmas-atomic emission spectrometry (ICP-AES) (Jovin Yvon, France, Model JY-38S). The reproducibility of all concentration measurements was within 3% (mostly, 1%). The rejection of metal ions was calculated by

$$rejection\ (\%) = 100 \times \left(1 - \frac{[M^{m+}]_{permeate}}{[M^{m+}]_{retentate}}\right)$$

Ion concentrations in the final retentate were also used to check whether mass balance was fulfilled (often, within 3%). The used membranes were immediately flushed with deionized water after NF, and then regenerated in order by rinsing with 0.1 mol/dm^3 NaOH, 1.4×10^{-3} mol/dm^3 NaOCl, and 0.01 mol/dm^3 HCl in an ultrasonic cleaner (Brandson B20, USA) for 10 min each.

RESULTS AND DISCUSSION

Rejection of Single Metal Ions Without Complexing Agents

In the absence of complexing agents, the effect of applied pressure on metal rejection using DK, DL, and GE membranes are shown in Figs. 1 through 3. It was found that the rejection increases with increasing the pressure and then reaches a plateau up to 65 psi. Bowen and Mukhtar[13]

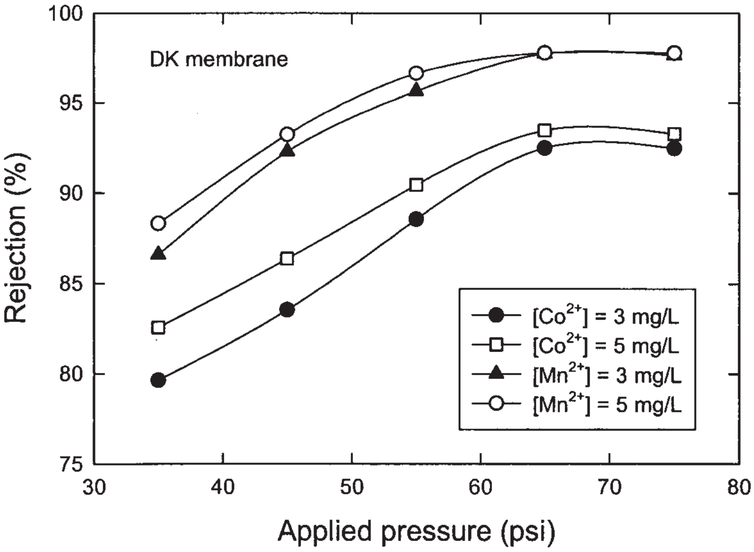

Figure 1. Effect of applied pressure and ion concentration on metal rejection in aqueous solutions using DK membrane (feed pH = 4.0).

reported a similar trend for NF of metal ions. In this work, the DK (MWCO, 150) and DL (MWCO, 300) membranes had nearly equivalent metal rejections. For example, a rejection of more than 90% was obtained for all metals studied at a pressure of 65 psi (see Figs. 1 and 2). However, the GE membrane (MWCO, 1000) revealed very low rejections (15 to 20%), even at 65 psi. Hence, the GE membrane was selected to further understand the role of added complexing agents because of its high permeate flux and incomplete rejection.

In NF, both charge and size effects as well as solution chemistry determine solute rejection.[12,14–16] The effect of ion valence on salt rejection follows the Donnan exclusion mechanism: the higher the valence of the co-ion, the higher the salt rejection; the higher the valence of the counter-ion, the lower the salt rejection. However, if the anion is univalent, salt rejection always increases with cation valence.[12] For the ions with the same valences or uncharged solutes, the size (steric) effect could be important.[14]

To examine the characteristics of metal rejection, experiments were conducted with various metal ions such as Co^{2+}, Ni^{2+}, Mn^{2+}, Fe^{2+}, and Cr^{3+}. The results are listed in Table 2. The ion size, estimated from partial molar hydration volume and hydration number,[17] decrease in the order $Cr^{3+} > Mn^{2+} > Fe^{2+} > Co^{2+} > Ni^{2+}$ (Table 3). This order is consistent with that of metal rejection with DL and DK membranes, as shown in Table 2. It was also

