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EFFECT OF SALT COMPOSITION ON THE SEPARATION PROFILE OF CONTAMINATED GROUNDWATER IONS BY ELECTRODIALYSIS

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ABSTRACT: Chloride, nitrate, sulfate, and fluoride ions are sometimes available in groundwater at an elevated level, which is harmful to humans and the environment. The membrane separation technique has been widely explored for controlling the level of those ions in drinking water, particularly the electrodialysis method. In the present study, divalent (AMX) and monovalent ion selective (ACS) anion exchange membranes were used to remove the high concentrations of chloride and other co-existent ions from groundwater samples. The study focuses on the effect of the composition of the salt in the groundwater on the reducing profile of ions, the number of deionized ions, and the rate of ion removal in the diluting compartment. To achieve these experimental objectives, two series of ion samples composed of sodium and calcium were designed. The compositions of the ions were NaCl, Na_2NO_3, Na_2SO_4 for the Na-compound, and $CaCl_2$, $Ca(NO_3)_2.4H_2O$, $CaSO_4{\cdot}2H_2O$, and CaF_2 for the Ca-compound. The electrodialysis experiment was run at a constant applied voltage (8 volts), using sodium nitrate as the electrode solution. In summary, the ED performance for all the investigated parameters was better in the case of ions composed of sodium. Furthermore, a change in the applied current against the electrodialysis operation time was also observed. The results indicated that the current requirement by ED to separate ions composed of calcium needed to be slightly higher than that for ions composed of sodium.

Keywords: electrodialysis, groundwater, a sodium compound, a calcium compound, deionized ions

1. INTRODUCTION

The rapid growth of the population that is not provided for by the provision of adequate living facilities in some developing countries can have a negative impact on the environment. Human activities in the household or in industrial areas can produce wastewater that can contaminate groundwater, which is one of the raw water sources for drinking water. In some parts of the world, groundwater containing chloride, nitrate, sulfate, and fluoride ions exceeds the water standard quality recommended by WHO [1][2][3]. A high presence of chloride in drinking water is undesirable as it will add saltiness to the water. No previous study has investigated the harmful effect of excess chloride on health. However, a serious impact will be felt by those with certain medical conditions; for instance, high blood pressure, or heart, kidney and liver disease. It was further reported that the existence of chloride could increase the electrical conductivity of water, which will enhance the corrosion properties of metal pipes during water distribution. Inside the metal material, chloride reacts with metal ions to form soluble salts and increases the concentration of metal ions in drinking water [4][5].

The chloride ions in groundwater do not stand alone, as they are usually accompanied by the presence of other elements, such as nitrate, sulfate, and fluoride. The content of these elements in groundwater in excess concentrations can alter the soil ecosystem, damage the environment, and affect human health [6,7]. Considering the magnitude of the impact on health and the environment, a surplus of chloride and other ions in water must be removed prior to being used as drinking water. A variety of methods have been introduced in the effort to remove excess chloride and its associated ions in water. Electrodialysis (ED) is one of the appealing separation techniques for the removal of elevated ions in the water [8–10]. This membrane-based separation process occurs on the principle of ion transfer due to the difference in the electrical potential applied to the system. The ED technique has been attracting increased attention from researchers and manufacturers and has been widely applied in the water treatment field due to its eminence in comparison to other membrane separation techniques, such as microfiltration, ultrafiltration, and nanofiltration. The main advantages of ion removal

by ED are a reduction in the fouling problems and its high removal rate.

A thorough study on the concept of the ion transfer process in the ED system has been carried out by a group of researchers from Japan [11–13]. Studies on the removal of high concentrations of chloride removal in groundwater samples with electrodialysis techniques have been reported by researchers. [8]. In this study, the authors studied the effect of two types of bipolar membrane electrodialysis (BMED) produced by two different companies. The efficiency of chloride ion removal by electrodialysis reached 99% using both types of BMED. The separation of nitrate ions in groundwater samples using the electrodialysis process has also been achieved by observing the effect of an applied voltage and nitrate concentration in the feed solution. The largest nitrate ion removal was obtained at 98.5% under the ED process conditions with an applied voltage of 20 Volts [7].

