

Electrodialysis to concentrate high-salinity solutions: The matching relation between cation- and anion-exchange membranes

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Abstract: The effect of the matching relation between the cation and anion exchange membranes on the electrodialysis (ED) concentration performance was investigated through evaluating the salt flux, water flux, the flux ratio of salt to water and the salt content of the ED concentrate. Results indicate that the water uptake of the cation exchange membrane (CEM) is a key factor for the ED concentration performance when the ion exchange capacity of CEM has a feasible value; while for the anion exchange membrane (AEM), the ion permeability maybe is more important compared with the ion exchange capacity and the water uptake for the ED concentration performance. Besides, CEM has a greater significance for the ED concentration performance compared with AEM when both membranes have a relatively high ion permeability.

Keywords: electrodialysis; cation exchange membrane; anion exchange membrane; matching relation; concentration performance

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1 Introduction

Electrodialysis (ED) is a mature desalination technology that can treat brackish water, seawater, RO concentrate, and high-salinity brines, in which water recovery and salt concentration can be achieved simultaneously^[1-4]. Due to this merit, ED has been used widely in the desalination and concentration of salinity streams^[2-4]. As is well known, ions, such as Na⁺ and Cl⁻ ions, are existed in the form of the hydrated ions in solutions^[5]. Therefore, the water molecular in the feed will be accompanied with ions to transport through ion exchange membranes (IEMs) to the concentrated solutions in the ED concentration process^[6]. Specifically, under a driving force of electrical potential difference cations and anions are transported from the dilute compartment to the concentrated one through the cation exchange membrane (CEM) and the anion exchange membrane (AEM), respectively^[1]. In this process, water transport will occur simultaneously^[7, 8]. This phenomenon is the so-called electro-osmosis. Generally, the water transport in the ED process refers mainly to electro-osmosis and

osmosis^[6, 9, 10]. The latter one is caused by the chemical potential difference (or osmotic pressure difference) between the dilute and concentrated compartments^[6]. Practically, a high water flux will dilute the concentrated solution in the ED concentration process, which is disadvantageous for the production of the concentrated salt solution^[6]. Hence, the water transport is a significant obstacle for the ED concentration process.

In the past decade, some reports have focused on the investigation of the water transport during the ED process^[6-14]. It was found that the water transport can be influenced by the intrinsic (i. e. membrane properties) and extrinsic factors (i. e. ED operating parameters). For instance, Berazina et al.^[11] reviewed the characterization of ion exchange membrane materials; properties vs structure and concluded that the water uptake in membranes is considered to be a key factor for the water transport in the electro-membrane system. Lu et al.^[7] investigated the water flow due to the osmosis effect during the ED process for preparing mineral source water from deep sea water, in which the concentrate and dilute tanks were filled with RO pure

water and LTVC (low temperature vacuum concentrator) concentrated brine respectively. They concluded that: ① for a lower DC voltage, the water flows from the concentrated to the dilute; ② for a higher DC voltage, the water flows from the concentrated to the dilute initially and flows reversely at a later stage. Izquierdo-Gil et al.^[12] investigated the effect of the membrane thickness and the electrolyte on the membrane water uptake. The authors found that the water uptake increases with the membrane thickness and decreases with the size of the cation. In other words, the membrane water uptake increases with decreasing membrane density and with the increasing hydration ability of the counterions. Rottiers et al.^[6] systematically investigated the influence of the type of AEM on the solvent flux (or the water transport) in ED of the concentrated NaCl solutions. The authors concluded that: ① electro-osmosis is the main contribution for the water transport, which depends only on the applied current density; and ② the concentration gradient between the concentrated and dilute solutions should be kept low to minimize the water transport caused by osmosis. Reig et al.^[14] found that the water transport is relatively low in a low temperature ED process, in which a relatively high concentration of the concentrated solution can be achieved but with relatively high energy consumption. Galama et al.^[13] investigated seawater pre-desalination with ED, and found that the water transport is influenced primarily by the membrane water uptake and an external salt concentration, and secondarily by temperature, the degree of crosslinking and the fixed charge density. Jiang et al.^[9] established a module for the water electro-transport (electro-osmosis) with hydrated cations in ED. The authors concluded that the water transport increases with an increase in the ion exchange capacity and hydrophilicity of membrane matrix materials. Han et al.^[8] investigated the ion hydrated number and electro-osmosis during ED of the mixed salt solution. The results indicated that the hydration numbers of Na^+ and Cl^- ions are 6 and 8 respectively for CMX and AMX membranes (Astom Co., Japan) at the studied conditions. It means that the flux ratio of water to NaCl ($J_{\text{H}_2\text{O}}/J_{\text{NaCl}}$) is around 14 in the ED separation process. Recently, Tedesco et al.^[10] established a module based on Nernst-Planck transport theory for water transport through IEMs in ED process. They showed, despite the extremely low water permeability of IEMs, the water flux in the membrane is not negligible and has the same order of magnitude of the counterions flux. Based on the above reports including experimental and theoretical investigations, it can be concluded that the water transport plays a significant role for the ED concentration performance,