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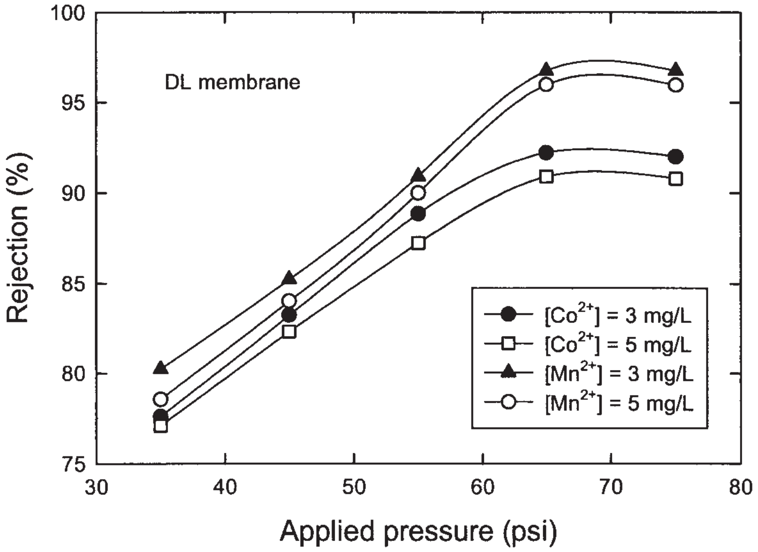

Figure 2. Effect of applied pressure and ion concentration on metal rejection in aqueous solutions using DL membrane (feed pH = 4.0).

found that the effect of ion size on ion rejection was not obvious with GE membrane but was more significant with DL and DK membranes, which is in good agreement with previous findings. Schaep et al.[14] studied the rejection of NaCl, Na_2SO_4, $MgCl_2$, $MgSO_4$, and $LaCl_3$ using NF40 (MWCO, 180), UTC20 (MWCO, 350), NTR7450 (MWCO, 600~800), and CA30 (MWCO, 1000) membranes. They found typical NF results using NF40 and UTC20 membranes: very high rejections for Na_2SO_4, $MgCl_2$, $MgSO_4$, and $LaCl_3$ and moderate rejection for NaCl. On the other hand, the rejection is lower with NTR7450 and CA30 membranes, and is mainly determined by the charge effect.

The dominant charge effect can explain why the rejection of Co^{2+} is higher than Mn^{2+} with a GE membrane but is reversed in the case of the DK and DK membranes (see Figs. 1 through 3), because Mn^{2+} has a larger size than Co^{2+}. Furthermore, the relatively high rejection of Cr^{3+} than the others with a GE membrane is probably due to the charge effect when the ions are much smaller than the pores.[18]

Figure 4 shows that the rejection with the GE membrane slightly decreased with increasing initial ion concentration. This is somewhat opposite to the earlier results for NF of divalent metal salts,[12] which is attributed to the use of a loose GE membrane in this work. However, Xu and Lebrun[19] studied

