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New insight into the performance analysis of flow-electrode capacitive deionization by varying the operation voltage

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KEYWORDS Voltage; Water desalination; Capacitive deionization; Flow electrode. Abstract. In the flow-electrode capacitive deionization (FCDI), the highly activated porous carbon electrodes of the slurry phase flows through the channels of current collectors adsorbs the salt ions when a voltage is applied. In this study, the effect of voltage on the performance of a FCDI cell is experimentally investigated. The voltage is applied on the top corner of a FCDI cell (V_{apply}) and simultaneously the voltage of the central cell (V_{cell}) is measured. The experiments were conducted by applying voltages from 0.6 to 3.9 V. The experimental results show that the difference between V_{apply} and V_{cell} is a function of salt concentrations of the feed water. The higher voltages $(V_{apply} > 1.2V)$ can be used for increasing the salt removal efficiency (E) for higher salt water concentrations without electrolysis. Also, the results show that E increases along with the applied voltage. A series of pH measurements were done for investigating the electrolysis point of the setup.

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1. Introduction

Capacitive desalination (CDI) is one of the most promising water desalination processes for brackish water of medium salinity [1]. CDI is an electrochemical water treatment technology based on the formation of an Electrical Double Layer (EDL) on the surface of porous electrode materials under an electrical field. CDI technology has increasingly received attention due to its small size, low energy usage, and low cost [2–16]. Many progresses have been made in

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capacitive deionization such as the development of carbon materials and their usage in this field [17–21], and the introduction of a new-family of metal-organic frameworks electrodes for capacitive deionization [22-26]. Flow-electrode capacitive desalination (FCDI) is an efficacious version of CDI for desalination of water with high salinity such as sea water [27]. FCDI technology is based on the principle of electric doublelayer capacitor [28,29] and it has recently come to be a serious subject in some studies, due to its easy scaleup for mass desalination. Unlike to CDI, FCDI can work in a continuous cycle by omitting the discharge step [30]. Coupling of neutralization deionization with flow electrodes (FCND) was investigated by Xu et al. They compared the performance of FCND and FCDI [31].

Slurry Activated Carbon (AC) in FCDI is used instead of the fixed electrode in a conventional CDI.

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The suspended AC adsorbs the ions when an external electric potential is applied to the current collector. The adsorbed ions are carried with the flow of electrodes. The cation and anion exchange membranes just allow one special ion to pass through membrane which is positive or negative. So, the FCDI cell generally separates the ions in the feed water [32-34]. The direct electrosorption of ions in salt water on porous electrodes in the CDI leads to a low operating voltage of CDI of about 1.2 V [2,3]. The heart of this technology is electric potential. Recently, some researchers investigated the applied voltage to FCDI cell [35]. Their study is focused on voltages greater than 1.23 V-the minimum voltage for water electrolysis-to enhance the driving force for ion migration and adsorption in an FCDI so as to achieve higher desalination rates. They have shown that when the voltage was raised from 0.6to 4.8 V, the desalination rate increased by nearly 7 times, but the charge efficiency reduced from 92% to 69.5% due to the occurrence of Faradaic reactions such as water electrolysis [35].

Jeon et al. have introduced FCDI [27], and some aspects of FCDI have been investigated by few researchers [18-19,24-25]. Recently, the authors have experimentally studied the structure of spacer in a FCDI [36]. In another study, the coupling of FCDI and solar energy was also investigated [37]. In the present study, the effects of voltage on the performance of FCDI cell are investigated. In this regard, at various applied voltage to the cell, the central cell voltage is measured and the maximum allowed voltage before electrolysis is determined.

2. Experiment

An FCDI unit cell is composed of a pair of graphite current collectors (Figure 1), cation- or anion-exchange membranes, a gasket, a spacer, and one pair of end plates [27–29]. The width, length, and height of the graphite current collectors are 110 mm, 66 mm, and 12 mm, respectively. The dimensions of the



Figure 1. Carved electrode channel on the current collector.

flow channel carved in the collectors are 2×2 mm. The column length of the flow channel is 30 mm, and the number of columns is 23. The thickness of the ion-exchange membranes is approximately 160 μ m (Neosepta CMX and AMX, Tokuyama, Japan).

