



Effects of operating parameters and ion characters on the adsorption capacity and energy consumption in membrane capacitive deionization

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ABSTRACT

Membrane capacitive deionization (MCDI) is an energy-efficient and environment-friendly technology for desalinating seawater or brackish water. In order to investigate effects of operating parameters and ion characteristics on the electrosorption capacity and energy consumption of MCDI, a series of experiments were conducted with single-electrolyte solutions (NaCl, KCl, MgCl₂ and CaCl₂) in this study. The results indicated that the working voltages, flow rates and initial concentration were the important factors for the performance of MCDI. Both electrosorption capacity and energy consumption were positive correlation to the three operating parameters, while specific energy consumption decreased with the increase of flow rate and initial concentration. Ions with smaller hydrated radius exhibited higher removal efficiency with lower energy consumption, and the difference of hydrated radius can be further enhanced when increase the working voltage. Besides, higher electrosorption capacity and lower energy consumption were observed when MCDI was operated with monovalent ion solution compared with using bivalent ion solution as inflow. This study clearly demonstrated the rule of electrosorption capacity and energy consumption in MCDI, hereby considerably providing a theoretical basis for practical application.

Keywords: Membrane capacitive deionization; Electrosorption capacity; Energy consumption; Operating parameters; Ion characters

1. Introduction

As the contradiction between shortage of freshwater resources and the increment of human demands is growing, purification of seawater or brackish water is one of the important challenges faced by human beings [1]. Prevailing desalination technologies such as reverse osmosis, electro-dialysis and thermal evaporation [2–4], which are applied to remove salt from seawater and brackish water, alleviate the pressure of freshwater supply but at the cost of high energy-consumption [5–7].

By contrast, membrane capacitive deionization (MCDI), also known as electrosorption or electrochemical desalination technology, is considered as a promising desalination

technology with the characteristics of energy-efficient and environment-friendly technology [8–10]. During the desalination, feedwater flows through the space between two electrodes where ion-exchange membranes are placed in front. The ions migrate towards the oppositely charged electrodes and are temporarily stored in the electrical double layers (EDLs), with a reduction of effluent conductivity to a certain level. Once the electrodes are saturated with ions, reverse voltage is applied to make the ions release to the spacer, which is designated as regeneration stage [11–13].

Compared with the conventional capacitive deionization (CDI), MCDI demonstrates great improvement in desalination efficiency by introducing ion-exchange membranes, resulting from the selective absorption of ions onto the electrode surface [14–17]. Besides higher ion removal rate and weaker coion repulsive effect in absorption stage, MCDI also

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exhibits promoted efficiency of electrodes regeneration by avoiding the reabsorption of desorbed ions in desorption stage [9,18,19].

In the previous literatures, studies about MCDI technology were mainly focused on the fabrication of electrode materials [15,20–22] or effect evaluation of operating parameters on the performance [18,23]. Recently, there are increased interests in studying energy-consumption of MCDI operation [24]. Zhao et al. [25] found that regardless the applied voltage and influent (feedwater) concentrations, MCDI was always superior to the CDI in aspect of specific energy consumptions (SECs). Zhao et al. [26] showed that MCDI required more energy with the rise of the water recovery or water flow rate when the removal efficiency was fixed. Choi [27] compared the energy consumption of two operation modes, constant current mode (CC mode) and constant voltage mode (CV mode) for sodium chloride solution purification, and observed higher adsorption capacity in CV mode and lower energy consumption in CC mode. Hou and Huang [28] found that the MCDI electrosorption performance was highly dependent on the charge, hydrated radius and initial concentration because of the double-layer overlapping. However, to our best knowledge, the influences of operation parameters and ion characteristics (such as the ion charge and hydrated radius) on the energy consumption in MCDI have not been investigated in detail.

The objective of this study was to identify the effect of operation parameters and ion characteristics on electrosorption capacity and energy consumption of MCDI. A systematic investigation was carried out to deionize the solution of cations with different ionic radius and charges. The cations selected for evaluation included sodium (Na^+), potassium (K^+), calcium (Ca^{2+}) and magnesium (Mg^{2+}). The influences of parameters (voltage, flow rate and initial concentration) and ion characteristics (ionic valence and hydrated radius) on electrosorption and energy consumption were investigated, which was expected to provide more reliable theoretical basis for MCDI operation.