and can be influenced by the CEM/AEM properties, such as the ion exchange capacity, water uptake, membrane structure, and membrane thickness, and operating parameters such as the feed concentration, running temperature and current density. Besides, the water transport can be influenced by the properties of cations/anions, such as valence, hydration number, and hydration free energy^[8]. Therefore, the AEM and CEM are different in controlling water transport.

However, few pay attention to the effect of the matching relation between CEM and AEM on the ED concentration performance especially for the water transport through IEMs. Which one of CEM and AEM plays a more important role on the ED concentration performance? Hence, in this work, different matching methods of CEMs and AEMs will be carried out to investigate the matching relation between CEM and AEM in the ED concentration process. Specifically, the salt flux (J_{NaCl}), water flux ($J_{\text{H}_2\text{O}}$) and the flux ratio of water to NaCl ($J_{\text{H}_2\text{O}}/J_{\text{NaCl}}$) will be investigated and discussed in detail.

2 Experimental section

2.1 Materials

Cation exchange membranes were CJMC-4, CJMC-5 and CJMC-5S, and anion exchange membranes were CJMA-4, CJMA-6, CJMA-7. The above membranes were provided by Hefei Chemjoy Polymer Materials Co., Ltd. Besides, the standard cation exchange membrane CMX (Astom Co., Japan) and the anion exchange membrane AMX (Astom Co., Japan) were used. Membrane properties are listed in Table 1. The chemicals used were of the analytical grade. Deionized water was used.

2.2 ED setup

Figure 1 shows the experimental setup and membrane stack for the ED concentration test. The setup was produced by Hefei Chemjoy Polymer Materials Co., Ltd. (CJED-9 × 21-5) and contained the following units: ① DC power supply (HSPY-60-10, Beijing Hanshengpuyuan science and technology Co., Ltd., China); ② ED membrane stack, in which electrodes were made of titanium coated with ruthenium; ③ electrode chamber fed with 500 mL 0.3 mol · L⁻¹ Na₂SO₄ as the electrode rinsing solution (ERS); ④ concentrate chamber fed with 200 mL 3% NaCl solution; ⑤ dilute chamber fed with 2000 mL 3% NaCl solution; and ⑥ peristaltic pumps (BT600L with 2 YT15 Pump head, Baoding Lead Fluid Technology Co. Ltd., China) for controlling the flow rate of various solutions precisely. Specifically, the membrane stack has six cation exchange membranes (CEMs) and five anion exchange

Table 1. Membrane properties.

