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Comparison of constant voltage (CV) and constant current (CC) operation in the membrane capacitive deionisation process

Jae-Hwan Choi

Department of Chemical Engineering, Kongju National University, 1223-24 Cheonan-daero, Seobuk-gu, Cheonan, Chungnam 331-717, South Korea, Tel. +82 41 521 9362; Fax: +82 41 521 2640; email: jhchoi@kongju.ac.kr

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ABSTRACT

In this study, desalination characteristics were systematically compared and analysed during operation of the membrane capacitive deionisation (MCDI) process in constant current (CC) and constant voltage (CV) modes. Operational characteristics in the MCDI process such as the change in effluent concentrations, the number of adsorbed ions, charge efficiencies and energy consumption were compared based on the power supply mode. When the cell potential was not varied, more ions were adsorbed in CV mode than in CC mode. However, it was determined that desorption proceeded rapidly when performing adsorption in CC mode. Regarding the charge efficiency, the process in CV mode exhibited a higher efficiency than in CC mode. In particular, the charge efficiency decreased beyond a cell potential of 0.8 V due to electrode reactions. In contrast, the energy consumption was greatly reduced when operating the MCDI process in CC mode compared with when operating the MCDI process in CV mode.

Keywords: Membrane capacitive deionisation; Constant voltage; Constant current; Charge efficiency; Energy consumption

1. Introduction

As the seriousness of energy and environmental issues increases worldwide, there has been growing interest in capacitive deionisation (CDI) technology [1–4]. CDI is based on the principle that ions around electrodes are adsorbed on the electrode surface due to electrostatic attraction when charging electrodes using an external power source. In comparison with conventional desalination technologies such as reverse osmosis, electrodialysis and ion-exchange methods, CDI has the advantages of lower energy consumption and environmental friendliness [5–8]. Accordingly, CDI has been evaluated as a promising desalination technology for the future.

Since the concept of CDI technology was introduced by Caudle et al. [9] in the mid-1960s, many studies have been performed in a variety of research areas. CDI technology utilises the adsorption reaction at the electrode surface. As a result, most studies have been carried out in the field of electrode manufacturing technology, and remarkable achievements have been reported. With the development of material technologies, various carbon materials with high specific surface areas and excellent electrical conductivities have been produced. Using carbon materials such as activated carbon powder, carbon nanotubes, carbon fibres, carbon aerogels and graphene, highly efficient carbon electrodes with enhanced adsorption capacities have been developed [10–14].

The technology of membrane capacitive deionisation (MCDI), which involves placing ion-exchange membranes in front of each electrode, has been introduced to improve the desalination efficiency in conventional CDI [15–19]. It was possible to dramatically improve the desalination efficiency of existing CDI using MCDI technology. Further, to enable MCDI, a composite carbon electrode has been developed, where ion-exchange polymers are coated onto the surface of the carbon electrode [20]. Recently, a selective electrode, where materials highly selective for specific ions are coated on the surface of the carbon electrode, has been developed and has further improved the desalination efficiency of the MCDI process [21].

To enable the widespread use of the MCDI process in the field of desalination, the development of carbon electrodes is important, but the technology to effectively operate the MCDI system is also very important. Because most studies to date have been conducted with a focus on the development of the carbon electrode, research on operating skills has remained relatively lacking. Depending on how one operates the MCDI process, the efficiency of the process may vary significantly. Therefore, the study of the desalination performance based on the operating mode of the MCDI system has significant importance.

MCDI adsorbs ions by charging the electrodes via an external power supply unit. Therefore, the power supply method is a very important factor that determines the desalination characteristics of the MCDI operation. For the power supply, a method of controlling the voltage or the current may be applied. One method of operating the MCDI process involves applying a constant voltage (CV) to the cell. When operating in this mode, the concentration of the effluent tends to vary in time based on the change in the amount of adsorption with time. It is also possible to operate the MCDI process by supplying a constant current (CC) to the cell. In general, the current flowing through the cell is proportional to the adsorbed amount. Therefore, it is possible to maintain a constant salt concentration in the effluent when operating in CC mode [22,23].