2. Materials and methods

2.1. Description of MCDI stack and desalination system

Fig. 1 shows the scheme of MCDI stack and the flow chart of MCDI setup. The MCDI stack contained six repeating cells that were sandwiched between two end plates

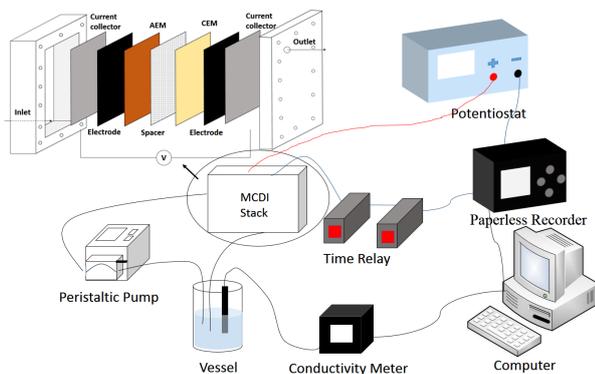


Fig. 1. The flow chart of MCDI setup and scheme of MCDI stack.

made of Plexiglass. In the MCDI cell, two parallel carbon sheets (Crystal Dragon Carbon Technology Co., Ltd., thickness = 1 mm, 20 cm × 10 cm) were set as the current collectors, in front of which activated carbon fiber felts (Jiangsu Kejing Carbon Fiber Co., Ltd., specific surface area = 1,200 m²/g, thickness = 1 mm) were attached closely to the anode and cathode. Cation-exchange membrane (AGC Asahi Glass Co., Ltd., thickness = 0.26 ± 0.02 mm) was placed in front of the cathode, and the anion-exchange membrane was correspondingly attached in front of the anode. The feedwater, which was pumped from the lower inlet of the end plate, flowed through the woven spacer (thickness = 1 mm) between two ion-exchange membranes. Then, the dilute solution released through the upper outlet of the opposite plate.

Operational parameters were investigated through batch experiments which were conducted in NaCl solution at different voltages, flow rates and initial concentrations. Ion characteristics were examined with a series of single-electrolyte solutions (including NaCl, KCl, CaCl_2 and MgCl_2) with the same initial molar concentration (10 mM), then the salt adsorption capacity and energy consumption are compared. The influent was continuously fed to the MCDI stack from a 500 mL beaker at a flow rate of 40 mL/min using peristaltic pump (YZ II 15, Longer Precision Pump Co., Ltd., China), and the effluent flew back to the beaker as recycle. The conductivity was analyzed by a conductivity meter (EC-1800, Jishen Technology Co., Ltd., China) and the data were logged into the computer at 5 s interval. When the feedwater flowed into the MCDI setup, a voltage was applied to the electrodes by the potentiostat (IT6720, ITECH Co., Ltd.). The adsorption and desorption time were controlled by the time relay (DH48S, Qinyang Electrical Co., Ltd., China). The interelectrode voltage and electrical current were measured automatically using a paperless recorder (THTZ408R, Penghe Electronics Co., Ltd., China) at 5 s interval.

2.2. Calculation of salt adsorption capacity and energy consumption

The salt electrosorption capacity was the amount of salt removed during the charging stage, which was calculated using the following equation [24]:

$$\text{The electrosorption capacity (mmol/g)} = \frac{(C_0 - C_e) \times V}{6m_e} \quad (1)$$

where C_0 and C_e are the initial and equilibrium concentrations (mM), respectively, m_e is the mass of each cathode ($m_e = 1.2$ g and the number = 6) and V is the volume of desalted water (about 0.4 L).

SEC was defined as the ratio of the applied electrical energy to the amount of removed ion. The amount of applied electrical energy was calculated by integrating current over charging time, and multiplying by the voltage using the below equation [24]:

$$\text{SEC (W} \cdot \text{h/mmol)} = \frac{v_e \times \int_0^t Idt}{(C_0 - C_e) \times V} \quad (2)$$

where v_e is the applied working voltage (V), I is the current (A) in the MCDI and t is the desalination time (h).

3. Results and discussion

3.1. Analysis of periodic experiment

Fig. 2 shows variations of conductivity, current and voltage for MCDI stack operated with CV mode. According to Fig. 2(a), during one operational cycle, a CV (1.2 V) was applied to the MCDI stack for 55 min as the adsorption process, providing under the electrostatic force whereby the ions were adsorbed to the electrode. Then in desorption process, -1.2 V voltage was applied for 40 min allowing ions to be released to the spacer under reserved electrostatic force. The corresponding variations of conductivity and current profiles are presented in Figs. 2(b) and (c).

