


## Investigation of effective parameters on brackish water desalination by flow-electrode capacitive deionization

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### ABSTRACT

Population growth and increasing global demand for freshwater have raised a serious challenge for the depleting sources of freshwater in the 21st century. Desalination technologies can be a reliable technique for providing freshwater. Capacitive deionization is one of the innovative desalination methods that has received increasing interest. Flow-electrode capacitive deionization (FCDI) (a new architecture of capacitive deionization) is one of the efficient, cost-effective, and environmentally friendly desalination methods for freshwater production. In this experimental research, the performance of an FCDI system was investigated and the influence of important parameters such as flow rate of flow-electrodes, electrolyte salt concentration of flow-electrodes, and initial feed water concentration will be assessed on the efficiency of desalination operation. In this study, the flow-electrodes operated in short-circuited closed-cycle operation (SCC) mode, and also the feed water operated similarly to the flow electrodes in closed cycle. Moreover, in all the experiments, the salt adsorption capacity (SAC) and salt removal efficiency (SRE) was calculated. Herein, by optimizing the above-mentioned parameters, the salt removal efficiency of 83% and a SAC value of 29.12 mg/g<sub>dry carbon</sub> were achieved in 5 hours.

**Key words:** flow-electrode capacitive deionization, salt adsorption capacity (SAC), salt removal efficiency (SRE), water desalination

### HIGHLIGHTS

- One of the newest desalination technologies with very low energy consumption, known as flow-electrode capacitive deionization (FCDI) is discussed.
- The components and materials used in this system (FCDI) and its modes of operation are introduced according to latest researches.
- The effect of different parameters on the efficiency of this system for brackish and salt water desalination is discussed.

## 1. INTRODUCTION

Water is a basic need for human survival and thus, access to safe and clean water is an important necessity for human societies. In recent decades, factors such as population growth, industrial and economic growth, improved standards of living, changing consumption patterns and the expansion of agriculture have increased global demand for water and thus pressure on water resources in many parts of the world. About two-thirds (4 billion people) of the world's population is affected by severe water shortages in at least one month of the year, nearly half of them in India and China. It is remarkable that half a billion people in the world suffer from severe water shortages all year round (Mekonnen & Hoekstra 2016). The United Nations Educational, Scientific and Cultural Organization (UNESCO) also estimates that nearly one-third of the world's population now lives in water-scarce areas, a figure that is expected to double by 2025 (Zhang *et al.* 2018).

Water resources cover about three-quarters of the Earth's surface. Nevertheless, the acute shortage of drinking water in Africa and the countries of the Middle East is extremely tangible. Only 2.5% of the Earth's water resources are freshwater in groundwater, lakes, rivers, surface water, and also in the form of ice and snow in mountainous and polar areas (inaccessible), and much of the resources as salt water are in seas and oceans and some lakes (Shatat & Riffat 2012; Youssef *et al.* 2014). To find a solution to the problem of water scarcity, desalination of sea and brackish water can be considered as a suitable and economical option.

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According to the guidelines of the World Health Organization (WHO), the permissible total dissolved solids (TDS) level of potable water should be less than 500 ppm but, in certain cases, it can be as high as 1,000 ppm. However, TDS concentration is reported in the range of 1,000–25,000 ppm for brackish water and 35,000–45,000 ppm for seawater (Shatat & Riffat 2012; Youssef *et al.* 2014). Therefore, the use of an efficient and effective desalination method can be very helpful and promising in reducing the urgent need for water around the world. Various factors are involved in the selection of a desalination process for the production of potable water, such as the quality of the feed water entering the desalination system, the concentration of the desalinated water (product), the amount of energy entering the system, the environmental impact (e.g., CO<sub>2</sub> emissions and secondary pollutants), and the cost.

The most common desalination technologies in water-scarce areas around the world include thermal phase change processes, membrane processes, and chemical desalination methods. Among the most important thermal phase change processes are multistage flash distillation (MSF), multiple-effect distillation (MED), mechanical vapor compression distillation (MVC), humidification–dehumidification desalination (HDH), solar distillation (SD) and freezing–melting (FM). It can be stated that the most important membrane processes include reverse osmosis (RO), forward osmosis (FO), electro-dialysis (ED), membrane distillation (MD), and nanofiltration (NF). In addition, to introduce chemical desalination processes, one can mention ion-exchange desalination (I.Ex), liquid–liquid extraction (LLE), and gas hydrate (G.Hyd). Most desalination facilities in the world are based on RO and multistage flash distillation (MSF) methods (Lee *et al.* 2018b). Due to the significant energy consumption involved in these technologies (RO: 2–4 KWh m<sup>-3</sup>, MSF: 55–80 KWh m<sup>-3</sup>), emerging technologies such as electrochemical desalination processes have attracted considerable attention (Lee *et al.* 2018b). Electrochemical desalination technologies can be introduced as an effective and promising solution for efficient energy management and the reduction of greenhouse gas emissions. An important group of electrochemical desalination processes, known as capacitive deionization process, consists of three main categories: capacitive deionization (CDI), membrane capacitive deionization (MCDI), and flow-electrode capacitive deionization (FCDI).

In the previous study, we discussed the assembly of a typical FCDI system and the removal efficiency of this system in desalination of brackish water (Dehghan *et al.* 2021). In this paper, we now discuss the effect of important and effective parameters on the efficiency of this system for the removal of salt ions from brackish groundwater using the batch mode FCDI experiments under a closed cycle. In other words, by conducting the FCDI experiments and plotting the comparison diagram, we investigate the effect of factors such as flow-electrodes flow rate, initial sodium chloride concentration of feed water, and salt concentration of electrolyte solution of flow-electrodes on the desalination efficiency of the FCDI system.

### 1.1. FCDI process definition

CDI was first introduced by D. D. Caudle (Caudle 1966) and proposed as an option for desalination of brackish water.

In capacitive deionization processes, the feed water is passed through the oppositely charged electrodes. In fact, the electrodes are charged by a potential difference (less than 1.23 V (Hatzell & Gogotsi 2016; Yang *et al.* 2017a)) under an electric field. As a result, the ions in the feed water migrate to the surface of the charged electrodes and are charged accordingly. In this process, the electrical double layer (EDL) actually forms in the electrode/electrolyte interface. After the charged ions reach the surface of the electrodes, they are accumulated inside the EDLs by electrostatic forces and are electrostatically held (Ratajczak *et al.* 2019). The performance of electrical double-layer capacitors (EDLCs) is similar to conventional capacitors. Conventional capacitors consist of a dielectric sandwiched between the two opposing electrodes, but EDLCs use a double layer for nanoscale charge separation (Ratajczak *et al.* 2019). In addition, the structure of electrodes in capacitive desalination processes is composed of nanostructured carbon materials. Among the properties that are expected of carbon materials used in the electrode structure in electrochemical desalination processes are highly extended surface area and pores matching the size of the electrolyte ions, appropriate pore distribution, excellent electrical conductivity, and good chemical stability (Zhang *et al.* 2018; Ratajczak *et al.* 2019).