To date, the method of applying a CV to the cell has been applied in the operation of MCDI in most cases. However, Zhao et al. [22] have recently reported the results of a desalination experiment performed by supplying a CC to an MCDI cell. They could adjust the concentration of the effluent to the desired level by adjusting the flow rate and the current supplied to the cell. In addition, they compared the energy consumption based on the salt concentration of the influent in the CV and CC modes.

The power supply method is a very important factor in the application and practical use of the MCDI process. However, research regarding the power supply method is insufficient. The operational characteristics in accordance with the CC and CV modes for the operation of the MCDI process were systematically analysed in this study. After operating the MCDI in the CV and CC modes while varying the voltage applied to the electrodes, the changes in the effluent concentration and the current and voltage flowing through the cell as a function of the each power supply method used were analysed. Further, the number of adsorbed ions, charge efficiency and energy consumption were compared between the CV and CC modes.

2. Experiment

2.1. Fabrication of carbon electrode

A carbon electrode was fabricated by mixing activated carbon powder with a polymer binder. After dissolving poly (vinylidene fluoride) (PVdF, M.W. = 530,000, Aldrich) in di-methylacetamide (DMAc, Aldrich), an organic solvent, and mixing it with activated carbon powder (CEP-21 K, PCT Co., Korea, BET surface area = 1,320 m^2/g), it was stirred in a planetary centrifugal mixer (AR-100, THINKY Co., Japan) for 30 min to produce a homogeneous electrode slurry. After placing the electrode slurry on a graphite foil (F02511, Dongbang Carbon Co., Korea), a current collector, and casting it to a uniform thickness using a doctor blade, it was dried in a drying oven at 50˚C for 2 h. To remove the remaining DMAc in the carbon electrode, it was dried for an additional 2 h in a vacuum oven at 50˚C. The mass ratio of PVdF, a polymer binder, was 10wt% in the produced carbon electrode. The thickness of the activated carbon layer was approximately 150 μm and the mass of the carbon powder was 0.91 mg/cm^2.

2.2. Construction of an MCDI unit cell

An MCDI unit cell to be used in the desalination experiments was prepared using the fabricated carbon electrode. The carbon electrodes were cut to a size of 10 × 10 cm, and they were used as the anode and the cathode. The total mass of the activated carbon in both of the electrodes was 1.82 g. An anion-exchange membrane (Neosepta AMX, Astom Co., Japan) was placed in front of the carbon anode electrode, and a cation-exchange membrane (Neosepta CMX, Astom Co., Japan) was placed in front of the carbon cathode electrode. Next, a 120-μm thick spacer (EX31-071/80 PW, NBC Meshtec Inc., Japan) was sandwiched between ion-exchange membranes for the influent to flow through. A hole with a 1-cm diameter was made in the middle of the carbon electrode so that the influent would flow in from the edge of the carbon electrode and flow out to the centre of the electrode through the spacer. A unit cell was produced by applying Plexiglas plates on both sides of the carbon electrode and subsequently bolting them together. The configuration diagram of the MCDI unit cell was described in detail in previous papers [20,24].

2.3. Desalination experiments using the MCDI unit cell

The desalination experiments were conducted using the fabricated MCDI unit cell while varying the power supply mode. The influent (10 mM NaCl) was continuously fed to the MCDI unit cell at a flow rate of 20 mL/min using a metering pump. Electric power was applied to the unit cell through a potentiostat (WPG100, WonA Tech Co., Korea). While supplying the voltage (or current) that was set in advance, adsorption and desorption experiments were conducted, and the voltage and current supplied to the cell were automatically measured using a computer at 3 s intervals.

In this study, the desalination characteristics measured while performing the adsorption process in CV and CC modes were compared. Therefore, desorption after adsorption was carried out under the same conditions (cell potential of 0.0 V for 300 s). First, to analyse the desalination characteristics in CV mode, a constant cell potential was supplied during the adsorption process. After adsorption occurred for 300 s at a constant cell potential, the cell potential was immediately changed to 0.0 V, and desorption occurred for 300 s. This adsorption/desorption cycle was repeated for five cycles under the same cell potential to reach a dynamic steady state in which the measured adsorption during one phase of the cycle was close to the measured salt desorption in the other phase of the cycle [22]. After the third cycle, we observed that operational characteristics such as the effluent concentration, current and the cell potential obtained a quasi-steady state and would repeat almost unchanged for a prolonged period. To analyse the effect of the cell potential, the desalination experiments were repeated by incrementing the cell potential by 0.2 V through the range of 0.4–1.4 V.