Sulfate ions with a concentration of 750 mg/L in the sample water were treated by means of the electrodialysis process. Ion removal of around 77% was all that could be achieved by the ED process after 25 minutes with an applied voltage of 15 V [14]. A higher concentration of sulfate ions in the sample (1000 mg/L) was treated using the electrodialysis process with various conditions of applied current (0.2, 0.3 and 0.4 A). The highest removal efficiency of sulfate ions was obtained with an applied current of 0.4 A; namely, 88.6% after the ED process had been running for three hours [15].

The ED concept has also been extensively studied for its applicable use in the treatment process of groundwater and high fluoride brackish water[16,17]. The effect of the *electric voltage* over time on the removal of fluoride and nitrate ions using an ED stack consisting of 7 cation and 6 anion exchange membranes was investigated. It was found that the duration of ED to achieve the maximum rate could be significantly reduced with an increase in the applied current [18]. Based on a review of earlier research results it can be concluded that the ED process is more effective in reducing chloride, sulfate, nitrate, and fluoride content in water. A comprehensive study on the effect of applied current and membrane configuration in the ED stack on the separation of sulfate, nitrate, and fluoride ions has been reported previously [2,19]. It can be understood that an increase in the applied current may accelerate the completion of the ED process, and hybrid-membrane configurations in the stack can save energy by increasing the removal rate in shorter ED operation times.

The effect of salt compounds in groundwater samples is also important to consider in designing the best ED performance for water treatment plants. So far, however, there has been little discussion about the utilization of ED for the removal of chloride and other ions in groundwater in view of the different salt compounds. This study attempts to further analyze the influence of salt compounds in the diluted compartment on the removal rate of chloride, nitrate, fluoride, and sulfate ions in electrodialysis with two types of cation-anion exchange membrane. Cl-, NO_3, SO_4, and F- samples were prepared in two series, which is compounds with sodium and calcium.

2. EXPERIMENTAL

2.1 Materials

Analytical grade chemicals were used for all the ion samples and electrolyte solutions. For the Na-compound solutions, sodium fluoride (NaF), sodium chloride (NaCl), sodium nitrate ($NaNO_3$), and sodium sulfate (Na_2SO_4) were employed, which were supplied by WAKO Pure Chemical Industry, Osaka, Japan. For the ions that combined with Ca, calcium difluoride (CaF_2), calcium chloride ($CaCl_2$), calcium nitrate tetrahydrate ($Ca(NO_3)_2 \cdot 4H_2O$), and calcium sulfate dihydrate ($CaSO_4 \cdot 2H_2O$) were used. These four chemicals were also supplied by WAKO Pure Chemical Industry, Osaka, Japan. The $NaNO_3$ used as the electrode solution was also obtained from WAKO. The CMX-AMX and CMX-ACS, which represent the non-selective cation-anion exchange membrane and monovalent anion-selective membrane used in this study, were provided by Astom Corp. Tokyo, Japan. CMX-AMX membranes contain a standard anion exchange membrane with an ion exchange capacity of 2 – 3 meq/dry-g., whereas the CMX-ACS membrane contains a selective monovalent anion exchange membrane with an ion exchange capacity of 1.5 – 2.5 meq/dry-g.

2.2 Preparation of the Sample Solutions

Regarding the objective of this study, i.e., to investigate the effect of salt composition on the removal profile of F, Cl, NO_3, and SO_4 ions by electrodialysis, the feed solution was prepared by dissolving two different compounds of ions into the de-ionized water. The components and the composition of each ion in each sample are tabulated in Table 1. Furthermore, the samples with the concentrations shown in Table 1 were put into both the diluted tank and the concentrated tank.

