



Performance optimization of a pilot-scale membrane capacitive deionization (MCDI) system operating with circulation process

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ABSTRACT

Membrane Capacitive Deionization (MCDI) is a water treatment technology that removes ionic substances from water. However, the MCDI process has low water recovery, which is primarily due to the utilization use of the same feed water for both the adsorption and desorption stages. In this study, we propose a novel operational method called the MCDI circulation process, designed to enhance the recovery rate. The MCDI circulation process involves recycling the concentrated water generated during desorption, leading to a method that yields a high product water to the feed water ratio. We have identified optimal operating conditions and energy consumption patterns by applying this method to a pilot-scale MCDI device equipped with various module array. The results indicate that employing a Quadruple array achieved an impressive total dissolved solids (TDS) removal efficiency of over 94 %. When considering varying flow rates, it was observed that lower flow rates led to higher adsorption efficiency, while higher flow rates were more favorable for desorption efficiency. Under these conditions, the energy consumption averaged approximately 0.797 kWh/m³, and the yield of the circulation process reached an impressive 90 % over the course of 10 cycles.

1. Introduction

Capacitive deionization (CDI) is a desalination technology that relies on electrodes made of carbon materials with a substantial surface area and excellent electrical conductivity [1,2]. This technique harnesses the electrostatic forces of the electric double layer formed on the electrode surface when voltage is applied, effectively adsorbing and removing ions from a solution into the micro-pores of the electrode [3,4,5]. However, in practice, charge movement can occur through ion repulsion from the electrodes, particularly when ions are not completely removed, resulting in reduced desalination efficiency [6,7]. To address this challenge and enhance performance, membrane capacitive deionization (MCDI) was developed, which combines CDI with ion-exchange membranes [8–10]. Since the mid-2000s, MCDI has been the subject of active research, with a focus on preventing the emission of ions sharing the same polarity as the electrodes, thereby improving desalination efficiency [11].

The choice of the charging method in MCDI is a critical factor in determining desalination characteristics. There are two primary methods: constant current (CC) and constant voltage (CV) [12–14]. The

CC method consumes less energy than the CV method but exhibits lower adsorption capacity within the same time frame [15,16]. In contrast, the CV method is faster and more efficient during desalination but may demonstrate varying adsorption rates [17,18]. Consequently, the CC and CV methods can be selectively operated based on the specific goals of the MCDI process [19].

Furthermore, selecting electrode type plays a pivotal role in MCDI systems. Two electrode structures are prominent: mono-polar and bipolar [20,10]. Mono-polar electrodes consist of stacked electrodes connected in parallel, operating at a voltage of 1.2 V and displaying increased current with higher influent concentration and flow rates [21,22]. However, these electrodes are complex to manufacture, costly, and exhibit relatively low energy efficiency [23,24]. In contrast, bipolar electrodes allow for stacking multiple units and feature a pair of metal electrodes at each end, minimizing electrical losses compared to mono-polar electrodes, resulting in improved desalination efficiency [25].

The typical operation of MCDI involves repeated cycles of adsorption, facilitated by the application of a positive potential, followed by desorption through negative potential application, while feed water is

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continuously supplied between the electrodes [26,27]. However, due to the necessity for a continuous feed water source, MCDI faces challenges associated with low recovery rates, meaning lower efficiency in terms of treated water volume compared to the feed water volume [28]. Various efforts have been made to enhance efficiency, such as reducing flow rates and velocity during desorption [29–31]. However, fundamentally, MCDI operates with repetitive adsorption and desorption cycles using the same feed water, which inherently limits the recovery rate. Additionally, the operational method has limitations, particularly when the ground water supply is restricted or when continuous desorption leads to wastewater generation [32].

The development of pilot-scale devices for MCDI is crucial for validating the scalability of the technology from the laboratory to real-world applications, assessing its performance under diverse conditions, and optimizing operational parameters such as flow rates and electrode materials [33,34]. These pilot-scale tests enable a comprehensive feasibility analysis, including economic viability and cost-effectiveness assessments, which are essential for decision-makers considering the adoption of MCDI in water treatment applications [18]. Moreover, pilot-scale demonstrations contribute to industry acceptance by showcasing the technology's effectiveness and reliability on a larger scale while identifying and addressing potential challenges that may arise during full-scale implementation [35]. In summary, pilot-scale testing is integral to the successful transition of MCDI from laboratory research to practical, industry-ready solutions [36–40].

In this study, we designed and implemented a pilot-scale device with practical applicability for on-site deployment [41]. The objective was to conduct experiments applying the MCDI circulation process to derive effective operational conditions at a pilot-scale magnitude. The device, capable of accommodating four MCDI modules, was constructed, and the adsorption and desorption characteristics based on module arrangements were assessed by monitoring Total Dissolved Solids (TDS) concentration variations. Utilizing the MCDI circulation process at the

pilot-scale allowed us to gain insights into optimal operating conditions and further explore the dynamic adsorption and desorption behaviors in response to varying module configurations.