Membrane	Support	Backbone	Ion exchange group	Thickness (μm)	IEC (meq/g)	Water uptake (%)	Area resistance ($\Omega \cdot \text{cm}^2$)	Transport number (%)
CMX ^[8]	Yes	DVB/PS	$-\text{SO}_3\text{Na}$	170–190	1.5–1.8	25–30	2.0–3.5	98
CJMC-4 ^[15]	No	PVDF	$-\text{SO}_3\text{Na}$	90–100	0.8–1.0	35–40	1.5–2.5	>93
CJMC-5 ^[4]	Yes	PVDF	$-\text{SO}_3\text{Na}$	150–160	1.0–1.2	25–27	2.0–2.5	>96
CJMC-5S ^a	Yes	PVDF	$-\text{SO}_3\text{Na}$	150–160	1.4–1.6	40–43	1.5–2.0	>96
AMX ^[8]	Yes	DVB/PS	$-\text{N}(\text{CH}_3)_3\text{Cl}$	160–180	1.4–1.7	25–30	2.0–3.5	98
CJMA-4 ^[15]	No	PVDF	$-\text{N}(\text{CH}_3)_3\text{Cl}$	90–100	0.5–0.6	15–20	3.5–4.5	>93
CJMA-6 ^[4]	Yes	Polyolefin	$-\text{N}(\text{CH}_3)_3\text{Cl}$	120–140	0.5–0.7	35–37	2.5–3.0	>98
CJMA-7 ^[16]	Yes	Polyolefin	$-\text{N}(\text{CH}_3)_3\text{Cl}$	140–160	0.9–1.0	28–30	1.5–1.8	95–98

[Note] ^a The data was collected from the product brochure provided by manufacturers.

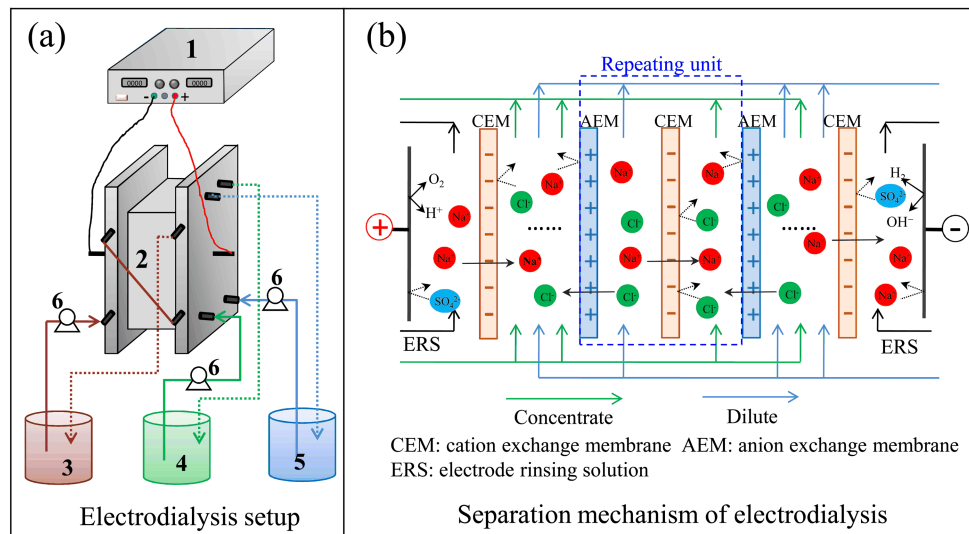


Figure 1. The experimental setup and separation mechanism of electrodialysis.

membranes (AEMs). Each membrane has an effective area of 189 cm^2 . The neighboring membranes were separated by a spacer with a thickness of 0.75 mm . Therefore, the membrane stack has five repeating units (five concentrated compartments and five dilute compartments).

During the ED test, the concentrated and dilute solutions were recirculated in the corresponding chambers at a flow rate of 567 mL/min (superficial velocity 3 cm/s). The constant current of 4.73 A (current density 25 mA/cm^2) was applied to the ED stack to concentrate NaCl solution. The conductivity of the dilute was monitored by a conductivity meter (DDS-307, Shanghai INESA & Scientific Instrument Co., Ltd., China). Each batch ED experiment was stopped once that the conductivity of the dilute solution decreased to 8 mS/cm . Then, the volume change of the dilute solution was measured to calculate water and salt

flux. Before each experiment, the ED stack was checked from leakage (external and internal) to make the experiment more accurate. Specifically, both concentrate and dilute chambers were recirculated with 200 mL 3% NaCl solution for 15 min . After that, the volume change of both chambers was checked. All the experiments were performed at the room temperature of $27.5 \text{ }^\circ\text{C} \pm 0.5 \text{ }^\circ\text{C}$.