Table 1 Compositions and concentrations of ions in water samples

Sample Series	Compounds	Ions	Concentration (mg/L)
1. Na-compounds	NaCl	Cl	100
	$NaNO_3$	NO_3	25
	Na_2SO_4	SO_4	500
	NaF	F	3
2. Ca-compounds	$CaCl_2$	Cl	100
	$Ca(NO_3)_2.4H_2O$	NO_3	25
	$CaSO_4 \cdot 2H_2O$	SO_4	500
	CaF_2	F	3

2.3 Design of Electrodialysis Module

A set of electrodialysis equipment using in this works is described in our previous paper [20,21]. The system consists of diluted and concentrated compartments for the sample and waste solution stock, an electrode compartment to supply the electrode solution, three resirculationrecirculation pumps, and a membrane stack. Ten sheets of cation-anion exchange membrane with an effective surface area of 55 cm2 were attached parralerlyparallelly in the stack. The system was also equipped with a flow meter (Flowmeter, Dwyer- RMB-SSV 5) and voltage/current recording devices. The separation process of four ions by electrodiayiselectrodialysis was carried out as the method previously described in our previous work [2,19]. The button controls the diluted solution, the concentrated solution, and the electrode solution flow was switched on simultaneously with the same flow rate of 1.2 L/min. The ED was run continuously at a constant applied voltage (8 volts) until the process ended when the applied current recorder showed the minimum (≈0.01A).

2.4 Reducing Ion Concentration

The ED performances were determined by measuring the reducing profile of all the ions in the diluting compartment and the increasing level of ions in the concentrating compartment. When the ED was running, one mL of the solution in the diluting compartment and concentrated compartment were collected every three minutes until the ED process was finished. The concentration of F, Cl, NO_3, and SO_4 ions in this solution were analyzed using an ion chromatograph (IA-300, DKK-TOA Corp., Japan). The ratio of the ions concentration in the diluted compartment at the time of ED running (C_{t_n}) to the ions concentration in the initial process (C_{t_0}) are calculated to describe the decreasing profile of the ions.

2.5 Deionized Ions

The number of deionized ions and migration from the diluting compartment is calculated using Eq (1).

$$n\ (\text{eq}) = Mt_0 - Mtn \quad (1)$$

Here, Mt_0 is the number of ions present in the feed compartment before the electrodialysis process is applied, and Mt_n is the number of ions present in the feed compartment when the electrodialysis proceeds at *n* minutes. Mt_0 and Mt_n are obtained by solving Eq. (2) and Eq. (3), respectively.

$$Mt_0\ (\text{eq}) = \left(\frac{C_{t0}}{M_w}\right) x\ V_{t_0} \quad (2)$$

$$Mt_n\ (\text{eq}) = \left(\frac{C_{t_n}}{M_w}\right) x\ V_{t_n} \quad (3)$$

In Eq. (2) and (3), Mw is the molecular weight of ions. Vt_0 is the volume of the solution in the feed tank before the ED process is executed, and Vt_n is the volume of the feed solution after the electrodialysis process operates at *n* minutes.

2.6 Separation Rate

In order to observe the rate of ion removal (η) in the overall ED process, the concentration of ions in the diluted tank at the end of the ED process (C_{t_f}) was also analyzed. The rate of ion removal was calculated using a formula proposed by other authors [15], as written in Equation (4). Here, V_{t_0} and V_{t_f} are denoted

as the initial volume (V_{t_0}) and the final volume (V_{t_f}) of the diluted solutions, respectively.

$$\eta\ (\%) = \frac{(C_{t_0} \times V_{t_0}) - (C_{t_f} \times V_{t_f})}{(C_{t_0} \times V_{t_0})} \times 100\ \% \qquad (4)$$

3. RESULTS AND DISCUSSION

3.1 Decreasing Ion Concentration in the Diluted Compartment

Figure 1 shows the reduction in concentrations of the Cl, NO_3, SO_4, and F ions in the diluted tank of the ED process with the ion samples in the form of sodium and calcium compounds. All the ED processes used either an AMX membrane (Fig. 1a) or an ACS membrane (Fig. 1b). In general, it can be clearly seen that, in all cases, the reducing concentration of Cl and other ions bound to sodium was easier than those bound to calcium. Observing Fig. 1 in detail, special note must be given to the fluoride and sulfate elements. The difference in the decreasing concentration of F ions between the Na and Ca compounds was significant in the ED process using the AMX membrane (Fig. 1a). Furthermore, the difference in the reducing concentration of SO_4 ions between the Na and Ca compounds was very large when the ED process was carried out with the ACS membrane (Fig. 1b).