According to Fig. 2(b), the conductivity dropped from 1,160 to 457 $\mu\text{S}/\text{cm}$, and increased to the initial value during the desorption process. The decline rate of conductivity slowed down over the adsorption time because of the gradual saturation of electrodes. There was a similar phenomenon during desorption process, where the conductivity rose sharply before it gradually approached the initial value with the similar increasing gradient. Noteworthy shorter time required for desorption process compared with adsorption

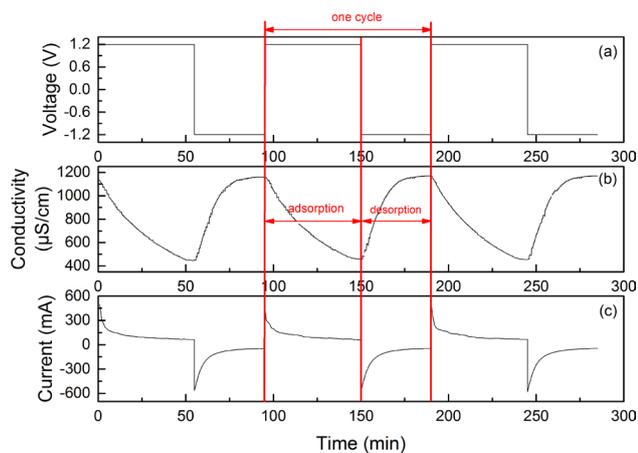


Fig. 2. Experimental results of (a) voltage, (b) conductivity and (c) current as a function of time at 1.2 V, feed concentration of 10 mM and flow rate of 40 mL/min.

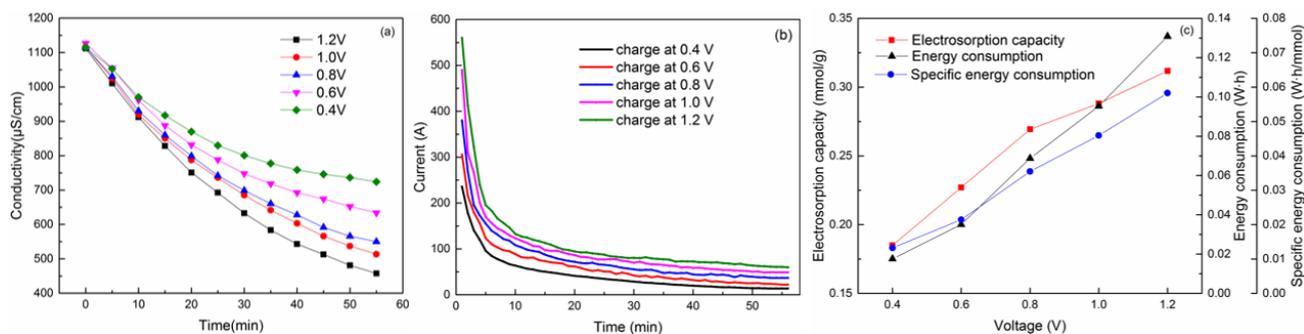


Fig. 3. Variations of (a) conductivity, (b) current and (c) electroadsorption capacity, energy consumption and specific energy consumption at different working voltages in the adsorption process.

process suggested the efficiency which would facilitate the practical MCDI application.

Fig. 2(c) presents the current variation in the working time. It can be observed that the electric current rapidly dropped from 540 to 150 mA during the initial 5 min and slowly decreased to 60 mA at 50 min. The initial quick decrease of current during adsorption process might be the result of ion migration from the solution in spacer compartment onto the electrodes, and its stable phenomenon after 50 min indicated the saturation of the electrode adsorption. Additionally, the electric current profile in desorption process was similar to that in adsorption process, but opposite in direction.

3.2. Effects of operating parameters upon performance of MCDI

3.2.1. Effects of voltages

To investigate the influence of voltage to the electroadsorption capacity and energy consumption of MCDI, the experiments at different voltages were conducted with NaCl solution of 10 mM (around 1,100 $\mu\text{S}/\text{cm}$) and flow rate of 40 mL/min. Fig. 3(a) shows the concentration variation of NaCl solution with operating time at voltages ranging from 0.4 to 1.2 V. As the applied voltage increased to 0.4, 0.6, 0.8, 1.0 and 1.2 V, the ions were adsorbed onto the surface of the oppositely charged electrodes under different electric field force, resulting the decrement of conductivity ending at 724.26, 633.72, 550.08, 513.67 and 457.58 $\mu\text{S}/\text{cm}$, respectively. The result indicated a negative correlation between conductivity and the working voltage.