2.3 Data analyses calculations

For the calculation of the salt content, a linear relation between the conductivity (κ , mS/cm) and salt content (δ , %) was regressed as Equations (1a) and (1b)^[4], while a linear relation between the salt content in % (δ) and in $\text{mol} \cdot \text{L}^{-1}$ (C) was regressed as Equations (2)^[4].

$$\delta = 0.000004\kappa^3 - 0.000212\kappa^2 + 0.063865\kappa - 0.034659, \\ R^2 = 0.999919, 0 < \kappa \leq 55 \text{ mS/cm} \quad (1a)$$

$$\delta = 0.000002\kappa^3 - 0.000224\kappa^2 + 0.086619\kappa - 0.870836, \\ R^2 = 0.999967, 55 < \kappa \leq 160 \text{ mS/cm} \quad (1b)$$

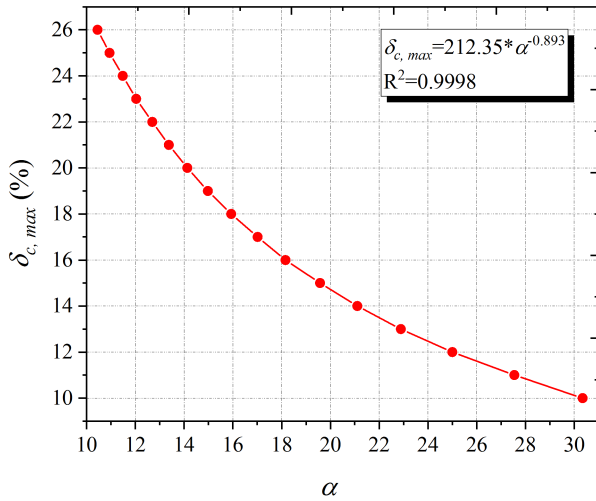


Figure 2. The relation between the flux ratio of water to salt (α) and the maximum salt content of the concentrated solution ($\delta_{c,\max}$).

$$C = 0.001387\delta^2 + 0.168291\delta + 0.005983, \\ R^2 = 0.999994, 0 < \delta \leq 26\% \quad (2)$$

The concentration of the concentrated solution can be calculated according to the material balance as^[4]:

$$C_{c,t} = \frac{C_{d,0}V_{d,0} - C_{d,t}V_{d,t} + C_{c,0}V_{c,0}}{V_{c,t}} = \frac{C_{d,0}V_{d,0} - C_{d,t}V_{d,t} + C_{c,0}V_{c,0}}{V_{c,0} + V_{d,0} - V_{d,t}} \quad (3)$$

where $C_{c,0}$ and $C_{c,t}$ are the NaCl concentrations of the concentrated solution at time 0 and t , respectively; $C_{d,0}$ and $C_{d,t}$ are the NaCl concentrations of the dilute solution at time 0 and t , respectively; $V_{c,0}$ and $V_{c,t}$ are the volumes of the concentrated solution at time 0 and t , respectively; and $V_{d,0}$ and $V_{d,t}$ are the volumes of the dilute solution at time 0 and t , respectively.

The salt flux through the cation and anion exchange membranes (J_{NaCl} , $\text{mmol}/(\text{m}^2 \cdot \text{s})$) is calculated by

the following equation^[4]:

$$J_{\text{NaCl}} = \frac{C_{c,t}V_{c,t} - C_{c,0}V_{c,0}}{2NA t} = \frac{C_{d,0}V_{d,0} - C_{d,t}V_{d,t}}{2NA t} \quad (4)$$

where N is the number of cell pairs ($N = 5$ in this study); A is the effective area of each membrane (189 cm^2); and t is the operating time.