As mentioned earlier, a conventional CDI cell contains two fixed electrodes located on either side of a spacer. In fact, the space between the two electrodes is considered as a water flow path. By applying an electrical voltage to carbon electrodes, cations (Na<sup>+</sup>) and anions (Cl<sup>-</sup>) in the feed water flowing in the spacer channel, migrate to carbon electrodes (cations are adsorbed by the cathodic electrode and anions by the anodic electrode). The ions are eventually adsorbed in the EDL and consequently, NaCl is removed from the feed water. After ion electrosorption (adsorption), the ion-saturated electrodes should either be replaced with a set of new electrodes or regenerated. Since the fixed electrodes used in conventional CDI have a limited ion adsorption capacity, the saturated electrodes can be regenerated during the ion desorption process by

short-circulating or reversing polarity. In this way, the adsorbed ions return to the brine stream and the electrodes regenerate their ion adsorption capacity (Lee *et al.* 2018a; Zhang *et al.* 2018). Saturation of the electrodes with ions, so that the electrodes do not have the capacity to adsorb more ions, is the main limitation of conventional CDI technology, as it limits the desalination of feed water with very high concentrations. Therefore, researchers have tried to overcome the limitations of conventional CDI using new technologies, such as static electrode architecture (e.g., flow-through electrode CDI (Suss *et al.* 2012), membrane CDI (Wal & Zhao 2012), inverted CDI (Gao *et al.* 2015), hybrid CDI (Lee *et al.* 2014), wire based CDI (Porada *et al.* 2012), and flow-electrode architecture (e.g., feed-in electrodes (Hatzell *et al.* 2014), feed-between electrodes (Suss *et al.* 2015), and membrane flow-electrodes (Jeon *et al.* 2013; Yang *et al.* 2017b).

Membrane capacitive deionization (MCDI) technology is considered a significant improvement over conventional CDI. MCDI is the product of mixing conventional CDI and ion-exchange membranes (IEMs). In MCDI, the IEMs are located in front of the CDI electrodes and, in fact, between the electrodes and the spacer chamber, which increases the adsorption and desorption of ions (compared to conventional CDI) and thus increases ion removal efficiency compared to conventional CDI. In other words, IEMs can be considered as porous separators. Also, the use of membranes is an effective method of reducing certain Faradaic reactions, such as the cathodic reduction of oxygen and the anodic oxidation of carbon (Omosebi *et al.* 2014). During the regeneration process, the membranes limit the adsorption of co-ions<sup>1</sup>, thereby improving the adsorption capacity of the electrodes after the regeneration stage.

As mentioned earlier, the presence of solid electrodes in CDI and MCDI results in salt adsorption being a discontinuous process (adsorption/desorption processes alternate). Moreover, since the solid electrodes are fixed and static on the current collector, the pores in the electrodes are limited. If the electrodes are saturated by the adsorption of ions, the deionization process cannot continue and the electrodes should be regenerated.

To overcome these limitations, the use of a flow-electrode capacitive deionization (FCDI) process can be considered as a suitable option for water desalination. Unlike conventional CDI and MCDI cells where solid-type electrodes are fixed in the cells, FCDI uses the flow-electrode (Yang *et al.* 2017b).

As shown in Figure 1, the components of an FCDI cell generally include a pair of current collectors, cation and anion exchange membranes (IEMs), gasket and spacer chamber (water chamber), and a pair of end plates. In some FCDI systems, the current collectors also play the role of end plates (Lee *et al.* 2018a).

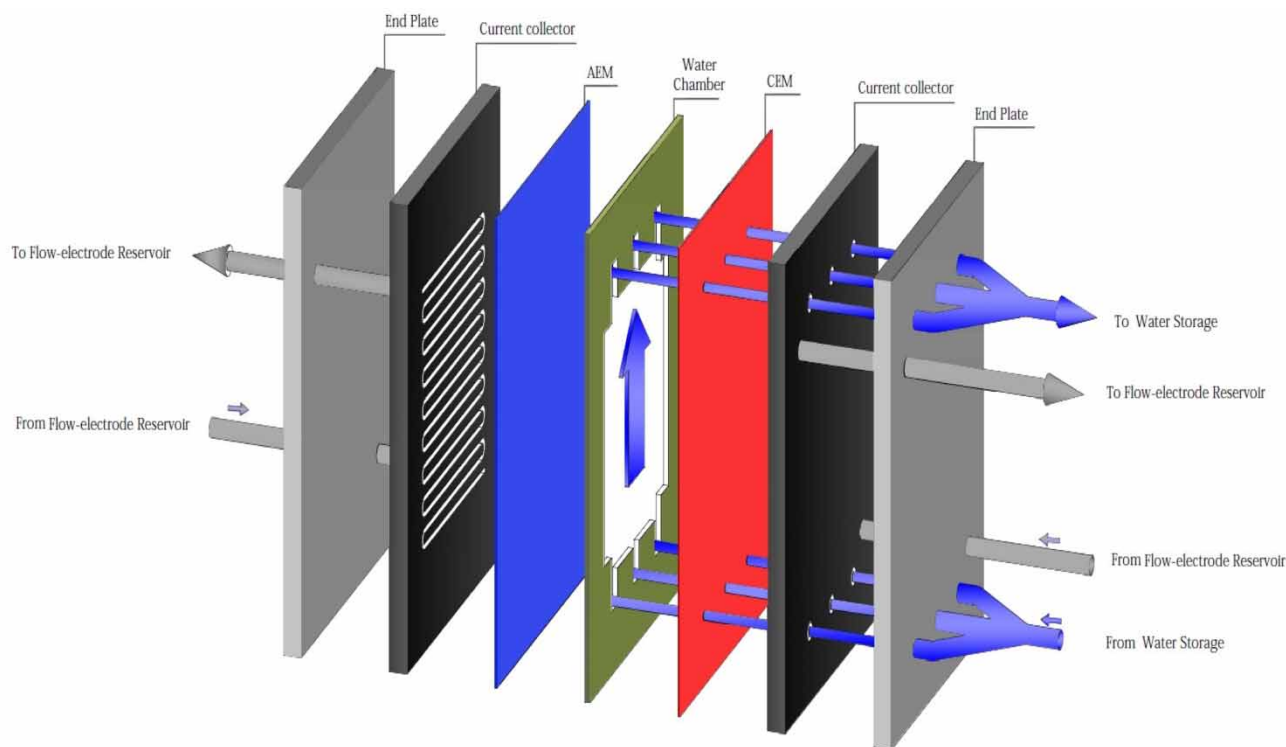
In FCDI, flow channels are carved on the current collector plates to provide a path for the movement of the flow-electrodes. In this process, the flow-electrodes flow between the IEMs and the current collectors (Yang *et al.* 2017b). In FCDI systems, the flow-electrodes can be used in two operational modes: open cycle operation (OC) mode and closed cycle operation (CC) mode.