The solutions of Na and Ca came from a strong base. The Na base strength was greater than that of the Ca so that the Na salt was more easily ionized. As a result, the ED operation took longer to separate the sulfate ions from the $CaSO_4$ compound than from the Na_2SO_4 compound. Investigations related to sulfate ions have been done by researchers from Ege University, Iran, to separate Na^+ and Ca^{2+} salts from a solution of Na_2SO_4 + $Ca(NO_3)_2$, and $NaNO_3$ + $Ca(NO_3)_2$ using an ED process with standard membranes and a potential of 10 V [22]. They concluded that to separate the Na^+ salt from the Na_2SO_4 solution the ED operation took longer than to separate it from the $NaNO_3$ solution. Thus, for the same ED operation time, the efficiency of the Na^+ ion separation was greater than the separation efficiency for the Ca^{2+} ion.

The decreasing tendency of this ion concentration may affect the operation time of the whole electrodialysis process. To acquire more detail concerning the relationship of the ion concentration reduction profile on the operation time, the requirement of the ED operation period to achieve a maximum of 90% ion reduction was calculated for the data shown in Fig. 1. The calculation of the time required to achieve a maximum 90% ion reduction is shown in Table 2.

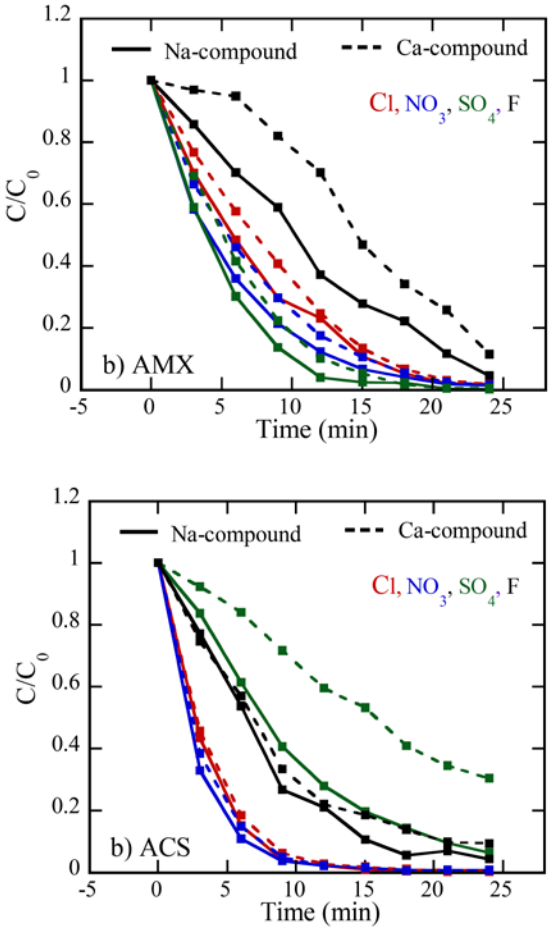

Fig. 1. Reducing the ion concentration in the diluted compartment against the ED operation time

Table 2 Electrodialytic time requirement to reject 90% of ions in the feed tank

Membrane Type	Ions	Time requirement (min) to reduce 90% of the ions	
		Na-Compound	Ca-Compound
CMX-AMX	Cl	15	16
	NO_3	13	16
	SO_4	10	13
	F	2	>24
CMX-ACS	Cl	7.5	8
	NO_3	5.5	7
	SO_4	20	>24
	F	18	21