2. Experimental

2.1. MCDI module

The MCDI module (O2&B Co., Seoul, Republic of Korea) used in this study, consisted of 250 porous carbon electrodes in a single module, as shown in Fig. 1. Each unit electrode was circular, with a diameter of 200 mm. Anion- and cation-exchange membranes were attached to the top and bottom of each carbon electrode, with a 180- μ m nylon spacer inserted between the ion exchange membranes to allow fluid to pass through. Within the electrodes, ion-exchange membranes, and spacers, there was a circular hole with a diameter of 25 mm. Feed water entering through the inlet is collected from all sides of the cell to the center through spacers, and exits through the outlet at the center of the module.

2.2. Pilot unit and operation conditions

The pilot unit consisted of two parts, as shown in Fig. 2. One part comprised a cartridge filter and an MCDI module, while the other part included a concentrate water tank, recycle pump, power supply, and PC for the MCDI circulation process. The feed water tank and feed pump for supplying the feed water, as well as the treated water tank, were installed outside the pilot unit. The MCDI modules in the pilot unit were designed for assembly in various configurations, accommodating up to four modules. In this study, four configurations were used employed: “Single,” with one module, “Serial” and “Parallel,” with two modules, and “Quadruple,” with four modules.

Feed water was pumped at various flow rates using a peripheral pump (HBI 4–30, Stairs, Taiwan). Given the vulnerability of the MCDI

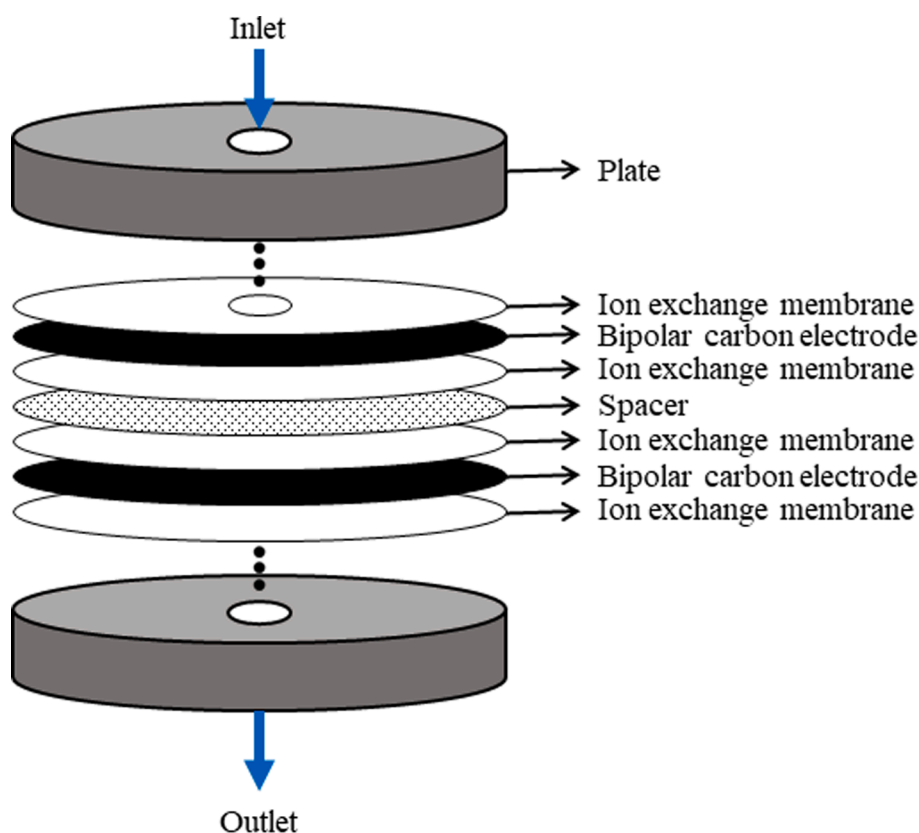


Fig. 1. Schematic representation of the MCDI module configuration.