The electroadsorption capacities, energy consumption and SEC at different voltages are shown in Fig. 3(c). The results illustrated that the electroadsorption capacity of activated carbon fiber electrodes ascended with working voltages because of the strengthening electrostatic interaction. Energy consumption also increased with the rising working voltage and reached 0.13 W h at 1.2 V, about seven times of that at 0.4 V.

Further profile for the effect of working voltage was illustrated through SEC, which is the energy consumption for per unit of electroadsorption capacity. It can be seen that the SEC presented an upward trend as the voltage rising from 0.4 to 1.2 V, suggesting that the energy efficiency declined with an increment of working voltage. When higher working voltage was applied, higher electric current occurred in the MCDI system (Fig. 3(b)), leading to more energy consumption

caused by the system resistance. Besides, more ions were adsorbed in the electrodes at higher voltage, which increased the repulsion between ions, making it more difficult for ions in spacer to migrate to electrodes.

3.2.2. Effects of flow rates

The adsorption capacity, energy consumption and SEC of MCDI at different flow rates (10–60 mL/min) are shown in Fig. 4, in which all initial NaCl concentrations are 10 mM. As can be seen, the electrosorption capacities of MCDI grew with the increase of flow rates and reached 0.34 mmol/g at 60 mL/min. A relatively low flow rate allowed limited amount of feed passed through the spacer during a certain period of operation and resulted in the low MCDI desalination efficiency. As feeding water was circulated, higher flow rate meant more chances for ions to be exposed under electrostatic field. Additionally, there was a slight rise of energy consumption in response to the increase of flow rates.

The SEC decreased from 0.073 to 0.059 W h/mmol, when the flow rates increased from 10 to 60 mL/min. The results indicated that the MCDI was more energy efficient when

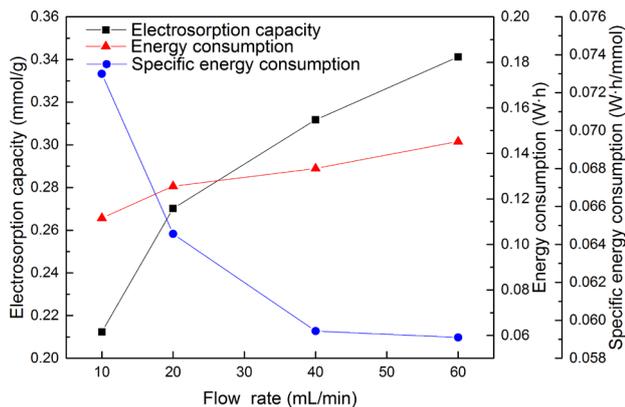


Fig. 4. Electrosorption capacity and energy consumption as a function of flow rates.

operated at higher flow rates. Although more energy was consumed at high flow rate, the manifest increase of adsorption capacity would outdo this drawback leading to the superiority as lower SEC of system. However, Zhao et al. [29] presented the opposite conclusion that less energy was consumed at a lower water flow rate when removing a given amount of salt ions. The contradictory results might be attributed to the different operation modes, in which the amount of removed ions was fixed and the time consumption was ignored [26] but the operating time was kept fixed in our experiments. Besides, the different phenomenon may also be caused by the difference between two MCDI modules.

3.2.3. Effect of initial concentration

The initial concentration was another factor affecting the adsorption capacity and energy consumption, and the desalination experiment was conducted at 1.2 V with initial concentrations ranging from 5 to 20 mM. As can be seen in Fig. 5(a), adsorption capacity was 0.16 mmol/g at the initial concentration of 5 mM and continued rising until it almost tripled when initial concentration was 20 mM. The result indicated that adsorption capacity grew with the increase of initial concentration. According to the theory of Gouy–Chapman–Stern, the amount of ion storage in the EDL increased with the increase of concentration of feedwater, but no further increase would occur when the concentration reached to an extreme value [30]. Conversely, the removal efficiency presented a downward trend when increasing the initial concentration as the consequence of saturated space of electrodes to store the adsorbed ions.