Overall, for all ionic parameters, the time required to reduce 90% of the ions using either an AMX or an

ACS membrane was longer when the ion sample was in the form of a Ca-compound than in the form of a Na-compound. Furthermore, Table 2 also shows that the time required to reduce 90% of the F ions was longer than for the other ions when the F ion was arranged in the form of a Ca-compound in the ED process using an AMX membrane. In addition, it took much longer for the reduction of 90% SO_4 ions when the sample was composed of a Ca-compound in the ED process using the ACS membrane. The difference in the rate of reduction of these ions was related to the ability of the ions to break away from the binding compound and migrate to the concentrating compartment. The order of the ion reduction rate in the diluted compartment for the AMX membrane is SO_4> NO_3> Cl> F, and for the ACS membrane it is NO_3> Cl> F> SO_4 [22,23].

3.2 Deionized Ions Profile

When the electrodialysis process is initiated, the chemical compounds present in the diluted compartment decompose into ions and migrate to the concentrated compartment as a waste solution. The number of ions moving from the diluted compartment to the concentrated compartment depends on the concentration and composition of the ions in the feed solution, the type of membrane used, as well as the applied voltage.

In this study, the effect of the composition and ionic compounds in the sample solution was studied regarding the type of membrane used for the deionizing ability of Cl, NO3, SO4, and F ions. The applied voltage was set constant at 8 volts for all the investigating variables. The amount of deionized ions from the diluted compartment in the electrodialysis process using two different types of the membrane with ionic compositions composed of sodium and calcium is shown in Fig. 2.

In the sample composed of sodium, the number of deionized Cl- ions in the ED process using an AMX membrane or an ACS membrane is approximately similar. Additionally, the number of deionized NO3, SO4, and F ions in the AMX or ACS system are almost the same. However, the number of deionized ions changes when the sample solution contains ions composed of calcium cations. In the ED process using an AMX membrane, there is a difference in the number of deionized ions for F ions. As can be seen from the detail of the enlarged Y-axis scale for the F ions in Fig. 2a, the smaller number of deionized F ion occurs when the sample solution contains ions with a Ca-compound. For the ED using an ACS membrane, it was found that the number of SO4 ions deionized was smaller when the sample was arranged in the form of Ca-compound (Fig. 2b).