The water flux ($J_{\text{H}_2\text{O}}$, $\text{mmol}/(\text{m}^2 \cdot \text{s})$) during the ED concentration process is calculated by the following equation^[4]:

$$J_{\text{H}_2\text{O}} = \frac{V_{c,t} - V_{c,0}}{36NA t} = \frac{V_{d,0} - V_{d,t}}{36NA t} \quad (5)$$

The flux ratio of water to salt (α) is calculated as^[4]:

$$\alpha = \frac{J_{\text{H}_2\text{O}}}{J_{\text{NaCl}}} \quad (6)$$

In fact, a given α is corresponding to a δ of the transported salt and water. We can consider this δ as the maximum salt content of the concentrated solution ($\delta_{c,\max}$) in the ED concentration process. Therefore, a linear relation between the α and the salt content ($\delta_{c,\max}$, %) was regressed:

$$\delta_{c,\max} = 212.35\alpha^{0.893}, R^2 = 0.9998 \quad (7)$$

In addition, the relation between α and $\delta_{c,\max}$ is clearly shown in Figure 2.

3 Results and discussion

Different CEMs and AEMs were matched and installed in the ED stack to concentrate high-salinity solutions at a current density of $25 \text{ mA}/\text{cm}^2$. After each batch experiment, the J_{NaCl} , $J_{\text{H}_2\text{O}}$ and α are calculated to evaluate the ED concentration performance of the matched membranes. The experimental data and the calculated J_{NaCl} , $J_{\text{H}_2\text{O}}$, α , δ_c (salt content of the ED concentrate), and $\delta_{c,\max}$ are listed in Table 2.

Table 2. The experimental data and the calculated J_{NaCl} , $J_{\text{H}_2\text{O}}$, α , δ , and δ_L at a different matching of the cation and anion exchange membranes.

CEM	AEM	Time (min)	$V_{d,0}(\text{L})$	$V_{d,t}(\text{L})$	J_{NaCl} ($\text{mmol}/(\text{m}^2 \cdot \text{s})$)	$J_{\text{H}_2\text{O}}$ ($\text{mmol}/(\text{m}^2 \cdot \text{s})$)	α	δ_c (%)	$\delta_{c,\max}$ (%)
CMX	AMX	65	2	1.80	1.24	15.07	12.11	13.64	22.90
CJMC-4	AMX	70	2	1.70	1.16	21.00	18.03	11.15	16.05
CJMC-5	AMX	67	2	1.70	1.22	21.94	18.03	11.15	16.05
CJMC-5S	AMX	78	2	1.58	1.05	26.38	25.02	9.23	11.98
CMX	CJMA-4	66	2	1.77	1.23	17.07	13.89	12.77	20.25
CMX	CJMA-6	65	2	1.76	1.25	18.09	14.49	12.51	19.51
CMX	CJMA-7	77.5	2	1.68	1.05	20.23	19.20	10.77	15.17
CJMC-5	CJMA-4	71	2	1.67	1.15	22.77	19.79	10.59	14.77
CJMC-5	CJMA-6	70	2	1.65	1.17	24.50	20.96	10.25	14.03
CJMC-5	CJMA-7	80	2	1.56	1.03	26.94	26.17	8.98	11.51