To study the desalination characteristics in CC mode, an adsorption experiment was conducted while supplying a constant current of 10 A/m^2 to the cell. To compare it with the results of the desalination process in CV mode, an adsorption experiment was conducted until the cell potential reached the cell potential applied in CV mode (0.4–1.4 V). As soon as the cell potential reached the set value, the cell potential was changed to 0.0 V and the adsorbed ions were desorbed for 300 s. As in the case of the experiment in CV mode, the adsorption and desorption processes were repeated for five cycles under the same conditions to reach a dynamic steady state.

An electrical conductivity sensor (CON-BTA, Vernier Software & Technology, USA) and a pH sensor (pH-BTA, Vernier Software & Technology, USA) were installed at the point where the effluent leaves the cell. The electrical conductivity and pH were automatically measured every 3 s using the interface (LABQUEST, Vernier Software & Technology, USA) connected to the computer. The concentration of the effluent was calculated from the correlation between the electrical conductivity and the NaCl concentration.

3. Results and discussion

3.1. Desalination characteristics in CV mode

Adsorption was performed by keeping the cell voltage constant for 300 s, and subsequently, desorption was performed by changing the cell voltage to 0.0 V. Fig. 1 shows the change in the effluent concentration as the cell potential is varied in the adsorption process. It can be observed that the concentration of the effluent rapidly decreases as soon as the cell potential is applied, and the ion-adsorption reaction proceeds very rapidly. Further, the concentration of the effluent in all cell potentials has slowly increased after going through the minimum value and

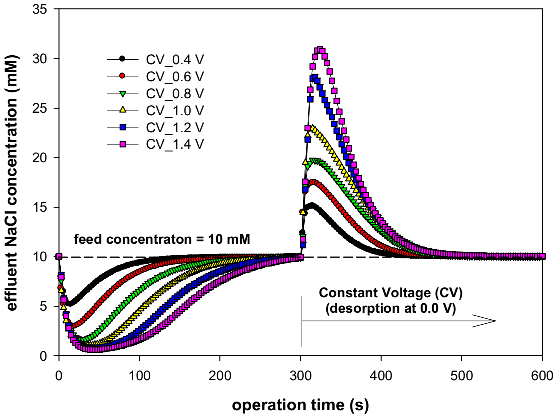

Fig. 1. Effluent NaCl concentrations at various cell voltages during the adsorption process. Desorption occurs at 0.0 V for 300 s. The feed solution (10 mM NaCl) was fed at a flow rate of 20 mL/min.

converges to the initial feed concentration (10 mM) as the adsorption capacity of the carbon electrode reaches saturation. As the cell potential increases, the adsorption rate of the ions increases and the minimum value of the effluent is reduced. When the cell potential is 0.4 V, the minimum concentration of the effluent is 5.2 mM; however, the minimum concentration of the effluent is reduced to 0.63 mM at 1.4 V.

The change in the effluent concentration when performing desorption at 0.0 V after the adsorption process was completed at each cell potential is shown in Fig. 1. During the adsorption process, the higher the cell potential was, the higher the adsorption rate was. Accordingly, the concentration of the effluent was significantly increased in the desorption process. When the cell potential in the adsorption process was 1.4 V, the concentration of the effluent in the desorption process was increased by up to 30.9 mM. A noteworthy point in the adsorption and desorption processes is that the desorption process proceeds more rapidly than the adsorption process. In the adsorption process, the time necessary for the adsorption capacity of the carbon electrode to reach saturation (the time necessary until the influent concentration becomes equal to the effluent concentration) varies depending on the applied potential (100–300 s). In contrast, in the desorption process, it can be observed that although there are some differences depending on the amount adsorbed to the electrode, most ions are desorbed after approximately 100–150 s.