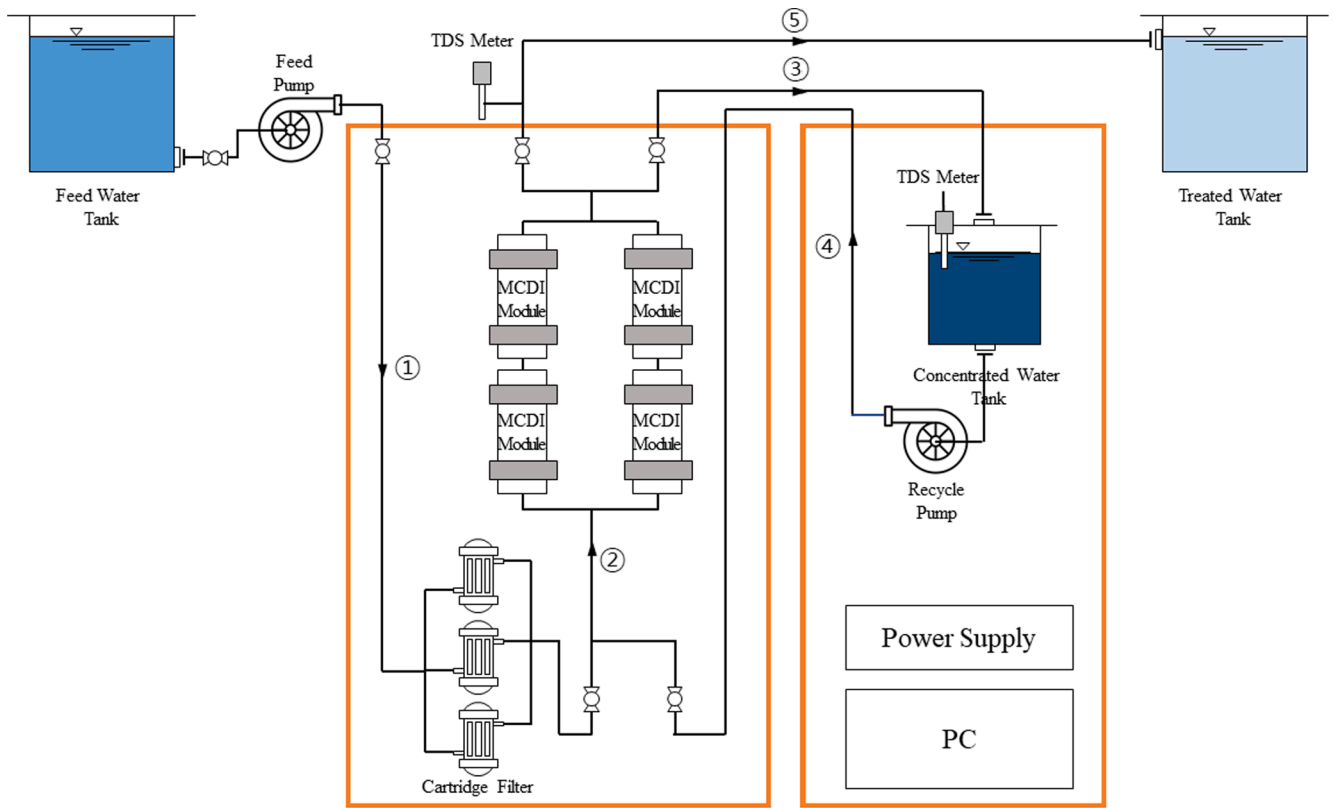


Fig. 2. Schematic diagram of the MCDI pilot unit.

process to organic matter, a 100- μm filter was used for water pre-treatment before passing through the MCDI module. The pilot unit operated in CV mode, with a DC power supply (SJ-1260AN, SENICS, Republic of Korea) providing a constant voltage. A stable voltage of 1.2 V per cell was supplied, considering that the MCDI module consisted of 250 bipolar electrodes, totaling 300 V. The current was limited to not exceed 30 A, and the actual current supplied to the MCDI module varied depending on the influent concentration.

The TDS of the treated water passing through the MCDI module and the concentrate were measured at 10-seconds intervals using TDS meters (3-2822-1 electrode, GF Signet, USA), with the data collected via a PC connection.

2.3. MCDI circulation process

The MCDI process typically involves of two continuous cycles: adsorption, where a constant voltage is applied to capture ions and produce water, and desorption, where a reverse voltage is applied to

release ions and clean the module. As shown in Fig. 3, the MCDI process repeats these two cycles to produce water. However, since the same feed water is used, the wastewater generated during the desorption process is typically discarded.

The experimental conditions of the MCDI circulation process used in this study are provided in Table 1. Unlike the MCDI process, the MCDI circulation process requires a preparatory step, because it does not use feed water during desorption [30]. The preparatory step corresponds to the first full cycle of the MCDI circulation process, which includes the first adsorption($① \rightarrow ② \rightarrow ③$) and desorption($④ \rightarrow ② \rightarrow ③$) cycles. During this step, the product water generated in the first adsorption cycle is stored in the concentrate tank and exclusively used for desorption. Once the preparation step is completed, feed water is used for product water generation during the adsorption process($① \rightarrow ② \rightarrow ⑤$), while the concentrated water from the concentrate tank is employed during the desorption process($④ \rightarrow ② \rightarrow ③$). In other words, unlike the MCDI process, the MCDI circulation process does not use feed water during desorption but instead concentrates and reuses the desorption solution.