In the OC mode, when the fresh flow-electrode is continuously injected into the FCDI cell, there is no reduction in the performance efficiency and the salt removal of FCDI cell, because in this mode, the flow-electrodes are continuously replaced and therefore, they have unlimited ion adsorption capacity.

Several laboratory studies in the field of FCDI have been conducted in the CC mode and exploited the limited capacity of flow-electrodes in the experiments. In practical operation of flow-electrodes in FCDI systems, the flow-electrodes must be operated in the CC modes using a limited capacity of flow-electrodes in FCDI systems due to high operation costs (Yang *et al.* 2017b).

The CC modes of the flow-electrodes used in FCDI systems can also be divided into two categories: isolated closed cycle operation (ICC) mode and short-circuited closed cycle operation (SCC) mode. In the ICC mode of FCDI, the anode and cathode flow-electrodes are isolated and the FCDI system has two separate reservoirs for collecting and storing flow-electrodes called anodic and cathodic reservoirs. In other words, in ICC mode, anions and cations are adsorbed separately by anode and cathode flow-electrodes in a continuous manner and accumulated in the anodic and cathodic reservoirs, respectively. Unlike ICC mode, the flow-electrodes in SCC mode are only collected and stored in one reservoir. In other words, in SCC mode, the anode and cathode electrodes are continuously mixed and accumulated in a common reservoir during FCDI desalination operation. During desalination operation, when the flow-electrodes with limited capacities operate under ICC mode in the FCDI cell, the ion adsorption capacity of the cathodic and anodic flow-electrodes is gradually saturated. The desalination efficiency of the FCDI system is thus significantly reduced, as the ions separated from the feed water

<sup>1</sup> The co-ion has the same charge as the electrode during ion removal, which is oppositely charged with the same electrode at the electrode regeneration stage.



**Figure 1** | General structure of an FCDI cell.

and transferred to the flow-electrodes are desorbed by the co-ions present in the flow-electrodes. This is similar to the desalination operation using the conventional CDI technology. In contrast to the ICC mode of flow-electrodes in FCDI systems, the SCC mode with a flow-electrode with a limited capacity has continuous desalination operation without degradation of desalting efficiency. This is because in SCC mode in FCDI systems, salt ions in charged flow-electrodes are neutralized by simply mixing anode and cathode flow-electrodes and then regenerated. Unlike the deterioration of the ICC mode of FCDI desalination performance, no degradation of desalination performance was observed in the SCC mode with a limited capacity of flow-electrode (Yang *et al.* 2017b).

The work of Nativ *et al.* (2018) is one of the new achievements in the field of flow-electrode capacitive deionization (FCDI). They managed to take advantage of the FCDI process to separate monovalent and divalent cations ( $\text{Na}^+$  and  $\text{Mg}^{2+}$ ) and anions ( $\text{Cl}^-$  and  $\text{SO}_4^{2-}$ ) from aqueous solutions. In fact, this process involves selective separation with ion-exchange and nano-filtration (NF) membranes (Nativ *et al.* 2018). This system consists of only one flow-electrode, which is recirculated between the holding vessel and two electrode compartments (flow-electrode). The electrode compartments are located between the electrodes and the membranes. The aqueous solution also contains  $\text{MgSO}_4$  and  $\text{NaCl}$ , which is recirculated between the holding vessel and the FCDI module.

Another important factor contributing to the efficiency of an FCDI system is the material that is used for flow electrode. Activated carbon is often used for this purpose. Some attempts have been also carried out to use other carbon materials such as carbon aerogels (Alam *et al.* 2019a, 2019b), activated charcoal (Nativ *et al.* 2017), activated charcoal/commercial carbon black (Kim *et al.* 2018) and activated carbon (AC)/ $\text{MnO}_2$  composite (Xu *et al.* 2017).

Those materials mentioned above together with numerous other recently published research works reporting on the FCDI demonstrate the capabilities of this novel methodology in terms of continuous operability, high salt absorption capacity, excellent efficiency, cost-effectiveness, and environment-friendliness, so that it can address many of the architectural problems suffered by the CDI, making it a promising technology for the future of water desalination industry.

In this article, an attempt was made to identify the parameters affecting salt removal efficiency in an FCDI system and further evaluate the effect of each parameter in terms of intensity.