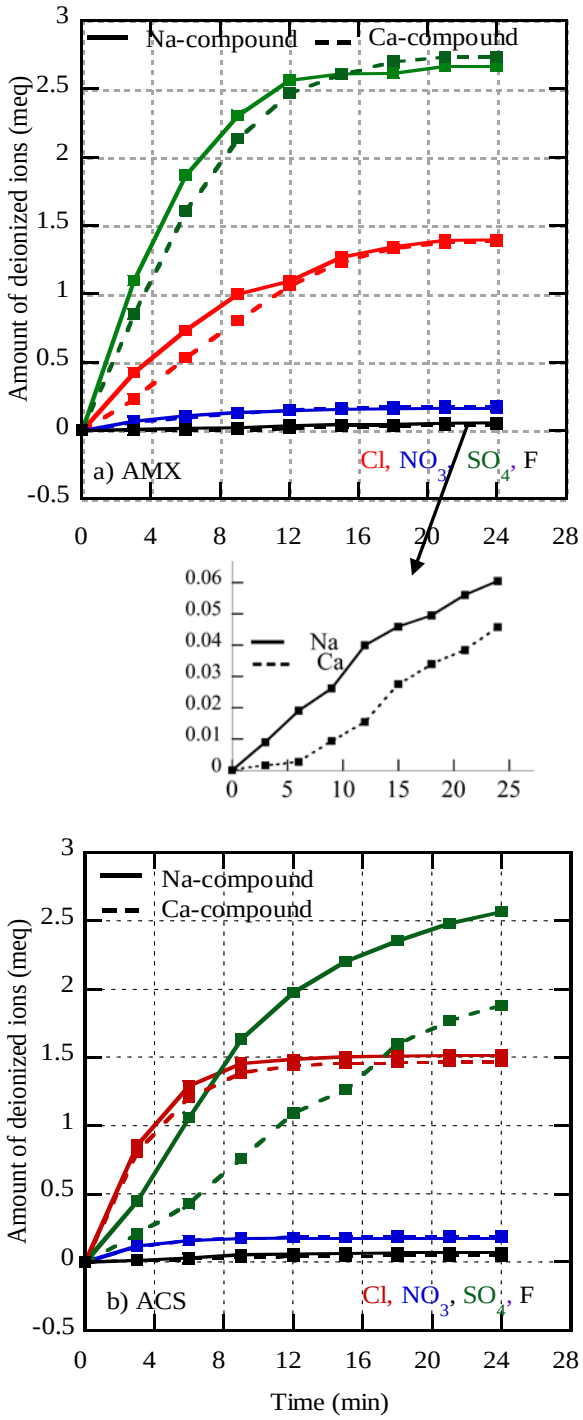

Fig. 2 Number of deionized ions versus ED operation time

3.3 Separation Rate

To determine the correlation between the type of compound and the removal efficiency of the Cl, NO_3, SO_4, and F ions, the ED experiment processes were designed to use the samples from series 1 and series 2; as shown in Table 1. The ED processes were stopped when the applied current was already at the minimum value, i.e., 0.02 A.

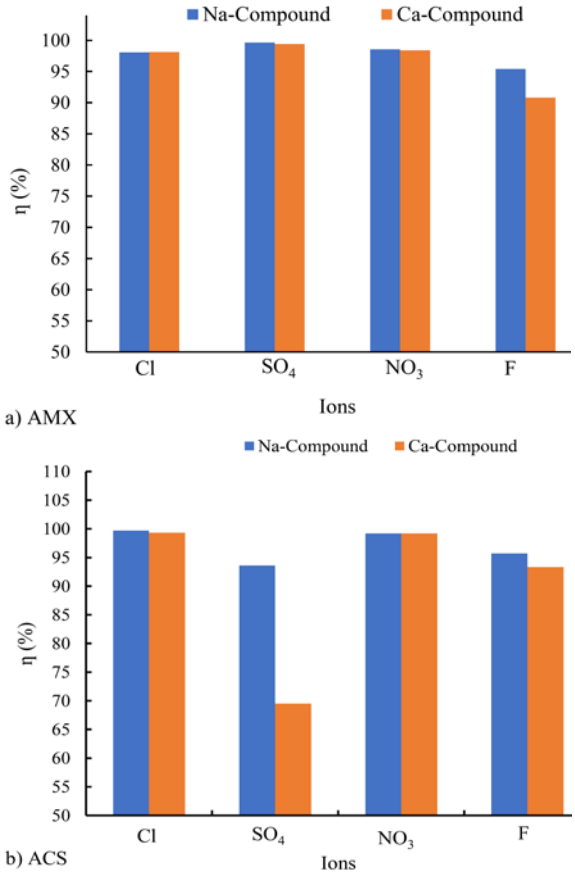

Fig. 3. The rate of ion removal with different sample compounds treated using AMX and ACS membranes

The removal efficiencies of all the ions with the samples of the sodium compounds and calcium compounds are shown in Fig. 3. Cl and NO_3 ions can be completely separated up to 98% through the ED process with the AMX membrane for either sample in the form of Na-compound or Ca-compound. In addition, both these ions can be eliminated by up to 99.30% from the diluted compartment in the ED process using an ACS membrane for both the Na- and Ca-compound samples. In both forms of sample composition, SO_4 ions can also be eliminated to a maximum of 99.40% by the ED process using an AMX membrane (Fig. 3a). However, SO_4 ion separation is very hard during the ED process using an ACS membrane, especially when the sample solution is in the form of a Ca-compound (Fig. 3b). In the case of F ions, the highest separation rate that can be obtained is 95% by the ED process using both types of the membrane for the sample in the form of a Na-compound. The F ion separation rate decreases as the sample used in the diluting compartment is a Ca-compound, especially when using an AMX membrane.