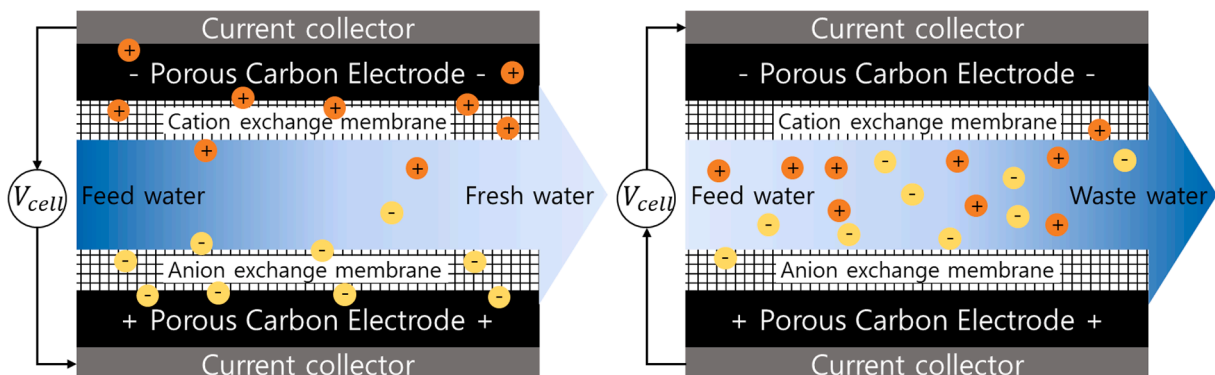


Fig. 3. Adsorption(left) and desorption(right) processes in MCDI.

Table 1

Experimental conditions of the MCDI circulation process experiment.

		Experimental Process	Water flow	Feed water (mg/L of NaCl)	Applied voltage (V)	Operation time (sec)
MCDI circulation process	Ready (1st cycle)	Adsorption	①→②→③	1,000	+300	120
		Desorption	④→②→③	Concentrated water	−300	120
	Process	Adsorption	①→②→③	1000	+300	120
		Desorption	④→②→③	Concentrated water	−300	120

The feed water used in the experiment, with a TDS of 1000 mg/L, was prepared by adding sodium chloride (99.5 %, Samchun, Republic of Korea) to distilled water. During desorption, the concentrated solution from the concentrated tank was used. Both the adsorption and desorption times were set to 120 s, with a voltage of +300 V applied during the CV mode and −300 V during desorption.

2.4. Data analysis

The TDS concentration removal efficiency was calculated using Eq. (1):

$$\text{TDS removal efficiency (\%)} = \frac{C_o - C}{C_o} * 100 \quad (1)$$

where C_o and C represent the initial and final TDS (mg/L) in the feed water and treated water, respectively.

The concentration ratio was calculated using Eq. (2):

$$\text{Concentration ratio (\%)} = \frac{C_{n+1} - C_n}{C_n} * 100 \quad (2)$$

where C represents the TDS (mg/L) in the recycle tank water at 240 s and n is the number of cycles (1–10).

The energy consumption was calculated using Eq. (3):

$$\text{Energy consumption (kWh/m}^3\text{)} = \frac{P * t}{V}$$

where P represents the power during the process, t represents the time of operation and V represents the volume of water treated.