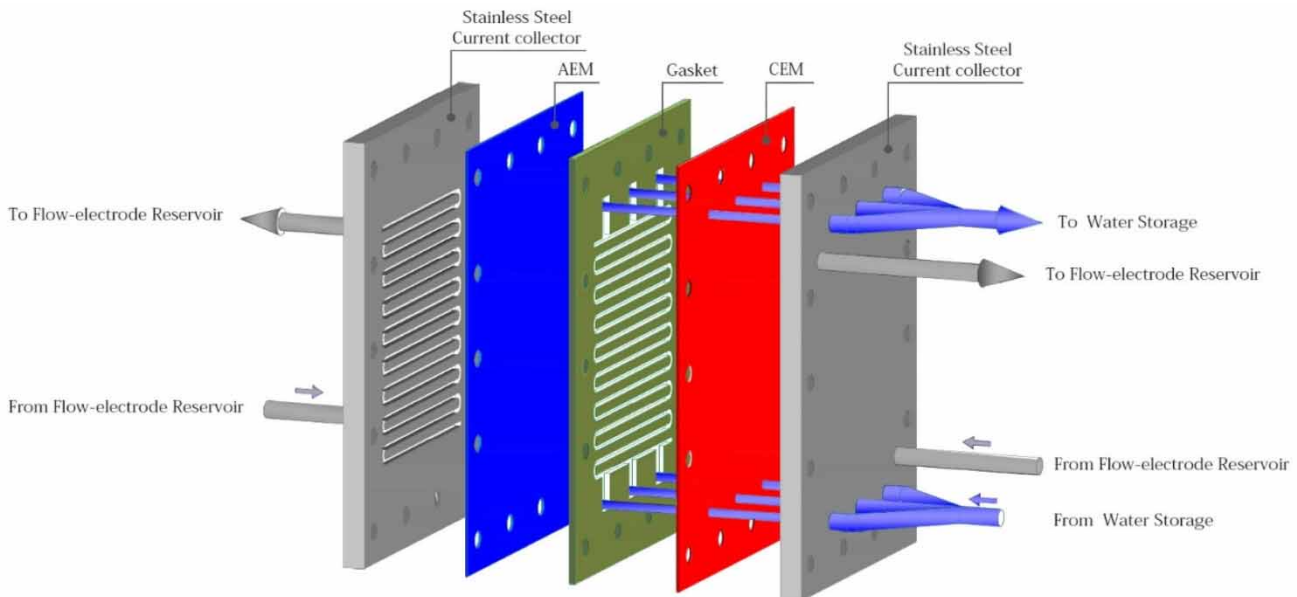
## 2. MATERIALS AND METHODS

### 2.1. FCDI setup and operating condition

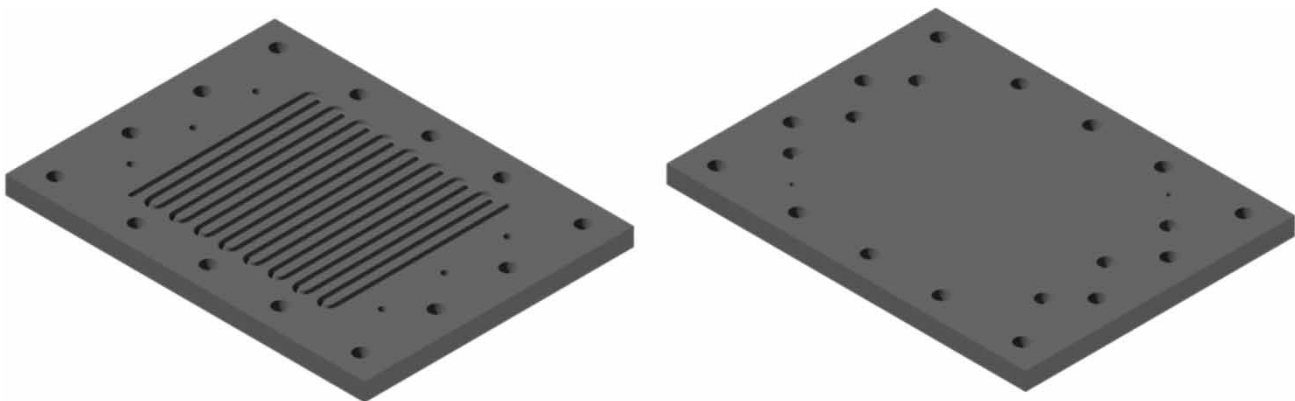
Figure 2 shows the schematic experimental FCDI setup that we used in this experimental study. In the previous work (Dehghan Mirbagheri & Alam 2021), we tried to fully describe the structures, materials, assembling details, and operation of an FCDI system. In this experimental work, the same FCDI cell (with slight modifications) was used.

As shown in the Figure 2, the unit cell of the FCDI system consists of a pair of stainless steel anodic and cathodic current collector plates (type 316) with dimensions 200\*150\*10 mm, a pair of cation and anion exchange membranes with a thickness of 130  $\mu\text{m}$  (Fumasep FKS-PET-130/ED-100 and Fumasep FAS-PET-130/ED-100 (Fumatech GmbH, Germany)), a spacer chamber or the water chamber including a PTFE (polytetrafluoroethylene) gasket (as shown in the Figure 2) at the center of the FCDI cell. The meandering flow channels were carved on the current collector plates using the CNC engraving machine.

The channels were intended as flow paths for suspension flow-electrodes. The width and depth of the flow channels are 2.5 mm, the column length of the flow channel is 102.4 mm, and each current collector plate consists of a total of 17 columns (as shown in Figure 3).



**Figure 2** | Schematic arrangement of our FCDI cell components.



**Figure 3** | Photographs of cathodic stainless steel current collector carved with meandering flow channels.



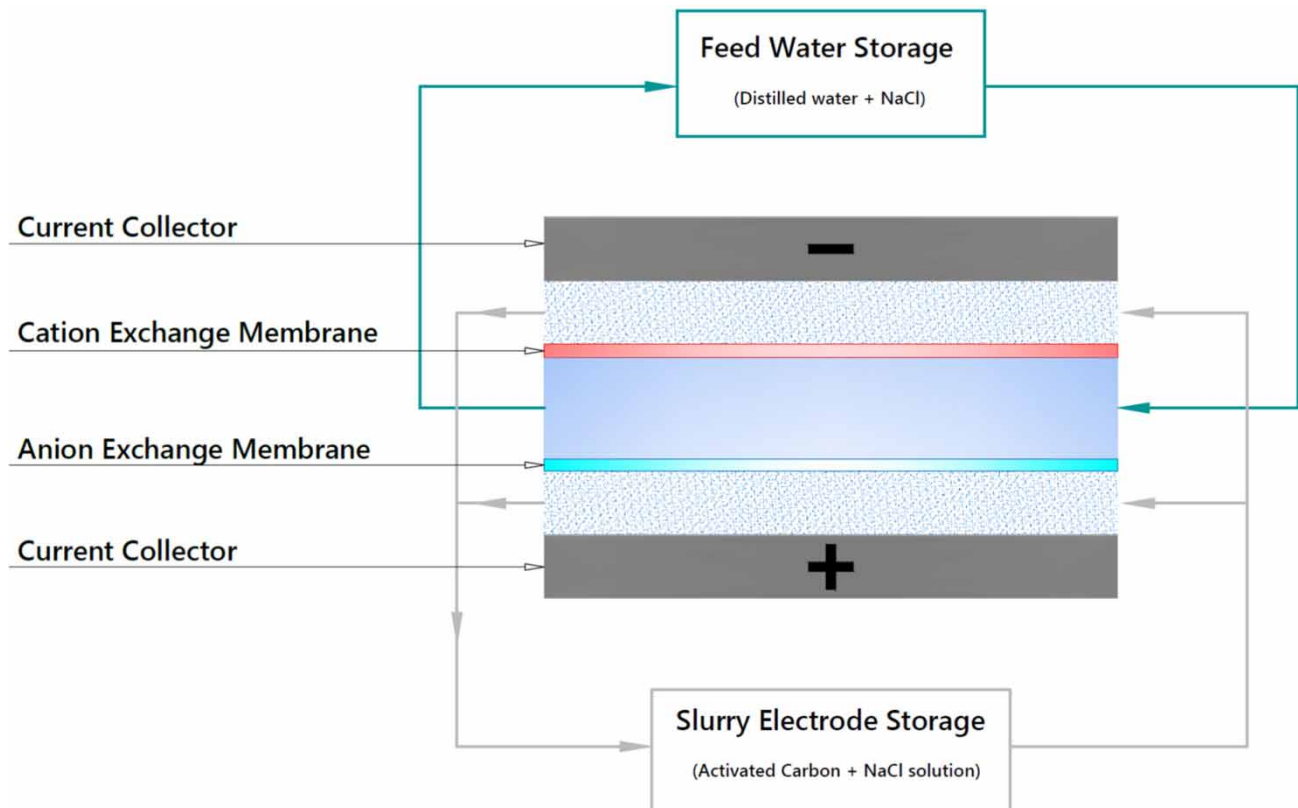
The contact area between the IEMs and the flow-electrodes is about 4,352 mm<sup>2</sup>. All components of the FCDI cell were assembled with stainless steel nuts and bolts in the order shown in Figure 2.