Fig. 2 shows the variations in the current density supplied to the cell when the adsorption (1.0 V) and desorption (0.0 V) experiments were conducted in CV mode. For the first 40 s after applying the cell potential, the current density was slightly decreased from $31.1 \mathrm{A/m^2}$ to $29.1 \mathrm{A/m^2}$, after which it rapidly decreased. After 200 s, the current density was reduced to $2.7 \mathrm{A/m^2}$. The current supplied to the cell in the MCDI process is proportional to the number of ions adsorbed. It can be confirmed that the change in the current density is in good agreement with the results of Fig. 1, which shows the change in the effluent concentration.

In contrast, when the cell potential was changed to 0.0 V for the desorption process, the current density exponentially decreased over time. Although the initial current density was $59.9 \mathrm{A/m^2}$, the current density became $2.7 \mathrm{A/m^2}$ approximately 135 s after the desorption process was started. As observed, for the change in the effluent concentration above, the desorption process proceeds rapidly compared with the adsorption process based on the change in the current density.

3.2. Desalination characteristics in CC mode

An adsorption experiment was conducted while supplying a constant current ($10 \mathrm{A/m^2}$) to the cell until the cell potential reached the set value. As soon as it reached the set value, the cell potential was changed to 0.0 V, and a desorption process proceeded. Fig. 3 shows the change in the effluent concentration. The concentrations of the effluents all exhibit the same value of 6.9 mM because a constant current density was supplied during the adsorption process.

When operating the MCDI in CC mode, the adsorption capacity of the carbon electrode increases as the final cell potential increases; thus, the adsorption time increases. When a constant current density of $10 \mathrm{A/m^2}$ was supplied, the time required for the

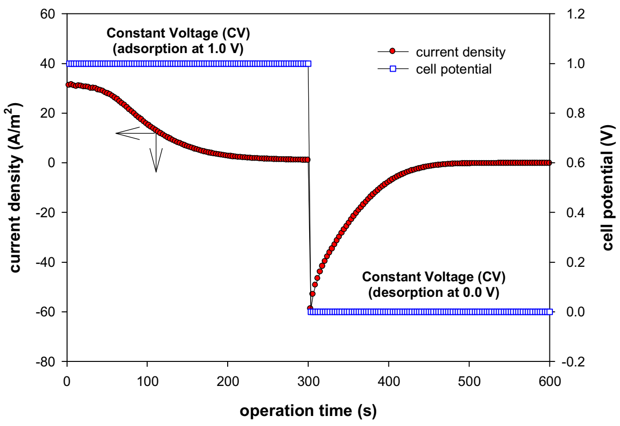

Fig. 2. The changes in the current densities at a cell voltage of 1.0 V for the adsorption process and at 0.0 V for the desorption process.

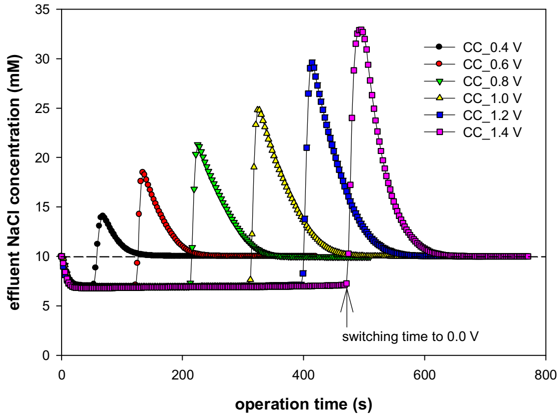

Fig. 3. Effluent NaCl concentrations at a constant current density of 10 A/m2 during the adsorption process. When the cell voltage reached a given value (0.4–1.4 V), the voltage was changed to 0.0 V for the desorption process.

cell potential to reach 0.4, 0.6, 0.8, 1.0, 1.2 and 1.4 V was 48, 120, 210, 309, 402 and 480 s, respectively. As soon as it reached the final cell potential, a desorption experiment was carried out by switching the cell potential to 0.0 V. It can be observed that the desorption process proceeds very rapidly, as shown in Fig. 3.