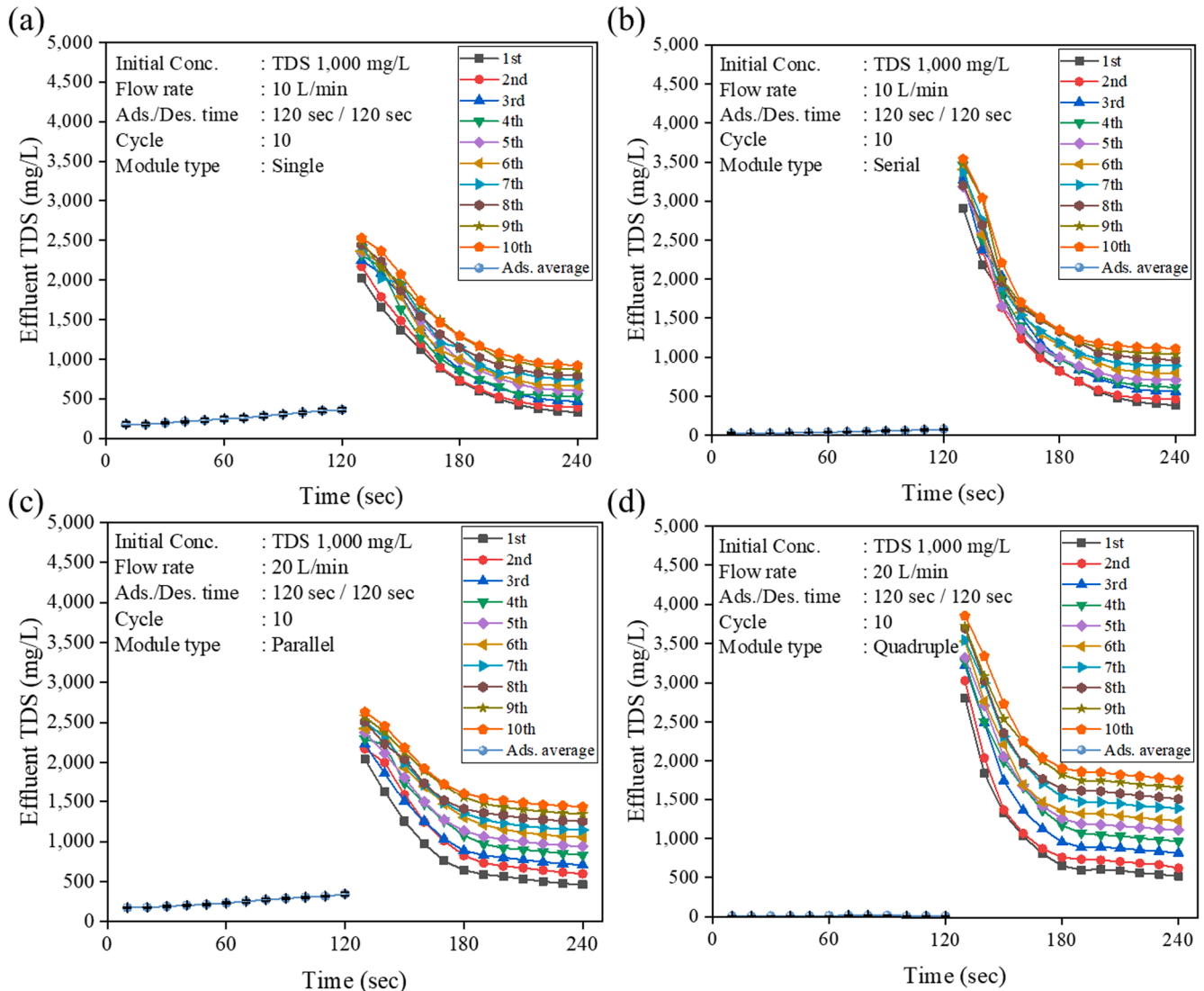


Fig. 4. Effluent TDS concentration for the (a) Single, (b) Serial, (c) Parallel, and (d) Quadruple array configurations.

These equations were used to assess the performance and efficiency of the MCDI circulation process in terms of TDS removal, concentration increase over cycles and energy consumption.

3. Results and discussion

3.1. Variations in TDS concentration according to module arrangement

In this study, we designed a pilot unit capable of accommodating four MCDI modules. We conducted experiments using different configurations: Single, Serial, Parallel, and Quadruple array, each processing a flow rate of 10 L/min through one module. The Single and Serial arrays operated at a flow rate of 10 L/min, while the Parallel and Quadruple arrays operated at 20 L/min. The experiment was carried out over 10 cycles, with adsorption and desorption times set at 120 s each. Fig. 4 (a)–(d) presents the experimental results for these four module arrangements.

The results showed that, even at the same flow rate of 10 L/min per MCDI module, the TDS concentrations during adsorption and desorption varied depending on the module array configuration. The Single and Parallel arrays exhibited similar TDS concentrations for adsorption, reducing removal efficiency from 83 % to 63 %. In contrast, the Serial and Quadruple arrays achieved a removal efficiency of over 90 %, surpassing that of the Single and Parallel arrays. This enhancement is attributed to the continuous connection of MCDI modules in the Serial and Quadruple arrays. In the Single and Parallel arrays, the fluid passes through a single module, whereas in the Serial and Quadruple arrays, it passes through a single module twice, allowing more ion adsorption. Therefore, the continuous connection of modules in the Serial and Quadruple arrays allowing the fluid to pass through a single module twice, resulting in improved ion adsorption.