In all experiments, each reservoir of feed water and flow-electrodes contained 100 ml NaCl solution and about 250 ml activated carbon suspension (as ion adsorbent), respectively. The particle size of the activated carbon powder (Norit, Netherlands) used in this experimental work is smaller than 5  $\mu\text{m}$ .

In this FCDI system, the feed water and the suspension flow-electrodes are continuously recirculated between the reservoirs containing water and electrodes and the FCDI cell by means of the peristaltic pumps at the specified flow rates in the CC mode. Consequently, each of the flow-electrodes enters the FCDI cell from the top of each current collector plate exits the cell after passing through the flow channels from the bottom of the current collector plates and returns to the same electrode reservoir. To neutralize and regenerate the flow-electrodes, both anode and cathode electrodes are returned to a reservoir and mixed. In other words, in this experimental study, the flow-electrodes operate in SCC mode. In each experiment, each of the flow-electrodes was divided equally between anodic and cathodic current collectors. The upper part of the flow-electrodes reservoir is the entry point of the two electrodes and the end of the reservoir is the exit point of the electrodes. Also, the feed water is pumped from the reservoir to the FCDI unit cell, and after passing through the flow channels, returns to the same water reservoir. A schematic layout diagram of flow-electrode capacitive deionization system in SCC mode is shown in Figure 4.

Therefore, the feed water entering the FCDI system operates similarly to the flow-electrodes in CC mode.

The closed cycle is repeated continuously for the feed water and the flow-electrodes to reduce the salt concentration of the feed water (electrical conductivity). In fact, the FCDI desalination system, which operates continuously in batch mode, can remove the salt ions from the feed water by means of the flow-electrodes until the desalination process reaches equilibrium (steady desalination). As the changes in feed water salt concentration indicate a successful desalination process, the efficiency



**Figure 4** | Schematic layout diagram of flow-electrode capacitive deionization system in short-circuited closed cycle operation mode.

of salt removal from the feed water can be calculated using Equation (1):

$$\text{SRE (\%)} = \frac{C_i - C_f}{C_i} * 100 \quad (1)$$

where  $C_i$  is the initial concentration of salt in the feed water and  $C_f$  is the final concentration of salt in the desalinated water.

Since the electrical conductivity of the feed water represents the concentration of salt in the feed water, the salt concentration of the desalinated water (water leaving the FCDI cell) was measured continuously using the conductivity meter (AZ 8603, Water Quality Meter, Taiwan). In addition, to supply the current collector plates with a cell voltage of 1.2 V in constant voltage mode (since voltages above 1.23 V lead to Faradaic reactions such as water hydrolysis (He *et al.* 2020)), we used GPS-3000 series constant voltage/current digital DC power supplies.

Pneumatic and silicon tubing (outer diameter of 4 mm and inner diameter of 2 mm) were used to establish a relationship between feed water and flow-electrodes reservoirs, peristaltic pumps, and FCDI cell.

## 2.2. Description of experiments

We conducted the FCDI experiments to investigate the effect of three operating parameters, namely flow rate of flow-electrodes, electrolyte salt concentration of flow-electrodes, and initial feed water concentration, on the efficiency of desalination operation. In all experiments, the flow-electrodes with a volume of approximately 250 ml and identical carbon mass loading (5 wt% activated carbon powder suspension electrode) were recirculated between the electrodes reservoir and the FCDI unit cell. Also, in all experiments, 100 ml of the feed water containing NaCl with the same constant flow rate of 20 ml/min was recirculated between the feed water reservoir and the FCDI cell. To investigate the effect of the three above-mentioned parameters on the desalination efficiency of the FCDI system, according to Table 4, we conducted six experiments and compared the results in the diagrams. The results of this comparison can be seen in section 3.

In all FCDI experiments performed in this study, we allowed the water and flow-electrodes to recirculate in the FCDI system for 15 minutes at the beginning of the work and after the FCDI system was turned on (the pumps of the feed water and flow-electrodes were turned on) and before the voltage was applied to the current collector plates. The circulation of water and flow-electrodes in the system slightly decreased the amount of salt ions in the water reservoir and reached a constant and unchanged concentration (equilibrium). This can be justified by two reasons:

1. At the end of each FCDI experiment, the system was washed with distilled water to prevent the activated carbon particles from accumulating in the system and, consequently, to avoid reducing the desalination efficiency of the system. Therefore, the distilled water remaining in the FCDI system influences the feed water concentration in the next experiment. In the experiments, the volume of distilled water in the system was estimated to be 18.4 ml.
2. The nature of the FCDI desalination process is defined mainly based on the electrosorption of ions on porous materials under an electric potential. Also, the efficiency of the desalination operation is influenced by the osmotic phenomenon caused by the difference in salt concentration between the flow-electrodes and the feed water (Jeon *et al.* 2013; Yang *et al.* 2016). However, the effect of chemical ion adsorption is negligible compared to desalination because of the voltage applied to the current collector plates.

To prevent clogging and deposition on the surface and in the channels of the current collector plates, a reverse voltage (polarity reversal) was applied to the current collector plates after removing the salt ions in the desired amount at the end of each experiment. This operation has two advantages: regeneration of the flow-electrodes, and improved performance of the current collector plates. By reversing the poles, feed water deionization is reversed and the ions removed from the feed water, return again to the feed water reservoir. Thus, the flow-electrodes, which previously adsorbed the salt ions, return the ions to water and are regenerated. At this stage, the activated carbon powder deposits that detach from the inner channel surface of the current collector plates return to the reservoir of the flow-electrodes.

In all six FCDI experiments, by applying a voltage of 1.2 V to the FCDI cell and starting the salt removal process in a fixed period of 5 hours, we achieved a certain amount of EC (NaCl) removal. Then, at the end of each experiment, we applied a reverse voltage (discharge at  $-1.2$  V) to the FCDI cell. We applied the reverse voltage in each experiment until the flow-electrodes returned all the salt adsorbed from the feed water back into the water channel and then into the water reservoir, and the salt concentration (EC) of the water in the reservoir reached the initial value. Thus, at the second stage of each experiment, the FCDI system was given time to recover all the NaCl ions removed at the first stage and to actually perform the

regeneration (neutralization) of the flow-electrodes. We repeated each of the six experiments four times and discarded the outlier data. In the FCDI experiments, laboratory errors (including the errors in the equipment used, measurement errors, laboratory conditions, and power fluctuations) can have a negative effect on the outcome of each experiment. Therefore, we tried to choose the most accurate and correct experiment to evaluate the efficiency of the FCDI system in desalination of the feed water.