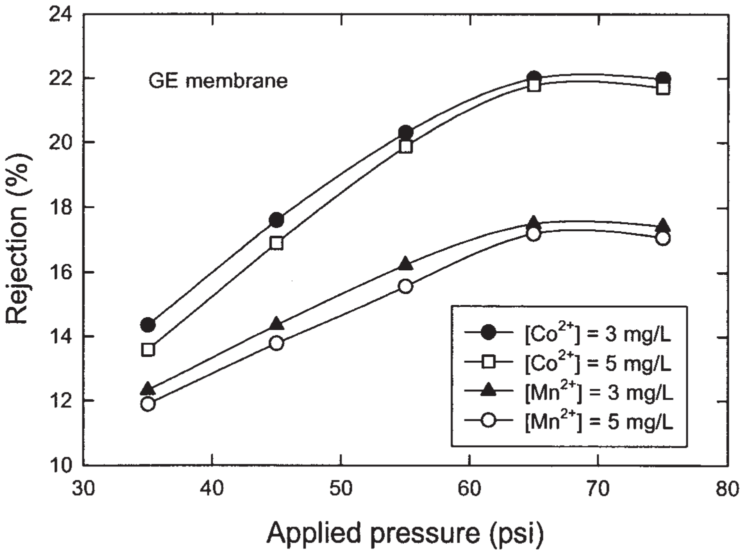

Figure 3. Effect of applied pressure and ion concentration on metal rejection in aqueous solutions using GE membrane (feed pH = 4.0).

ion separation by charged NF membranes and found that the rejection strongly depends on ion concentration; that is, there is a higher rejection at lower ion concentrations. In the NF of single electrolytes, Bowen and Mukhtar[13] also observed that metal rejection with PES5 membrane decreases with increasing electrolyte concentrations (NaCl and Na_2SO_4). On the other hand, Chio et al.[20] investigated the effect of co-existing ions and surface characteristics of NF membranes on the removal of nitrate and fluoride. They found that nitrate rejection with NTR7250 membrane is relatively stable from 95% to 94.1% when Mg^{2+} concentration increases from 2×10^{-4} to 8×10^{-4} mol/dm^3, whereas it changes from 96.7% to 87.5% with NTR7450 membrane.

Table 2. Rejection of Co^{2+}, Ni^{2+}, Mn^{2+}, Fe^{2+}, and Cr^{3+} using the three NF membranes.[a]

Membrane	Ni^{2+}	Co^{2+}	Fe^{2+}	Mn^{2+}	Cr^{3+}
DK	92.6	93.6	95.6	97.8	98.0
DL	91.2	95.8	96.2	96.8	97.6
GE	18.4	21.8	31.8	17.2	44.2

[a] Initial ion concentration = 20 mg/L, pH 4.0.

Table 3. Partial molar hydration volume and hydration number of the metals studied.[17]

Metal	Partial molar hydration volume (cm^3/mol)	Hydration number
Co^{2+}	169.6	11.5
Ni^{2+}	147.8	10.3
Fe^{2+}	174.5	10.3
Mn^{2+}	189.6	12.2
Sr^{2+}	156.7	10.4
Cr^{3+}	219.5	15.5

Rejection of Multimetal Ions with Complexing Agents

The effects of solution pH on metal rejection with a GE membrane in the presence of equimolar EDTA, NTA, and citrate are illustrated in Figs. 5 through 7, respectively. Although the rejection of EDTA alone is not given, it is expected to be nearly 100%, as shown in Fig. 5 at pH 11. The rejection of the complexed metals increased with increasing solution pH, and reached 1 when

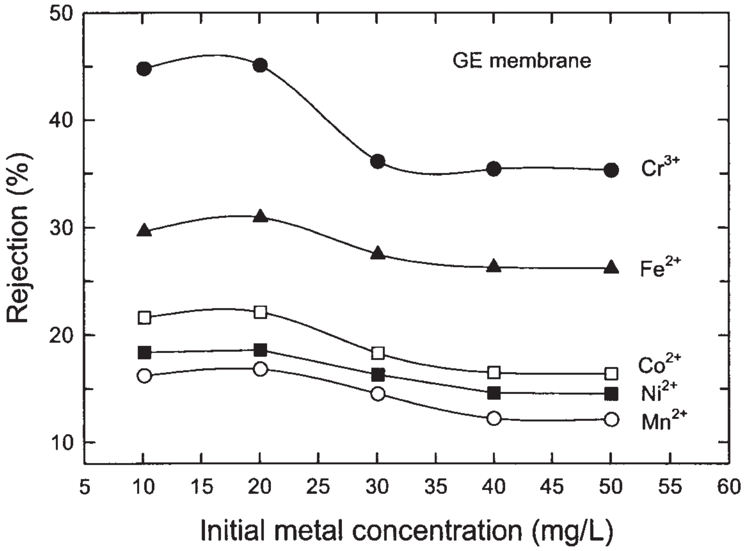

Figure 4. Effect of ion concentration on metal rejection in aqueous solutions using GE membrane (feed pH = 4.0, applied pressure = 65 psi).