In terms of energy consumption and SEC, Fig. 5(b) illustrates an increasing trend of energy consumption with the increment of initial concentration, while the SEC presented oppositely. As the current through the MCDI was subject to the intension of ion transference between the two parallel electrodes, higher concentration thus allowed greater current and led to a high energy consumption. In spite of this upward trend of energy consumption, SEC decreased because of the more manifest rise of adsorption, suggesting

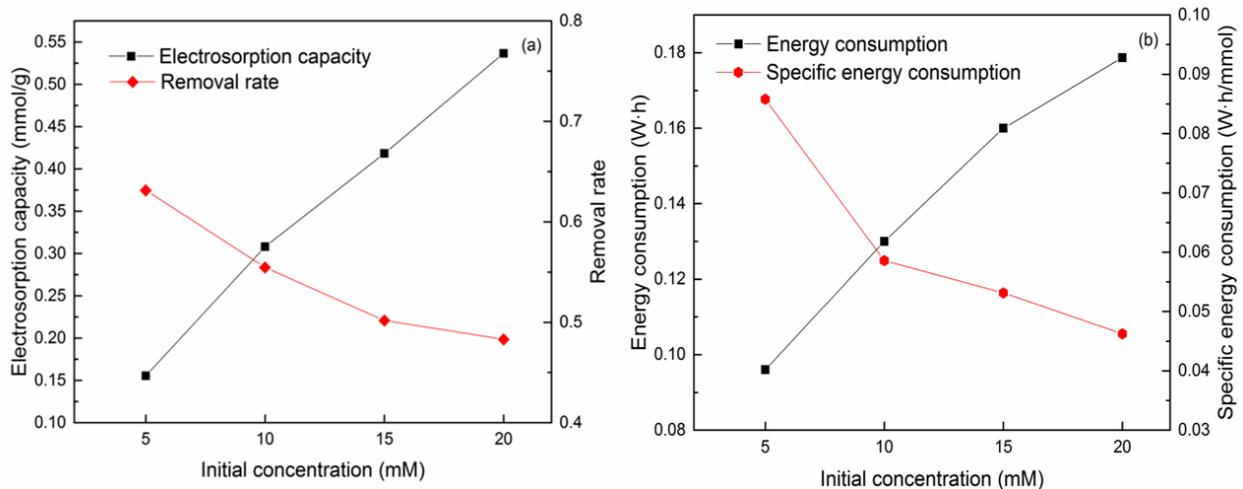


Fig. 5. The variation of (a) electrosorption capacity and removal rate and (b) energy consumption and SEC as a function of initial concentration.

MCDI to operate with concentrated solution as an efficient strategy.

3.3. Effect of ion characters on the performance of MCDI

3.3.1. Electrosorption capacity

To demonstrate the effects of ion properties on the salt adsorption capacity and energy consumption, monovalent Na^+ , K^+ and divalent Ca^{2+} , Mg^{2+} were adopted in this section. The ionic radius of cations were in the order of K^+ (0.133 nm) > Ca^{2+} (0.099 nm) > Na^+ (0.095 nm) > Mg^{2+} (0.065 nm), and the order of hydrated radius was K^+ (0.331 nm) < Na^+ (0.358 nm) < Ca^{2+} (0.412 nm) < Mg^{2+} (0.428 nm) [31]. All electrolyte solution had a common anion Cl^- .

The experiment was conducted with the same initial concentration (10 mM), and Fig. 6 shows the electrosorption capacities of different ions under working voltages ranging from 0.4 to 1.2 V. The electrosorption capacity of Na^+ and K^+ both presented a continuous increase as the working voltage rising in Fig. 6(a). The adsorption capacity of K^+ was less than 0.20 mmol/g at 0.4 V then increased to 0.33 mmol/g when the applied voltage rose to 1.2 V, closely followed by Na^+ which also presented an upward trend with the rising working voltage. Also, the difference between K^+ and Na^+ gradually manifested as the working voltage increasing. This was due to that ions with shorter hydrated radius occupied less space thus can screen the electrode surface more efficiently, and this phenomenon would be more obvious under higher voltage. Hou and Huang [28] revealed that ions with larger hydrated radius can cause double-layer overlapping easily for the size-exclusion and confinement effect of charged nanopores, which reduced the storage of ions with larger hydrated radius.