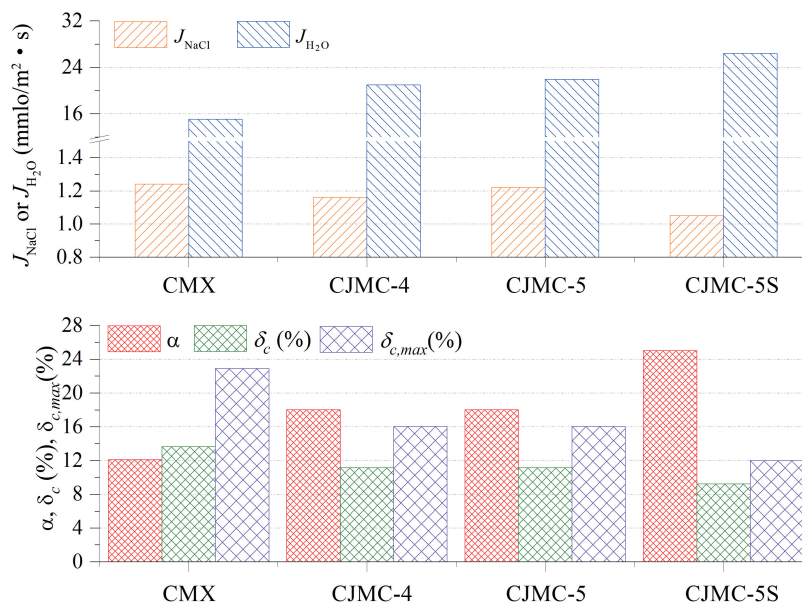


Figure 3. Effect of the cation exchange membrane on the concentration performance with AEM of AMX.

Firstly, four types of CEMs (i. e. CMX, CJMC-4, CJMC-5, and CJMC-5S) were matched with AMX to investigate the effect of CEM on the ED concentration performance. For Chemjoy CEMs, the CJMC-5 and CJMC-5S membranes have the same polymer matrix and functional groups but with different ion exchange capacity and water uptake, while CJMC-4 membrane has a different membrane polymer matrix compared with the CJMC-5 and CJMC-5S membrane as shown in Table 1. The ion exchange capacity and water uptake of the CJMC-5S membrane are higher than that of the CJMC-5 membrane. From Figure 3 we can see that a higher J_{NaCl} , a lower J_{H_2O} , a lower α and a high salt content of concentrate can be achieved for the CMX membrane compared with Chemjoy CEMs. When the Chemjoy CEMs were matched with the AMX membrane, ① the J_{NaCl} decreases slightly from 1.24 (CMX membrane used) to 1.16 and 1.22 mmol/(m² · s) for CJMC-4 and CJMC-5 membranes, respectively, and further decreases to 1.05 mmol/(m² · s) for the CJMC-5S membrane; ② the J_{H_2O} increases from 15.07 (CMX membrane used) to 21.00 and 21.94 mmol/(m² · s) for CJMC-4 and CJMC-5 membranes, respectively, and further increases to 26.38 mmol/(m² · s) for CJMC-5S membrane due to the higher water uptake of CJMC-5S membrane compared with that of CJMC-5 membrane, because the water transport is significantly influenced by the membrane water uptake^[11,13]; and ③ the α increases from 12.11 (CMX membrane used) to 18.03 for both CJMC-4 and CJMC-5 membranes, and further increases to 25.02 for CJMC-5S membrane due to the lowest J_{NaCl} and the highest J_{H_2O} as aforementioned. Besides, it should be noted that the concentration performance of

CJMC-5S is lower than that of CMX though both membranes have a similar ion exchange capacity, which means that the ion exchange capacity is less significant for the ED concentration performance when the composition of the membranes is different. The above results indicate that the water uptake of CEM is a key factor for the ED concentration performance when the ion exchange capacity of CEM has a feasible value such as in the range of 0.8–1.6 meq/g. This can be ascribed to the reason that the water transport including osmosis and electro-osmosis occurs when cations migrate from the dilute compartment to the concentrated one through CEMs^[9]. The CMX membrane is of a superior concentration performance (low α , 12.11; high δ_c , 13.64%; high $\delta_{c,max}$, 22.90%) due to the reason that this membrane has a high ion permeability, a dense structure (low water uptake, 25%–30%) and thus a low water permeability. If a CEM has no dense structure^[12] or has high water uptake^[9], the α will increase, resulting in a decrease in the ED concentration performance (low δ_c and $\delta_{c,max}$).