To investigate the effect of flow rate of flow-electrodes, initial concentration of feed water, and electrolyte salt concentration of flow-electrodes on NaCl adsorption efficiency, the first to sixth experiments were compared in separate diagrams, as shown in Tables 1–3.

### 3. RESULTS AND DISCUSSION

In this section, we review, explain and compare the results of the FCDI experiments and perform the statistical and graphical analysis.

#### 3.1. Diagram and results of FCDI experiments conducted in this study

##### 3.1.1. Effect of flow rate of flow-electrodes on salt adsorption (removal) efficiency of FCDI system

To investigate the effect of flow rate of flow-electrodes on salt adsorption (removal) efficiency of FCDI system, we compared the results of three pairs of experiments. The results are shown in Figures 5–7.

According to Table 4, since the two experiments 1 and 2 differed only in the flow rate of flow-electrodes, and the results of these two experiments were almost the same, it can be concluded that the flow rate of flow-electrodes at low initial concentrations (1,000 mg NaCl/l) of feed water and 0.1 M NaCl concentration in the electrolyte had no significant effect on salt removal efficiency of the FCDI system.

**Table 1** | Effect of flow rate of flow-electrodes on salt adsorption (removal) efficiency of FCDI system

Test no.	Purpose of comparison
1 and 2	Effect of flow rate of flow-electrodes on salt adsorption efficiency in low feed water concentrations and low electrolyte salt concentration of flow-electrodes
3 and 4	Effect of flow rate of flow-electrodes on salt adsorption efficiency in high feed water concentrations and low electrolyte salt concentration of flow-electrodes
5 and 6	Effect of flow rate of flow-electrodes on salt adsorption efficiency in high feed water concentrations and high electrolyte salt concentration of flow-electrodes

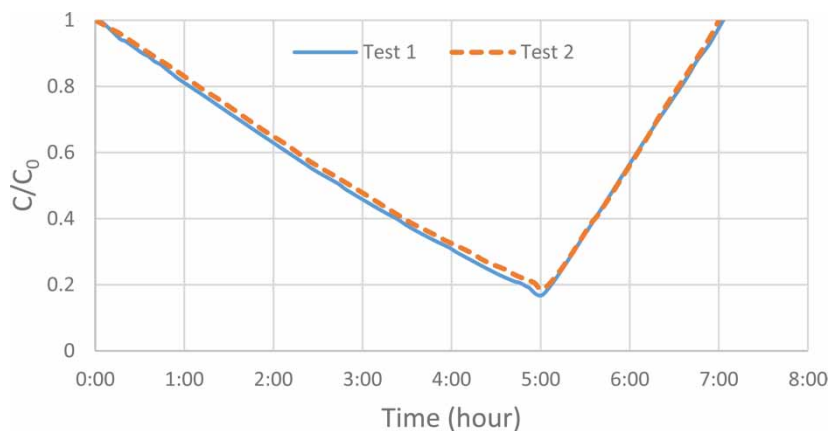
**Table 2** | Effect of initial sodium chloride concentration of feed water on salt adsorption (removal) efficiency of FCDI system

Test no.	Purpose of comparison
2 and 4	Effect of initial sodium chloride concentration of feed water on salt adsorption efficiency at low rates of flow-electrodes and low electrolyte salt concentration of flow-electrodes
1 and 3	Effect of initial sodium chloride concentration of feed water on salt adsorption efficiency at high rates of flow-electrodes and low electrolyte salt concentration of flow-electrodes

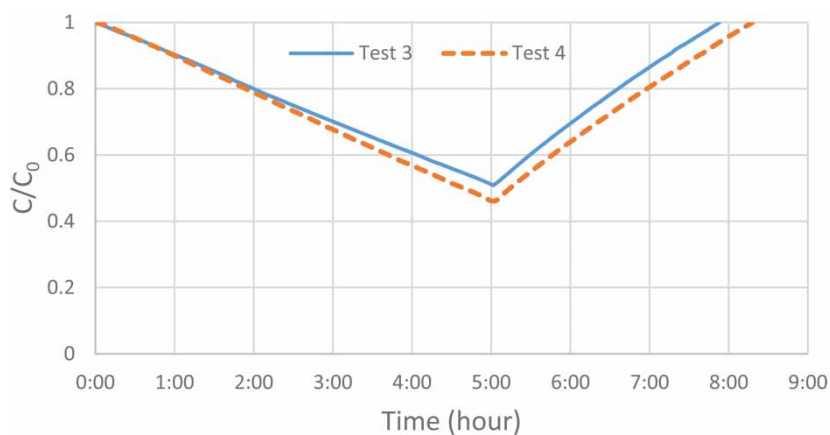
**Table 3** | Effect of salt concentration of electrolyte of flow-electrodes on salt adsorption (removal) efficiency of FCDI system

Test no.	Purpose of comparison
3 and 6	Effect of salt concentration of flow-electrodes on salt adsorption efficiency in high feed water concentrations and high flow rate of flow-electrodes
4 and 5	Effect of salt concentration of flow-electrodes on salt adsorption efficiency in high feed water concentrations and low flow rate of flow-electrodes

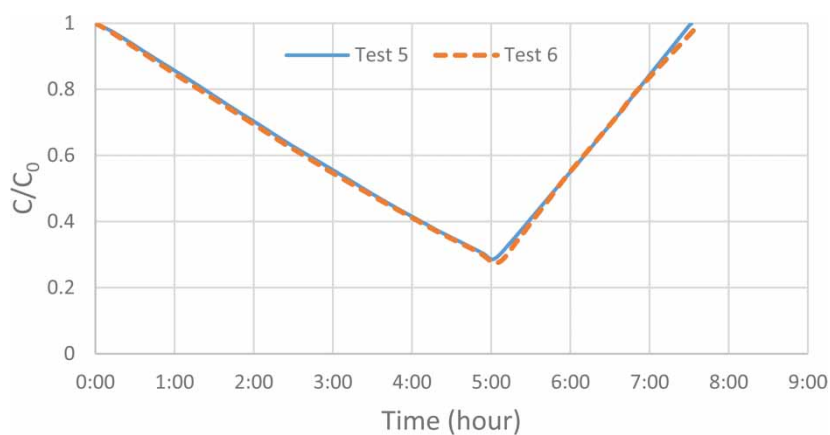




**Figure 5** | Result comparison of experiments 1 and 2.



**Figure 6** | Result comparison of experiments 3 and 4.



**Figure 7** | Result comparison of experiments 5 and 6.

By comparing the results of two experiments 3 and 4, we examined the effect of flow rate of flow-electrodes on the adsorption efficiency of NaCl at high initial concentrations (5,000 mg NaCl/l) of feed water and 0.1 M NaCl concentration in the electrolyte of flow-electrodes. In contrast to experiments 1 and 2 for the feed water with a low initial concentration