Fig. 4 shows the changes in the current and voltage of the cell in the desorption process at 0.0 V after the adsorption process proceeded until the cell potential reached 1.0 V at a current density of 10 A/m^2. When the current was applied to the cell, the initial cell potential was 0.16 V and proceeded rapidly for approximately 40 s. The cell potential then increased at a constant rate. It can be interpreted that the initial 0.16 V was a result of the electrical resistance of the spacer and the ion-exchange membranes. In other words, it can be observed that the electrical resistance of the spacer, CMX membrane and the AMX membrane in the MCDI cell is 1.6 Ω.

After the cell potential reached 1.0 V, the initial current flowing in the cell was 60.5 A/m^2 when the cell potential was immediately switched to 0.0 V. Additional current was supplied compared with when in CV mode. The amount of current flowing through the cell is proportional to the number of ions transported. Thus, it is confirmed that conducting adsorption in CC mode is advantageous when attempting to increase the desorption rate.

3.3. Comparison of adsorption amounts in CV and CC modes

The adsorption amount was calculated from the change in the concentration of the effluent. Fig. 5 shows the adsorption amount based on the cell potentials. Additional ions were adsorbed when conducting adsorption in CV mode compared with CC mode.

The adsorption amount of the carbon electrode is proportional to the electrode potential. When supplying a CV to the MCDI cell, a large amount of current initially flows. However, when the adsorption capacity of the electrode achieves saturation, current seldom flows to the cell. Thus, the cell potential corresponds to the electrode potential at the surface of the carbon electrode. In contrast, while current is continuously supplied to the cell as it is adsorbed in CC mode, a voltage drop due to the resistances of the solution in the flow channel and in the ion-exchange membranes occurs. Thus, the electrode potential always has a lower value than the cell potential. As a result, the amount of adsorption is higher in CV mode than in CC mode. However, as observed in Fig. 5, the difference in the adsorption amount decreases as the cell potential increases.

3.4. Comparison of charge efficiency in CV and CC modes

Fig. 6 shows the total charge supplied to the cell during the adsorption and desorption period while operating the MCDI cell in CV and CC modes. The amount of adsorption (or the amount of desorption) of ions during MCDI operation is proportional to the amount of charge supplied to the cell. The amount of charge flowing to the cell shows a tendency similar to the adsorption amount in Fig. 5. Similar to the ion-adsorption rate, more charge was supplied in CV mode compared with CC mode.

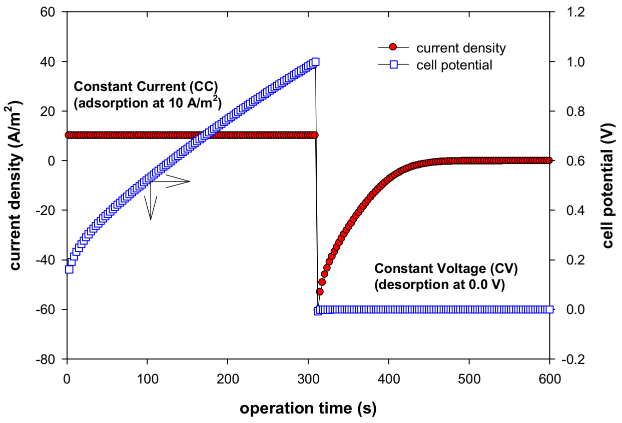

Fig. 4. The changes in the cell voltage and the current supplied. During the adsorption process, a constant current was applied until the cell voltage reached 1.0 V. The cell voltage was maintained at 0.0 V for the desorption process.

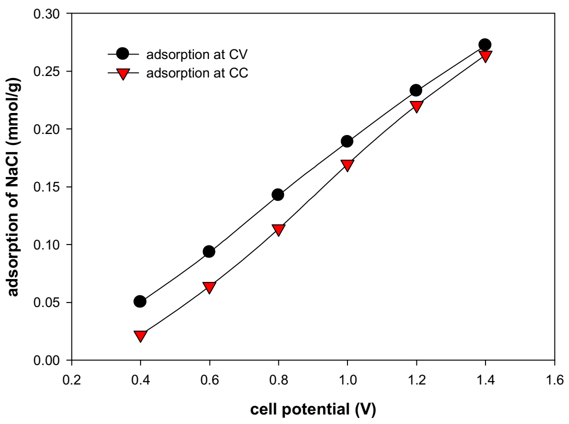

Fig. 5. The amount of NaCl adsorption as a function of cell voltages under constant voltage and constant current operation. The cell voltages under constant current operation indicate the final cell voltage during the adsorption process.