Similar trends of electrosorption capacities for Ca^{2+} and Mg^{2+} were observed (Fig. 6(b)). Electrosorption capacity of Ca^{2+} was higher than that of Mg^{2+} at voltage of 0.4 and 0.6 V, because of the smaller hydrated radius of Ca^{2+} which made it easier to be adsorbed onto the electrode. However, electrosorption capacity of Mg^{2+} began to be approaching that of

Ca^{2+} when higher voltages was applied (>0.6 V) and finally exceed Ca^{2+} at 1.2 V, possibly because of the dehydration reaction (the inverse process of hydration) occurring during the adsorption. Moreover, with the increment of working voltage, dehydration actions were more likely to occur within the electrode pores and the Mg^{2+} with smaller ionic radius presented higher amount of electrosorption capacity. By comparing the electrosorption capacities for monovalent and divalent cations, it was noticeable that the electrosorption capacities of MCDI were higher when treating monovalent cation solution. Ji et al. [32] also reported that the removal rate was best for Na^+ and followed by Ca^{2+} , which showed the same trend as our results.

3.3.2. Energy consumption

The comparison of energy consumption for NaCl , KCl , MgCl_2 and CaCl_2 solution is shown in Fig. 7, in which five

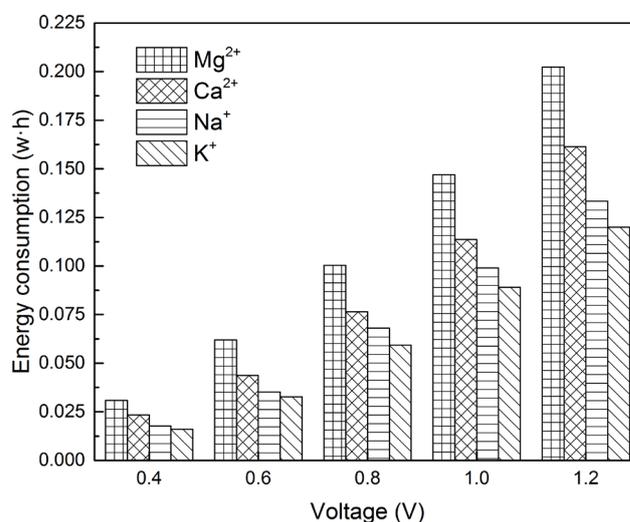


Fig. 7. Energy consumption of MCDI for monovalent and divalent cations at different working voltages.

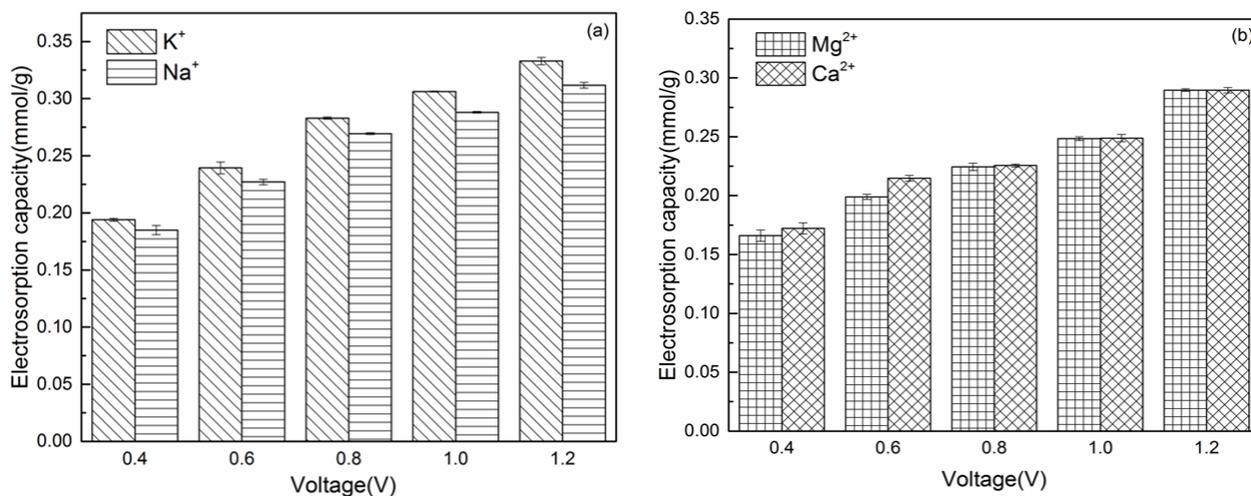


Fig. 6. Electrosorption capacities of (a) monovalent (Na^+ and K^+) and (b) divalent (Ca^{2+} and Mg^{2+}) cations at different working voltages ranging from 0.4 to 1.2 V.