**Table 4** | Summary of specifications and results of FCDI experiments conducted in this experimental study

Test no.	Initial concentration of feed water (mg/l)	Initial electrical conductivity of influent saltwater ( $\text{mS cm}^{-1}$ )	Flow rate of flow-electrodes (ml/min)	Concentration of NaCl in electrolyte of flow-electrodes (M, molarity)	Salt removal efficiency	Salt adsorption capacity (SAC) ( $\text{mg/g}_{\text{dry carbon}}$ )	Duration of reverse voltage applied to FCDI system (hr)
1	1,000	2.09	80	0.1	83%	7.36	2
2	1,000	2.07	40	0.1	81%	7.2	2
3	5,000	8.01	80	0.1	49%	18.8	3
4	5,000	8.02	40	0.1	54%	20.64	3.25
5	5,000	8.02	40	0.3	71%	26.96	2.5
6	5,000	8.05	80	0.3	72%	29.12	3

(1,000 mg NaCl/l), the salt removal efficiency in experiments 3 and 4 showed a meaningful difference and the lower flow rate (40 ml/min) of the flow-electrodes resulted in a higher salt removal efficiency (54%).

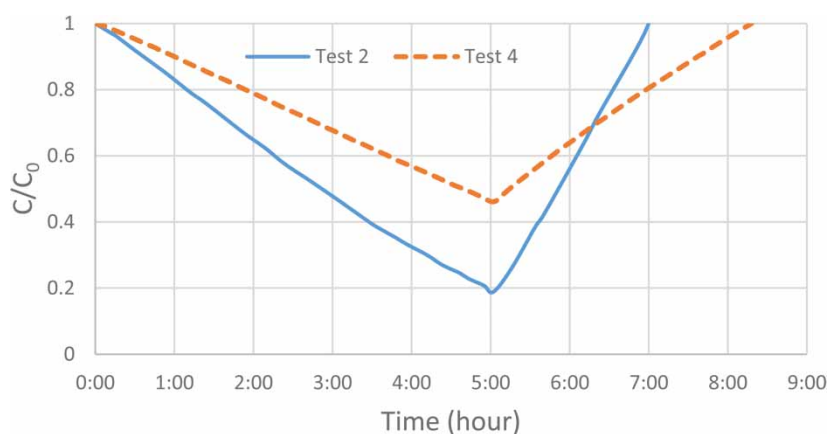
In FCDI cells, when the potential was applied to the current collector plates, ions move forward to the electrodes. The current in the FCDI cell is carried by ions. As it was mentioned, the main difference between these experiments (experiment pairs 1 and 2 and experiment pairs 3 and 4) was the initial salt concentration of feed water. Higher salt concentration results in more ions in solution for carrying the current and therefore higher current efficiency could be achieved. In this condition, the flow rate of flow-electrode, could play more important role. It means that more residence time for flow-electrodes (40 ml/min) in the FCDI system, results in a higher salt removal efficiency because much more ions carry the current. In other words, the ion concentration in feed water in experiments 1 and 2, plays a limiting role and the current efficiency is affected by feed conductivity.

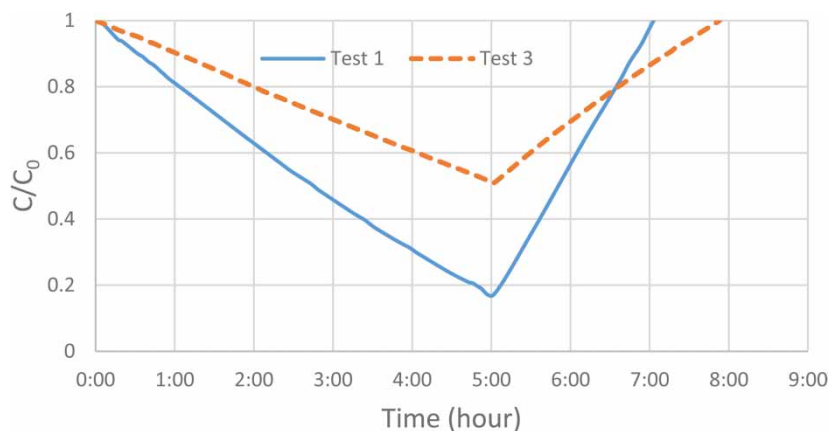
Considering the agreement between the results of both experiments 5 and 6, it can be concluded that although increasing the NaCl concentration in electrolyte of the flow-electrodes increases the salt removal efficiency of our FCDI system, the results did not differ at either the low or high flow rate of the flow-electrodes. This means that in the high concentrated electrolyte, the residence time has no significant effect on current efficiency and there is no need to decrease the flow rate of flow-electrodes for improving the performance of FCDI system.

### 3.1.2. Effect of initial sodium chloride concentration of feed water on salt adsorption (removal) efficiency of FCDI system

To investigate the effect of initial sodium chloride concentration of feed water on salt adsorption (removal) efficiency of FCDI system, we compared the results of two pairs of experiments. The results are shown in Figures 8 and 9.

According to Figure 8, the NaCl removal efficiency in experiment 2 (81%) was much higher than that in experiment 4 (54%), indicating the significant effect of the initial feed water concentration on the NaCl adsorption efficiency of the

**Figure 8** | Result comparison of experiments 2 and 4.



**Figure 9** | Result comparison of experiments 1 and 3.

FCDI system. In both experiments, the flow rate of the flow-electrodes was 40 ml/min and the concentration of NaCl in the electrolyte of the flow-electrodes was 0.1 M. Since the slope of the NaCl removal diagram in the first phase of experiment 2 is higher than in experiment 4, the diagram slope in the second phase of the FCDI experiment, which involves polarity reversal and electrodes regeneration, is higher in experiment 2 than in experiment 4. In the electrode regeneration phase, the flow-electrodes in experiment 2 were able to return the same amount of salt that they had adsorbed in the first phase in a shorter time. This is because the flow-electrodes adsorbed less salt in experiment 2 than in experiment 4 and then returned to water, and consequently consumed less ion adsorption capacity. In this way, the process of ion adsorption and electrode regeneration was represented in the diagram with a steeper slope.

In experiment 4 the feed water has much more salinity, although the amount of salt that is removed over a period of time is much higher, but the salt removal percentage is significantly lower than in experiment 2. According to Table 4, the amount of SAC in experiment 4 was approximately three times that in experiment 2, since the initial feed water concentration is five times more.