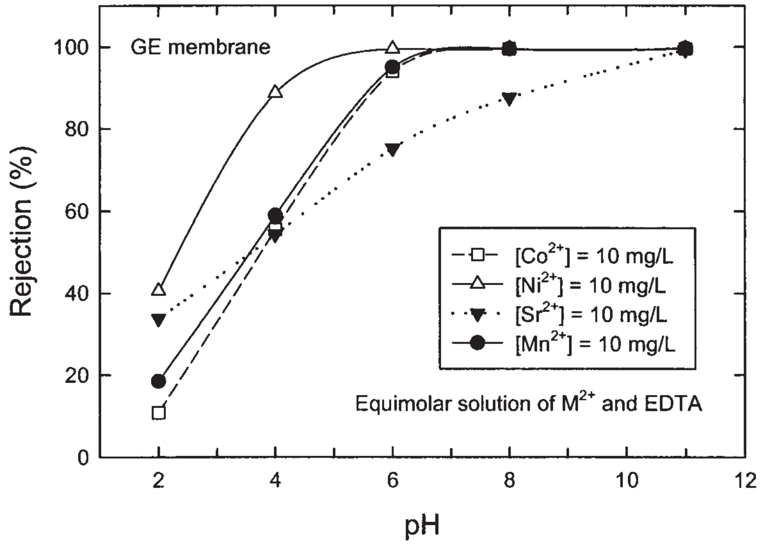

Figure 5. Effect of pH on metal rejection in the multi-ion solution containing EDTA using GE membrane (applied pressure = 65 psi).

the pH exceeded a threshold value. For example, the threshold pH was 4.5 for Ni^{2+}, 6.0 for Co^{2+} and Mn^{2+}, and 11 for Sr^{2+} in the EDTA systems (see Fig. 5). The behavior of Sr^{2+} was somewhat different from other metals because Sr^{2+} does not tend to react with OH^-.[21] The threshold pH values in the NTA and citrate systems were equivalent to those in the EDTA systems, as shown in Figs. 6 and 7. The pH-dependent nature of metal rejection can be understood from pH trends of species distribution. The overall formation constants (K_f) between metal ions and anionic ligands at zero ionic strength and 25°C are listed in Table 4.

It is known that EDTA, NTA, and citrate exist in a number of protonated forms in aqueous solutions.[21] They readily form stable complexes with most metal ions in a 1 : 1 molar ratio. The pH diagrams of species distribution can be calculated from a set of mass-balance equations.[22,23] In an equimolar solution of Cu^{2+} and EDTA (H_4L), it was reported that species CuL^{2-} predominates at $pH > 3.2$ and $CuHL^-$ at $pH < 3.2$.[22] Free Cu^{2+} ions were absent in the pH range tested. The anions CuL^- predominated absolutely at pH 2.0–11 and free Cu^{2+} ions at $pH < 2.0$ in the NTA (H_3L) system.[23] In the citric acid (H_3L) system, on the other hand, the dominant species were free Cu^{2+} ions at $pH < 4.0$ and CuL^- at $pH > 6.8$.[22] The neutral species CuHL dominated within pH 4.0 and 6.8.