Secondly, four types of AEMs (i. e. AMX, CJMA-4, CJMA-6 and CJMA-7) were matched with CMX to investigate the effect of AEM on the ED concentration performance. For Chemjoy AEMs, the CJMA-4, CJMA-6 and CJMA-7 have different membrane structures, and thus different membrane properties such as the ion exchange capacity and water uptake as can be seen in Table 1. Specifically, the water uptakes of CJMA-4 and CJMA-6 membranes are higher than that of the standard AMX membrane, while the water uptake of CJMA-7 is the same as that of the AMX membrane. Figure 4 shows the ED concentration

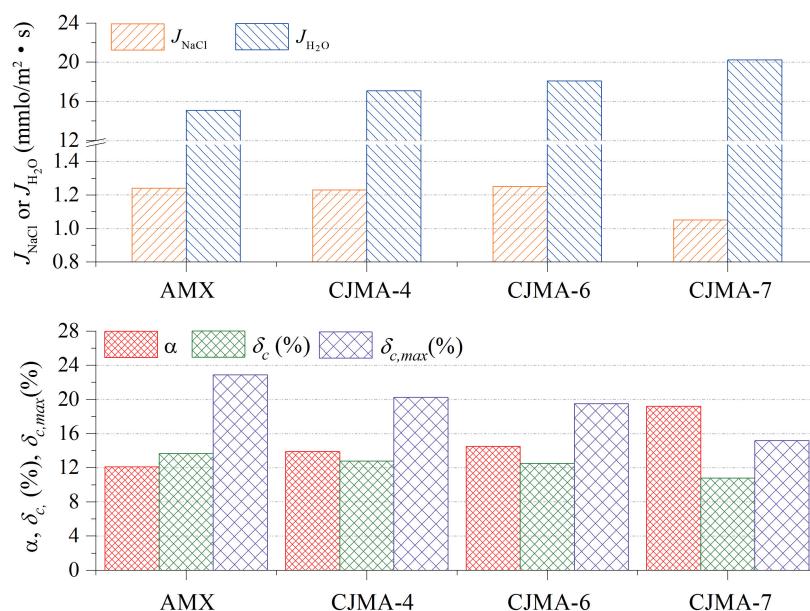


Figure 4. Effect of AEM on ED concentration performance with CEM of CMX.

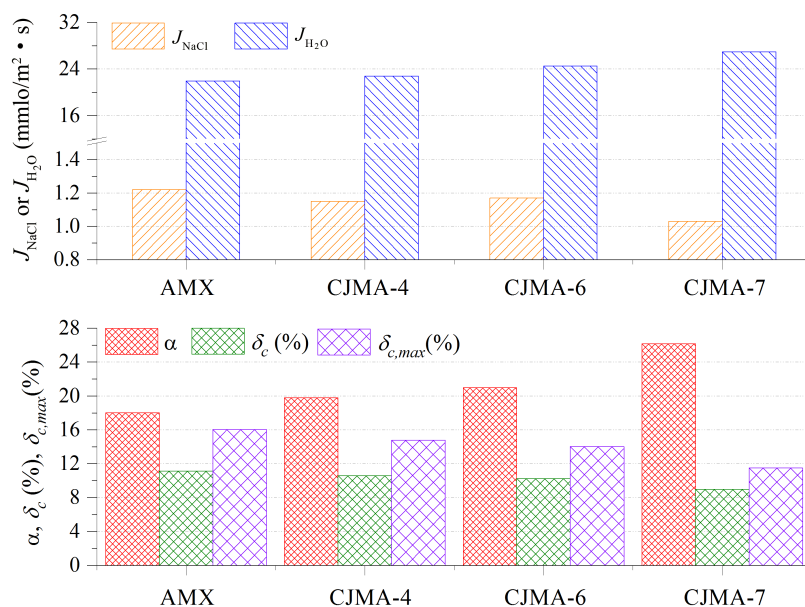


Figure 5. Effect of AEM on ED concentration performance with CEM of CJMC-5.