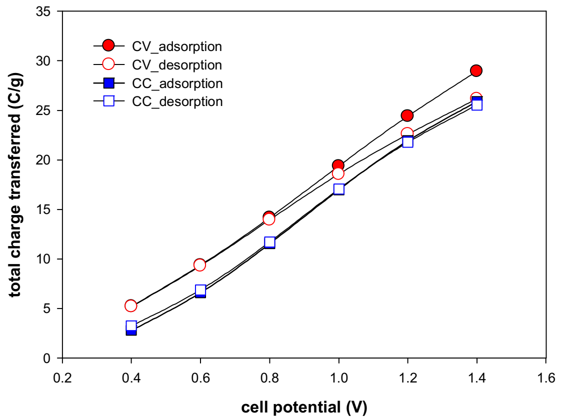

Fig. 6. Total number of charges transferred through the cell for various cell voltages during the adsorption process under constant voltage and constant current operation.

An important feature is noticed when comparing the amount of charge between the adsorption and desorption processes. If all ions adsorbed on the carbon electrode are desorbed, the total amount of charge in the adsorption and desorption processes must be equal. There is little difference in the amount of charge in the adsorption and desorption processes up to a cell potential of 1.0 V. However, when the cell potential becomes greater than 1.0 V, the amount of charge in the adsorption process becomes higher than in the desorption process, and the difference gradually increases as the cell potential increases. In particular, when operating the MCDI process in CV mode, it shows an additional difference compared with when operating the process in CC mode.

In general, if the MCDI process uses a non-faradaic current, then an electrode reaction does not occur. If an electrode reaction occurs at a high cell potential, the current flowing at this time cannot be utilised for adsorbing ions. Instead, it may adversely affect the operation of the process. The reason that the amount of charge in the adsorption process at a cell potential of 1 V or higher is greater than that in the desorption process is determined to be due to the occurrence of oxidation and reduction reactions at the electrodes.

The efficiency of the desalination process can be analysed by calculating the charge efficiency () from the results of the operation of the MCDI process. The charge efficiency is the ratio of the number of ions adsorbed to the number of charges supplied, which is expressed by the following equation [20]:

$$\wedge\ (\%) = \frac{(C_o - C_{\mathrm{eff}})V \cdot F}{\int_{iAdt}} \times 100,$$

where C_o denotes the initial concentration of the influent, C_{eff} is the average concentration of the effluent, V is the total solution volume passing through the cell over the adsorption time, F is Faraday's constant, i is the current density and A is the area of the carbon electrode.

Fig. 7 shows the charge efficiency as a function of the cell potential. It shows the tendency for the charge efficiencies to increase until the cell potential reaches 0.8 V, after which it decreases again. The charge efficiency in CV mode exhibited the highest efficiency of 97.2% at 0.8 V, after which it decreased to 90.7% as the cell voltage increased to 1.4 V. In contrast, the charge efficiency in CC mode exhibited the highest efficiency of 96.4% at 1.0 V and slowly decreased to 94.6% at 1.4 V. The charge efficiency is low at a low cell potential region because the electrolyte within the carbon electrode is adsorbed on the carbon surface. Studies by Biesheuvel and co-workers [17,25] showed similar results in the low cell potential region. However, a decrease in the charge efficiency was not observed in their studies at higher cell potentials.

The charge efficiency decreases in the high cell potential region because the electrode reactions proceed and because faradaic currents are supplied. As mentioned above, the electrode potential at the same cell potential has a higher value in CV mode than in CC mode. Accordingly, when operating the MCDI process at the same cell potential, a higher faradaic current may be found in CV mode compared with CC mode. As a result, the decrease in charge efficiency in CV mode is larger than in CC mode in the high cell potential region.