During desorption, the TDS concentration in the concentrate tank differed among the array configurations after 10 cycles. The TDS concentrations for the Single, Serial, Parallel and Quadruple arrays were 920, 1,110, 1,435, and 1,760 mg/L, respectively. Contrary to expectations, the Serial and Quadruple arrays, which demonstrated superior removal efficiency during adsorption, exhibited higher concentrate concentrations than the Single and Parallel arrays. This discrepancy can be attributed to the higher flow rates in the Parallel and Quadruple arrays, resulting in relatively higher concentrations than those achieved with the Single and Serial arrays. This is believed to be because increasing the flow rate to rapidly desorb the ions adsorbed on the cell is more

influential in obtaining concentrated water than having a continuous arrangement of MCDI modules for brine collection. As a result, the concentrated TDS of Parallel and Quadruple arrays with high flow rates was high.

3.2. Adsorption and TDS removal

Building upon the adsorption results discussed above, further experiments were conducted using the Serial and Quadruple arrays, which showed > 90 % removal efficiency at flow rates of 10, 15, and 20 L/min Fig. 5.

Fig. 5a shows the results for the Serial configuration. As the flow rate increased, the TDS concentration in the treated water increased, resulting in lower removal efficiency. At a flow rate of 10 L/min, the TDS removal efficiency was highest, at approximately 92 %. However, at flow rates of 15 L/min and 20 L/min, the removal efficiency dropped to approximately 88 % and 86 %, respectively. This decline can be attributed to the extended residence time of water within the MCDI system at lower flow rates, allowing for increased interactions between ions in the water and the electrode surfaces, thereby promoting higher adsorption efficiency.

Fig. 5b shows the results for the Quadruple array. For this configuration, the TDS concentration in the treated water remained relatively consistent across different flow rates. At all flow rates tested, the removal efficiency consistently exceeded 94 %. The 20 L/min condition for the Quadruple array involves dividing the flow into 10 L/min per MCDI unit module, which is the same as the 10 L/min condition for the Serial array. However, the TDS removal efficiency was higher with the Quadruple array. This indicates that, the number of electrodes used affects the desalination efficiency even at the same flow rate.

3.3. Concentrated water TDS concentration and ratio

The MCDI circulation process involves continuous cycling of adsorption and desorption processes, potentially affecting subsequent adsorption. Understanding the TDS concentration and characteristics of the concentrated water is therefore crucial. Fig. 6 illustrates changes in TDS concentration and concentration ratio in the concentrated water tank over 10 cycles, based on the MCDI module array. As shown in Fig. 4, the TDS concentration in the concentrated water followed this order: Single < Serial < Parallel < Quadruple. The concentration ratio was 12.19 % for the Single, 12.58 % for the Serial, 13.59 % for the

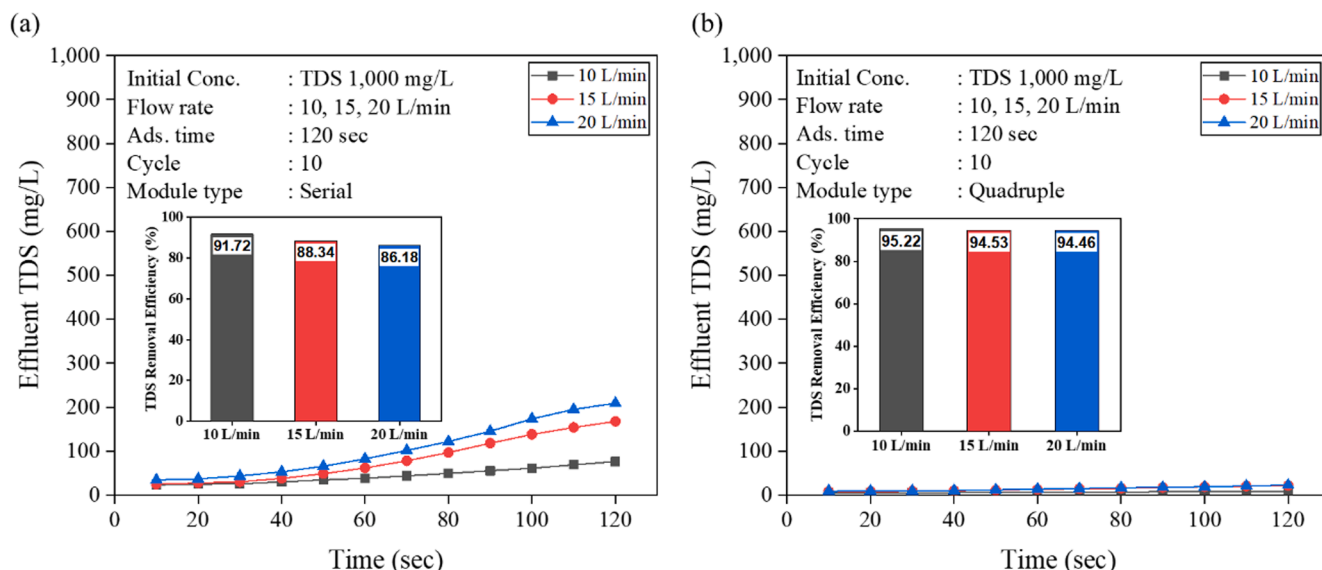


Fig. 5. Adsorption and TDS removal efficiency of the (a) Serial and (b) Quadruple arrays.