The SO_4 ion sample used in this study was derived from the Na_2SO_4 and $CaSO_4.2H_2O$ compounds. The SO_4 ions of Na_2SO_4 compounds are more easily deionized than those derived from $CaSO_4.2H_2O$ compounds. The reason for this is because $CaSO_4.2H_2O$ is a hydrate compound that binds some water elements. In order to ionize $CaSO_4.2H_2O$, this compound needs to release the hydrated compound first. In the case of F ions, when this element binds to Na, the system requires less ionization energy than when binding to Ca^{2+}. Ca^{2+} requires 2 moles of element F^- whereas Na takes only 1 mole of F^-; thus, to release the bond of this compound requires less energy.

3.4 Current Requirement

Electrodialysis was designed at a constant applied voltage so that the applied current would change automatically during the ED process from the maximum to the minimum level depending on the concentration of ions in the diluted tank. The change in electric consumption was noted by the current recording devices (Astom Corp. Tokyo, Japan) every three minutes during the ED. The current requirement during the ED process to achieve the maximum level of ion reduction in the diluting compartment is shown in Table 3. The reducing ion concentration in the diluted compartment against the operational time of the ED process brought about a decrease in the current requirement. A small difference in the current needed was found in the ED process when using an AMX membrane vs. an ACS, or when using ion samples with different salt compounds. Similar observations of the current needed were also presented in our previous investigation of electrolysis for groundwater treatment [2,19].

Table 3 Current requirement during ED process using AMX and ACS membranes with different ion compounds

ED run time (min)	AMX membrane (A)		ACS membrane (A)	
	Na-Compound	Ca-Compound	Na-Compound	Ca-Compound
0	0.14	0.14	0.14	0.14
3	0.12	0.12	0.10	0.10
6	0.09	0.09	0.08	0.08
9	0.05	0.06	0.06	0.06
12	0.03	0.04	0.04	0.05
15	0.02	0.02	0.03	0.04
18	0.01	0.02	0.03	0.03
21	0.01	0.02	0.02	0.03
24	0.01	0.02	0.02	0.03

4. CONCLUSIONS

The separation of Cl, NO_3, SO_4, and F ions from water samples composed of Na- and Ca-compounds was performed by electrodialysis using commercial AMX and ACS membranes. Investigation of the decreased ion concentration profile in the diluted compartment shows that the reduction of all the ions composed of the Na-compound occurs faster than for the ions composed of Ca-compound. In the ED process using an AMX membrane, the separation of F ions from the NaF compound is much faster than of the F ions composed of CaF_2 compounds. The F ion of the NaF sample can be reduced by 90% in the diluted compartment after the ED process has run for 22 min, while it takes over 24 min to reduce 90% F ion from the CaF_2 sample. The same tendency occurs for the SO_4 elements in the ED process using the ACS membrane. For ions from the $NaSO_4$ sample, the SO_4 ions can be reduced to 90% after the ED process runs for 8 minutes, whereas after 24 minutes, the SO_4 ion reduction of the $CaSO_4.2H_2O$ sample has not reached 90%.

The difference in the composition of the salt compounds in the groundwater samples also had an impact on the total ion removal rate in the ED process. Cl, and NO_3 ions could be separated from the diluted compartment by as much as 98% using both the AMX and ACS membranes for samples from the sodium and calcium compounds. SO_4 ions from the Na-compound and Ca-compound could be removed completely (99.40%) from the diluted compartment using the ED technique when using the AMX membrane. However, the rate of SO_4 removal was significantly lower when the ED process used the ACS membrane, particularly for the ions composed of the Ca-compound (69.5%). Overall, the design of the ED in this research was effective for use in reducing the Cl, NO_3, SO_4, and F ions from groundwater because it could remove ions with minimal current requirements (0.14 A).

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