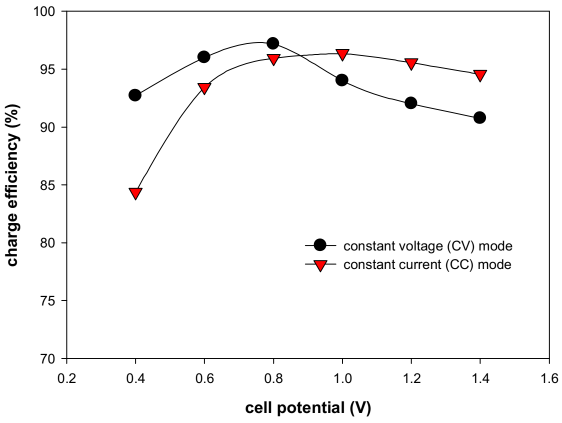

Fig. 7. The variation of the charge efficiencies as a function of the cell voltages under constant voltage and constant current operation.

3.5. Comparison of energy consumption in CV and CC modes

Energy cost is a very important factor when comparing efficiencies in desalination processes. The energy consumption was calculated by multiplying the cell potential and the current and integrating over the duration of the adsorption period. We used kT per ion removed as a unit of energy consumption [20].

Fig. 8 illustrates the energy consumption in the CV and CC modes depending on the cell potential. As the cell potential increases, the energy consumption increases. In addition, it is determined that the energy consumption is much higher in CV mode than in CC mode. The energy consumption in the CV and CC modes at a cell potential of 1.0 V was 20.6 and 13.0 kT per ion removed, respectively. As a result, the energy consumption was approximately 60% higher in CV mode than in CC mode. It is considered that the energy consumption is high because a substantial amount of current is supplied during an initial adsorption process when operating the MCDI process in CV mode. In addition, it is considered that the electrode reactions occur at a greater than 1.0 V cell potential, and this increases the energy consumption.

The MCDI process is recognised as a process whose energy consumption is lower than other conventional desalination processes. Through this study, it is confirmed that when operating the MCDI process in CC mode, the energy consumption can be further reduced. Therefore, in terms of energy costs, the MCDI process is seen as a highly competitive desalination technology compared with conventional desalination processes.

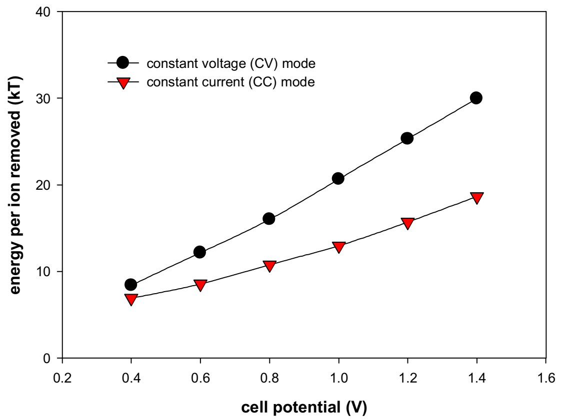

Fig. 8. Comparison of energy consumption under constant voltage and constant current operation.

4. Conclusion

The characteristics of the desalination process with different power supply modes using the adsorption process of MCDI were systematically analysed in this study. After conducting adsorption experiments in which the cell potential was varied under constant voltage and constant current conditions, desalination characteristics such as the adsorption of ions, charge efficiency and energy consumption were compared for different power supply modes.

When operating the MCDI in CV mode, more ions were adsorbed compared with when operating in CC mode. However, the charge efficiency was higher in CC mode than in CV mode. In particular, the higher the cell potential was, the higher the electrode reaction rate in CV mode, resulting in a greatly reduced charge efficiency. In addition, the energy consumption was significantly lower when operating the MCDI in CC mode than in CV mode.

It is very important to determine the optimal operating conditions when operating an MCDI system. The optimum conditions of the MCDI process may vary depending on the salt concentration of the influent, the desalination rate and the concentration of the effluent. It is believed that the results of this study will provide useful information for determining the optimal operating conditions in the operation of MCDI systems.

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