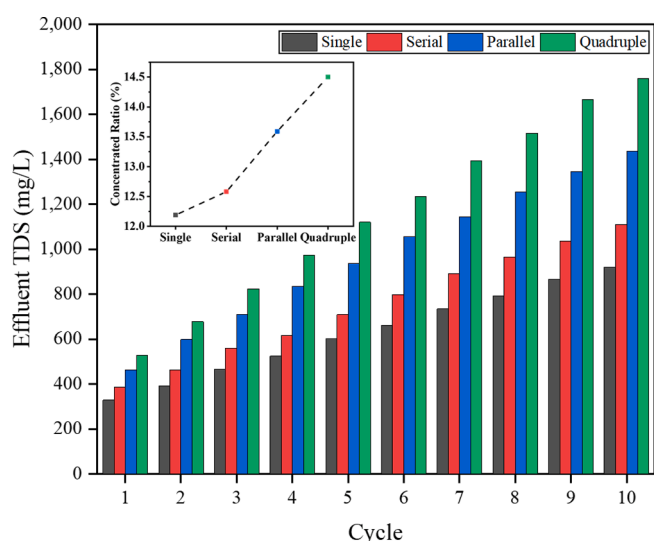


Fig. 6. Differences in TDS concentration and concentration ratio among the MCDI module arrangements over 10 cycles.

Parallel, and 14.5 % for the Quadruple array. This suggests that the TDS concentration in the concentrated water is proportional to the rate at which ions are desorbed and concentrated in the cells, depending on the array.

Fig. 7 presents the TDS concentrations in the concentrated water at different flow rates for the Serial and Quadruple arrays, which showed > 90 % removal efficiency during adsorption. Experiments were conducted at flow rates of 10, 15, and 20 L/min. The results indicated that, as observed during adsorption, the desorption characteristics were similar at the same flow rate, regardless of the module array. However, the effluent TDS concentration at flow rates of 15 L/min and 20 L/min appeared similar. This suggests that, similar to the above findings for adsorption, during desorption, rapidly removing ions from the cells through higher flow rates has a greater impact on the concentrated water than the MCDI module arrangement.

3.4. Energy consumption and recovery rate

Fig. 8(a) and 8(b) present the energy consumption and recovery rate for the MCDI circulation process. Fig. 8(a) compares the energy consumption over 10 cycles for different MCDI module arrays. The Single

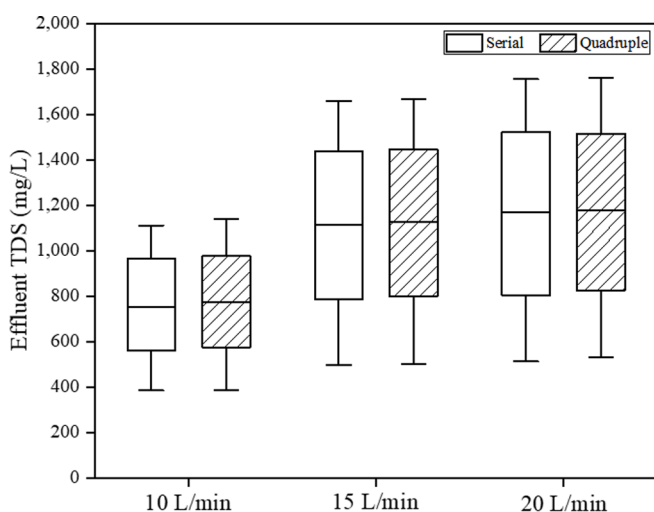


Fig. 7. TDS concentrations in the concentrated water at different flow rates for the Serial and Quadruple arrays.

array had the lowest energy consumption, at 0.438 kWh/m³, while the Quadruple array had the highest energy consumption, at 0.797 kWh/m³. This pattern is similar to that of the TDS concentrations in the concentrated water. In the MCDI circulation process, the reuse of concentrated water during desorption results in the influent supplied to the MCDI device containing concentrated ions. This leads to a gradual rise in the TDS concentration and, consequently, an increase in power consumption. Therefore, in the case of the Quadruple array, it exhibited the highest energy consumption due to the presence of a higher concentration of concentrated ions in the MCDI cells.