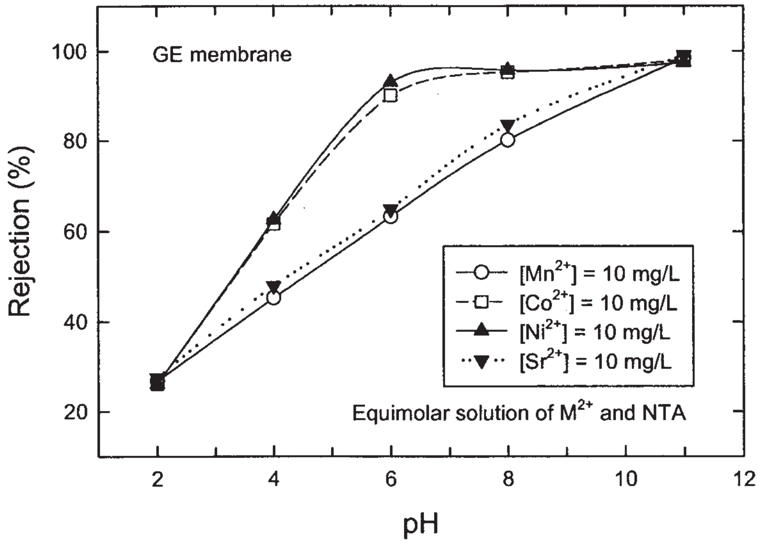

Figure 6. Effect of pH on metal rejection in the multi-ion solution containing NTA using GE membrane (applied pressure = 65 psi).

Although the speciation diagrams as a function of pH for Ni^{2+}, Mn^{2+}, Co^{2+}, and Sr^{2+} systems are unavailable from the literature, one can construct these diagrams from given overall formation constants.[22,23] It is expected that they are basically similar to those in the Cu^{2+} systems except that the dominating pH will somewhat shift because $\log K_f$ is different, as shown in Table 4. The complexing ability of metals with EDTA (i.e., $\log K_f$) decreases in the order $Ni^{2+} > Co^{2+} > Mn^{2+} > Sr^{2+}$. This means that the pH required when the 1 : 1 complex (ML^{2-}) predominates decreases in the order $Sr^{2+} > Mn^{2+} > Co^{2+} > Ni^{2+}$, which exactly reflects the order of the above threshold pH value. This is also the case in the NTA and citrate systems (see Figs. 6 and 7). This can be understood in that the rejection of metals obeys the steric effect when the metals are fully complexed to possess voluminous sizes.[10–12] It should be noted that the low rejection at low pH values is a result of a small extent of complexation of metal ions.

Comments on Separation Strategies

According to the present results, two separation strategies are proposed for the treatment of waste effluents containing complexing agents. First, if

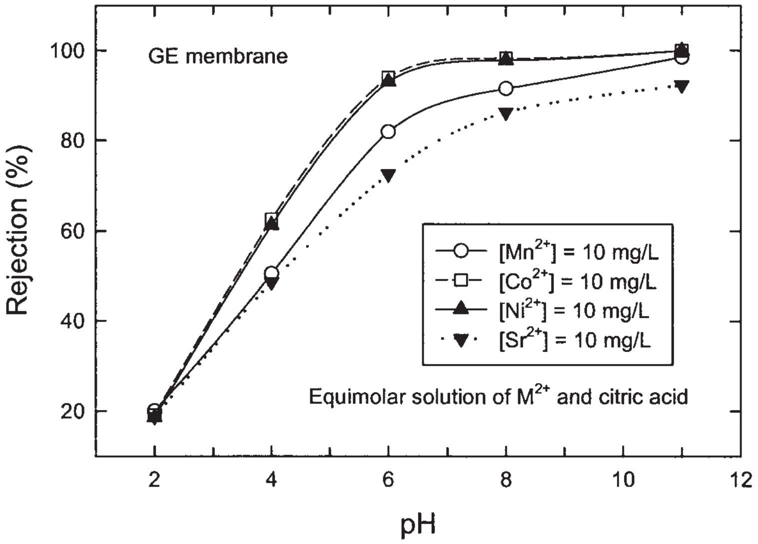

Figure 7. Effect of pH on metal rejection in the multi-ion solution containing citrate using GE membrane (applied pressure = 65 psi).

we plan to separate metal ions only from the solutions, NF will be performed at a sufficiently low pH using large-MWCO NF membranes (e.g., GE membrane). In this case, the metal ions pass through the membrane and the complexing agents are retained. Second, if we want to remove both metal ions and complexing agents, a high enough pH is needed. In this situation, the permeate can be directly discharged and the retentate should be treated by other alternatives, such as an electrochemical membrane process.[24] On the other hand, selective separation of multimetal ions by NF is possible by controlling solution pH with the help of suitable complexing agents.