performance of different AEMs matched with CMX membranes. As compared with the standard AMX membrane, CJMA-4 and CJMA-6 membranes have a similar concentration performance. For instance, the values of J_{NaCl} of CJMA-4 (1.23 mmol/(m²·s)) and CJMA-6 (1.25 mmol/(m²·s)) membranes are nearly equal to that of AMX (1.24 mmol/(m²·s)); the values of $J_{\text{H}_2\text{O}}$ of CJMA-4 (17.07 mmol/(m²·s)) and CJMA-6 (18.09 mmol/(m²·s)) membranes are slightly higher than that of AMX (15.07 mmol/(m²·s)); the values of α of CJMA-4 (13.89) and CJMA-6 (14.49) membranes are slightly higher than that of

AMX (12.11); and the values of δ of the ED concentrate of CJMA-4 (12.77%) and CJMA-6 (12.51%) membranes are slightly lower than that of AMX (13.64%). Therefore, it can be concluded that there is no obvious influence on the ED concentration performance when using AEM with a relatively high water uptake. However, the ED concentration performance is relatively poor for the CJMA-7 membrane. The reason can be ascribed to the low ion permeability of the CJMA-7 membrane, because the J_{NaCl} is relatively low though the standard CMX membrane has a superior performance. Even so, CMX/CJMA-7 membranes have a better ED concentration

performance than CJMC-6/AMX membranes as can be seen in Figures 3 and 4. It indicates that AEM has a weaker influence on the ED concentration performance compared with CEM.

Additionally, AMX, CJMA-4, CJMA-6 and CJMA-7 membranes were matched with CJMC-5 membrane instead of the CMX membrane to consolidate the above statement that AEM has a weaker influence on the ED concentration performance. Figure 5 shows that the ED concentration performance has no obvious decrease when the standard AMX membrane was replaced by CJMA-4 and CJMA-6 membranes. But the ED concentration performance decreases a lot when the CJMA-7 membrane was matched. This phenomenon is the same as the above mentioned, because the CJMA-7 membrane has a low ion permeability.

Hence, we can conclude that the matching relation between CEM and AEM is significant for the ED concentration performance. For CEM, the water uptake is a key factor for the ED concentration performance when the ion exchange capacity of CEM has a feasible value; while for AEM, the ion permeability (or salt flux) maybe more important compared with the ion exchange capacity and water uptake, because the ED concentration performance is relatively poor when AEM with a low ion permeability was matched with the standard CMX that has a superior performance. Additionally, CEM is more significant for the ED concentration performance than AEM when both CEM and AEM have a relatively high ion permeability.

4 Conclusions

This study reports the significance of matching relation between CEM and AEM for the ED concentration performance. Results indicate that the water uptake of CEM is a key factor for the ED concentration performance when the ion exchange capacity of CEM has a feasible value; while for AEM, the ion permeability maybe more important compared with the ion exchange capacity and water uptake. Besides, CEM is more significant for the ED concentration performance than AEM when both membranes have a relatively high ion permeability. We hope the above conclusions can provide guidance for the membrane development in the ED concentration process.

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Conflict of interest

The authors declare no conflict of interest.

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电渗析浓缩高盐溶液: 阴阳离子交换膜匹配性关系

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摘要: 研究了阳离子交换膜和阴离子交换膜的匹配性对电渗析浓缩性能的影响, 主要考察了电渗析过程中的盐通量、水通量、水盐通量比和电渗析浓缩液的浓度. 实验结果表明, 当阳离子交换膜具有合适的离子交换容量时, 阳离子交换膜的水含量对电渗析的浓度性能有着重要的影响; 相比于离子交换容量和水含量, 阴离子交换膜的离子渗透性对电渗析浓缩性能有着更加重要的影响. 此外, 当阳离子交换膜和阴离子交换膜均具有较高离子渗透性时, 阳离子交换膜对电渗析浓缩的影响更为重要.

关键词: 电渗析; 阳离子交换膜; 阴离子交换膜; 匹配性关系; 浓缩性能