Comparing the experiments 1 and 3 (Figure 9), it is evident that the salt adsorption efficiency was higher (83%) in experiment 1 than in experiment 3 (49%). The amount of SAC was 7.36 mg/g dry carbon in experiment 1 and 18.8 mg/g dry carbon in experiment 3. The marked difference between the two values obtained is also due to the marked difference between the initial NaCl concentration in the feed water in experiments 1 and 3.

As mentioned before, according to the high level of salinity in the electrolyte of the flow-electrodes, higher feed water concentration resulted in higher current efficiency and therefore much more ions can move from feed water to the electrodes and higher SAC can be achieved.

To effectively remove salt from the FCDI system, the ions in the feed water must first pass through the ion-exchange membrane and enter the flow-electrodes. The ions are adsorbed in the microporous and mesoporous spaces of the activated carbon and finally removed from the FCDI system. A number of previous studies on the electrical adsorption of ions on the microporous spaces of carbon electrodes reported that the electrical adsorption increased as salt concentration in the flow-electrodes increased. This is due to the overlapping effect of the EDL on the microporous spaces and the reduced ionic resistance at higher salt concentrations.

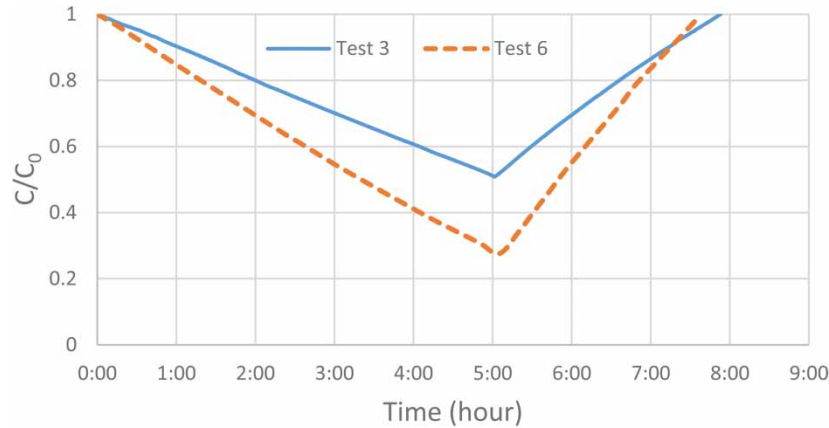
It was found that sodium chloride removal increased with increasing feed water concentration, resulting from stronger electrostatic interactions, higher concentration gradient, and less double-layer overlapping effect (Hou *et al.* 2013).

Therefore, EDL overlapping is a key factor affecting capacitive behavior and electrosorption capacity (Hou *et al.* 2006).

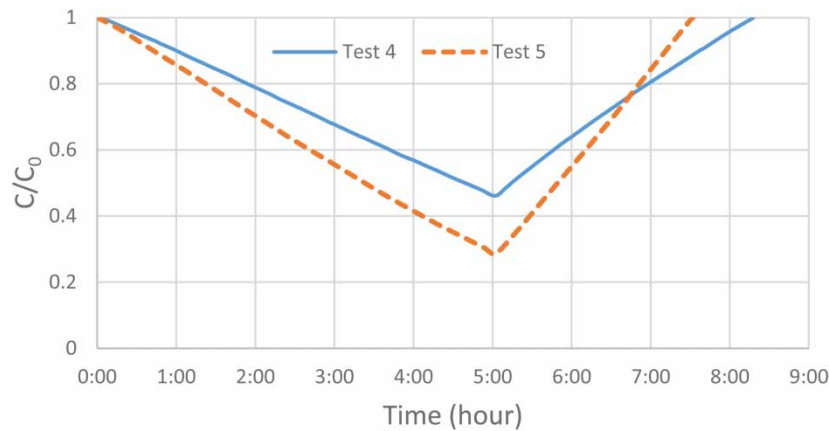
### 3.1.3. Effect of salt concentration of electrolyte of flow-electrodes on salt adsorption (removal) efficiency of FCDI system

Comparing the results of experiments 3 and 6 and experiments 4 and 5 in a diagram (Figures 10 and 11) illustrates the importance of the salt concentration in the electrolyte of the flow-electrodes (molarity) on the salt adsorption efficiency of the FCDI system.

As shown in Table 4, the initial concentration of the feed water (5,000 mg NaCl/l) and the flow rate of the flow-electrodes (40 or 80 ml/min) are the same in both pairs of experiments. The two experiments differed only in the salt concentration



**Figure 10** | Result comparison of experiments 3 and 6.



**Figure 11** | Result comparison of experiments 4 and 5.

(molarity) in the electrolyte of the flow-electrodes. These difference in the molarity of the electrolyte, made a significant difference in NaCl adsorption efficiency (about 20%) and the amount of SAC (more than 6 mg NaCl/gr<sub>dry carbon</sub>). In fact, the desalination efficiency of the FCDI system was influenced by both the electrical potential and the osmotic phenomenon caused by the difference in the salt concentration of the flow-electrode and the salt water. Higher difference results in higher salt removal and therefore higher SAC can be achieved.

#### 4. CONCLUSIONS

The aim of this study was to experimentally explore the performance of a pilot FCDI system at the laboratory scale and evaluate the effects of three parameters of flow rate of flow-electrodes, electrolyte salt concentration of flow-electrodes, and initial feed water concentration on the desalination efficiency. Six experiments were carried out on this FCDI system under closed-cycle continuous mode of operation. The salt concentration of feed water and the electrolyte of flow-electrodes plays an important role in the performance of FCDI systems, because of their impact on carrying the ions from feed water to the electrodes and providing enough current efficiency for salt removal. The flow rate of the flow electrode did not make a considerable change in the salt removal efficiency and its impact was only observed when the initial concentration of the feed water and the salt concentration of the electrolyte (in the flow-electrodes) has a meaningful difference. At the mentioned conditions, the low flow rate of the flow-electrodes slightly enhanced the salt removal efficiency.

The highest salt removal efficiency (83%) was observed in the experiment involving a 5 wt% suspension electrode with the flow rate of 80 ml/min, low salt concentration (0.1 M NaCl) in the electrolyte of flow-electrode, and 100 ml feed water with an initial concentration of 1,000 mg NaCl/l and flow rate of 20 ml/min in 5 hours. The highest SAC (29.12 mg/g<sub>dry carbon</sub>) was

also achieved in the experiment which involved 5 wt% suspension electrode with the flow rate of 80 ml/min, high NaCl concentration (0.3 M NaCl) of the electrolyte of flow-electrode, and 100 ml feed water with an initial concentration of 5,000 mg NaCl/l and flow rate of 20 ml/min in 5 hours. Regarding the reversibility of all the experiments, Faradic reaction could be ignored.

## DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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