CONCLUSION

The rejections of Co^{2+}, Ni^{2+}, Mn^{2+}, Fe^{2+}, Cr^{3+}, and Sr^{2+} from the solutions with and without EDTA, NTA, and citrate by NF were measured. In the absence of complexing agents, metal rejection increased with increasing applied pressure and reached a plateau up to 65 psi. When the membrane MWCO was beyond 300, metal rejection significantly decreased. The effect of ion size on ion rejection was more important with DL and DK membranes

Table 4. The overall formation constants ($\log K_f$) for selected complexes of metals and anionic ligands (L) at 25°C and zero ionic strength.[21] [a]

Ion	$L = OH^-$	$L = \text{citrate}^{3-}$	$L = \text{NTA}^{3-}$	$L = \text{EDTA}^{4-}$
H^+	HL (14.0)	HL (6.40)	HL (10.33)	HL (11.12)
		H_2L (11.16)	H_2L (13.27)	H_2L (17.80)
		H_3L (14.29)	H_3L (14.92)	H_3L (21.04)
				H_4L (23.76)
Sr^{2+}		SrL (4.1)	SrL (6.3)	SrL (10.5)
				SrHL (14.9)
Mn^{2+}	MnL (3.4)	MnL (5.5)	MnL (8.7)	MnL (15.6)
	MnL_2 (5.8)	MnHL (9.4)	MnL_2 (11.6)	MnHL (19.1)
Fe^{2+}	FeL (4.5)	FeL (5.7)	FeL (9.6)	FeL (16.1)
	FeL_2 (7.4)	FeHL (9.9)	FeL_2 (13.6)	FeHL (19.3)
Co^{2+}	CoL (4.3)	CoL (6.3)	CoL (11.7)	CoL (18.1)
	CoL_2 (9.2)	CoHL (10.3)	CoL_2 (15.0)	CoHL (21.5)
Ni^{2+}	NiL (4.1)	NiL (6.7)	NiL (12.8)	NiL (20.4)
	NiL_2 (9.0)	NiHL (10.5)	NiL_2 (17.0)	NiHL (24.0)
Cr^{3+}	CrL (10.0)			CrL (26.0)
	CrL_2 (18.3)			CrHL (28.2)
Cu^{2+}	CuL (6.3)	CuL (7.2)	CuL (14.2)	CuL (20.5)
	CuL_2 (11.8)	CuHL (10.7)	CuL_2 (18.1)	CuHL (23.9)

[a] K_f is defined as $K_f = [M_xL_y(OH)_z]/([M^{m+}]^x[L^{n-}]^y[OH^-]^z)$ e.g., for the complexation reaction $xM^{m+} + yL^{n-} + zOH^- \Leftrightarrow M_xL_y(OH)_z$. The concentration units used to calculate K_f are in mol/dm^3.

compared to the use of loose GE membrane (MWCO, 1000). The relatively high rejection of Cr^{3+} than other divalent ions with GE membrane was due to the charge effect.

In the presence of equimolar EDTA, NTA, or citrate, the metal rejection using GE membrane increased with increasing pH and reached 1 as the pH exceeded a threshold value. For example, the threshold pH was 4.5 for Ni^{2+}, 6.0 for Co^{2+} and Mn^{2+}, and 11 for Sr^{2+} in the EDTA systems. The ability for forming 1 : 1 complexes between metals and a given complexing agent reflected the order of the threshold pH value. The low rejection at a low pH value was due to negligibly small extent of complexation of metal ions.

Two NF separation strategies via solution pH control were proposed for treatment of solutions containing complexing agents. Metal ions could be separated from complexing agents only when NF was performed at a low pH using large-MWCO NF membranes. Otherwise, a sufficiently high pH was needed to remove both metal ions and complexing agents.

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