The MCDI circulation process differs from the conventional process in that it does not use the same feed water during adsorption and desorption. In the conventional process, the same feed water is used for adsorption and drain during desorption, resulting in a recovery rate of 50 % because half of the feed water is recovered as product water. In contrast, the MCDI circulation process secures product water equivalent to the incoming water; hence, the recovery rate increases as the cycles progress. Fig. 8(b) illustrates this behavior. In this process, the product water from the first cycle is stored in the concentrated water tank for desorption. As a result, the recovery rate is 0 % in the first cycle, but from the second cycle onwards, the product water obtained increases, and over 10 cycles, a recovery rate of 90 % is achieved.

4. Conclusions

In this study, we investigated the TDS removal and concentration efficiencies of the MCDI circulation process. Experiments were conducted by configuring MCDI modules into Single, Serial, Parallel, and Quadruple arrays. The results revealed variations in TDS removal efficiency during adsorption and concentration efficiency during desorption, depending on the configuration.

Regarding adsorption, the Serial and Quadruple arrays demonstrated higher TDS removal efficiencies than the Single and Parallel arrays. This is likely due to the continuous passage of ions through two modules, enabling greater ion adsorption. When we conducted experiments with different flow rates using the Serial and Quadruple arrays, both showed similar desorption characteristics. This suggests that increasing the flow rate to rapidly remove ions adsorbed in the cells significantly impacts concentrate recovery more than the module array. The Quadruple array exhibited the highest power consumption, which is consistent with the increase in the TDS concentration during desorption, as the concentrate is reused for desorption in the MCDI circulation process. However, recovery rates in the MCDI circulation process increased as the cycles progressed because the influent water differed between the adsorption and desorption processes.

The proposed MCDI circulation process not only secures a substantial amount of production water compared to the influent water but also enables the distinction of TDS removal and concentration efficiencies based on module configurations. Additionally, the MCDI technology proposed in this study can be applied to various areas, including households or small communities relying on brackish water sources, as well as in regions where brackish water is utilized for irrigation, where MCDI technology can effectively reduce the salinity of the water. Therefore, based on its proven versatility, effectiveness and as well as high recovery rate, the MCDI circulation process stands out as a promising technology for a wide range of water treatment applications.

CRedit authorship contribution statement

Changseog Oh: Writing – original draft, Investigation, Data curation, Conceptualization. **Bokjin Lee:** Writing – original draft, Investigation, Data curation, Conceptualization. **Jusuk An:** Writing – review & editing, Funding acquisition. **Seungjae Yeon:** Writing – review & editing, Funding acquisition. **Hyun Je Oh:** Writing – review & editing, Funding acquisition.

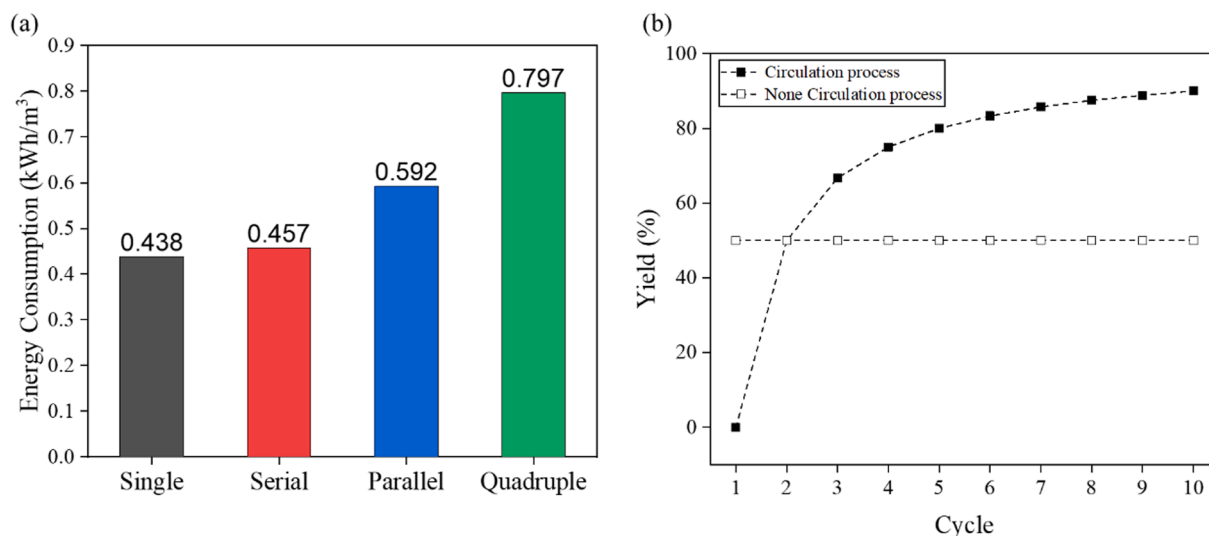


Fig. 8. (a) Energy consumption and (b) recovery rates in the MCDI circulation process.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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