levels of voltages (0.4, 0.6, 0.8, 1.0 and 1.2 V) are investigated. It can be seen that for both Na^+ and K^+ solutions, the energy consumption of MCDI increased as the working voltage rises. At 0.4 V, the energy consumption for Na^+ and K^+ was almost the same, which were 0.018 and 0.016 W·h, respectively. When the voltage increased to 1.2 V, the energy consumption for Na^+ increased to 0.130 W·h, approximately 0.010 W·h higher than that for K^+ . It was because that there was more resistance existing when Na^+ with the larger hydrated radius transferred to the inner of electrode under the electrostatic force. Besides, as the working voltage increased, the difference of energy consumption between Na^+ and K^+ increased as well, suggesting that the effects of hydrated radius on energy consumption were also subject to the applied voltages.

As observed, energy consumption for Ca^{2+} and Mg^{2+} at 0.4 V was 0.023 and 0.030 W·h, respectively, both of which rose with the increase of working voltages as well. Within the studied voltage range (0–1.2 V), the energy consumption of Mg^{2+} was always higher than that of Ca^{2+} , and the difference was greater at higher voltage. The maximum energy consumption for Mg^{2+} was about 0.21 W·h at 1.2 V, which was approximately 25% higher than that of Ca^{2+} . As discussed earlier in monovalent ions, the ion with larger hydrated radius consumed more energy in MCDI operation system due to more resistance during the migration process. Therefore, the energy consumption of Ca^{2+} here was less than that of Mg^{2+} .

The SEC results for Na^+ , K^+ , Ca^{2+} and Mg^{2+} at different working voltages are presented in Fig. 8. The SEC of both monovalent and divalent cations grew with voltages from 0.4 to 1.2 V, while for the divalent cations, the SEC increased more rapidly. Moreover, the SEC for the four studied cations was in the order $\text{Mg}^{2+} > \text{Ca}^{2+} > \text{Na}^+ > \text{K}^+$. This means that the SEC was related to the ionic charge of solution, and the SEC was higher in MCDI when treating the solution containing cations with higher ionic charge. The reason was that more repulsion occurred between divalent ions compared with monovalent ions, resulting in lower energy efficiency when treating divalent ions.

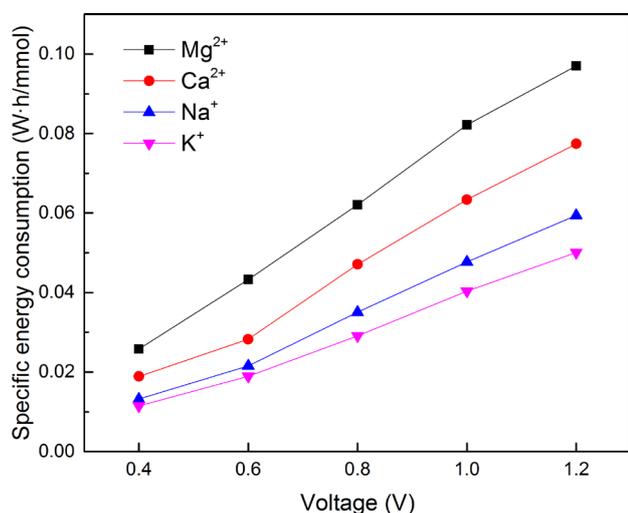


Fig. 8. Comparison of specific energy consumption for monovalent and divalent cations at different applied voltages.

4. Conclusion

In this study, the electrosorption experiment was conducted with single-electrolyte solution to investigate the effects of operating parameters and ion characteristics on performance of MCDI. The obtained results revealed that the electrosorption capacity grew with the increase of applied voltages due to stronger electric field force, while the energy consumption was even higher at high voltage as a result of more energy loss. The electrosorption capacity and energy consumption rose as the flow rates increasing, while the SEC presented an opposite trend. Furthermore, ions with smaller hydrated radius presented larger electrosorption capacity and lower energy consumption, and the difference manifested at higher working voltage for both monovalent and divalent cations. Specially, the electrosorption capacity of Mg^{2+} was slightly higher than that of Ca^{2+} at 1.2 V, while the hydrated radius of Mg^{2+} was larger. More electrosorption capacity and less energy consumption of MCDI were observed when treating the monovalent ions compared with divalent ions.

Acknowledgments

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