The contact area between the ion-exchange membranes and flow-electrode is 12.7 cm^2 . A silicone gasket and a 0.3 mm thick polyester spacer are used between the cation- and anion-exchange membranes for assembling the FCDI unit cell. All parts are held together using polyvinyl chloride (PVC) end plates.

Figure 2 shows the assembly of an FCDI setup. The salt water is processed under open-cycle conditions with two reservoirs, i.e. salt water and desalinated water reservoirs. The flow rate of the flow-electrode, operated in a closed cycle, was maintained constant at 21.5 mL min⁻¹. The flow-electrode runs along the flow path on the current collector. As shown in Figure 2, the used cathode and anode flow-electrodes are assembled and terminated into the same flow-electrode reservoir. As shown in previous studies, this operation method originated from the automatic release of ions electrostatically adsorbed on the surface of AC after desalting by mixing and neutralization of the charged cathode and anode flow-electrodes [38-39].

Each desalting experiment was performed for 30 A time interval between two consecutive minutes. experiments was introduced for attaining the initial electrical conductivity of the effluent after cutting out the applied voltage. Each desalting experiment was repeated for three times. The voltage was applied to the FCDI unit cell using a power supply (E3630A, Triple output DC power supply, HEWLETT PACKARD, Republic of Korea) for desalting experiments. The voltage at the center of the FCDI cell, V_{cell} , was measured by a multimeter (Fluke, 170 digital multimeter). The salt concentration in the output of salt water was obtained from its measured electrical conductivity, using a conductivity meter (F-74, LAQUA, HORIBA scientific, Japan).

The flow-electrode, which comprised a slurry of activated carbon, was made from a homogeneous dispersion of commercial AC (Maxsorb MSC-30, Kansai Coke & Chemicals Co. Ltd, Japan) in a 0.1 M aqueous solution of NaCl. The mass ratio of AC per distilled water was 1:10 [39]. The mixtures were stirred using a magnetic bar for 24 hours for achieving homogeneous carbon suspension. The AC used in the experiments was the same as in previous investigations [27,39–40].

3. Results and discussion

In this study, the effects of electric potential on the salt removal efficiency were investigated. The results were obtained for several volume flow rates of salt water (FR_w) . Salt removal efficiency is defined as the



Figure 2. Experimental setup.

following:

$$E = \frac{C_{in} - C_{out}}{C_{in}} \times 100, \tag{1}$$

where E is the salt removal efficiency in %, and C_{in} and C_{out} are the concentration (g/L) of inlet and outlet of salt water, respectively.

The thickness of the polyester spacer was 0.3 mm and its porosity was 0.56. The concentration of the inlet salt water, C_{in} , was 35 g/L, which is the same as sea water. The volume flow rate of electrode (FR_e) was kept constant at 21.5 mL/min. The amounts of applied voltage (V_{apply}) , measured cell voltage (V_{cell}) , and salt removal efficiency for flow rates of $FR_w = 2, 2.5, 3$, and 5 mL/min are shown in Tables 1 to 4, respectively.

From Tables 1 to 4 it can be seen that the salt removal efficiency increases with increasing the voltage. For example, in the case of $FR_w = 2 \text{ mL/min}$ and $V_{apply} = 3.89 \text{ V}$, the efficiency reaches 78%. This value is quite high for this size of a FCDI cell with a small contact surface area and continuous mode (or open

1.21.611.75 $\mathbf{2}$ 2.643.9 $V_{a\,pply}$ (V) 1 1.5 ΔV (V) 0.270.3050.350.410.450.50.640.9 E%2023.73132.435.841.35778

Table 1. Effect of voltage on the salt removal efficiency, $FR_w = 2 \text{ mL/min}$

Table 2. Effect	of volt	age on	the salt	removal	efficien	cy, FR_v	v = 2.5	mL/min.
V_{apply} (V)	1	1.2	1.5	1.6	1.74	2	2 .64	3.8
$\Delta V (\mathrm{V})$	0.26	0.3	0.37	0.4	0.44	0.5	0.64	0.8
E%	15	18.4	23.5	23.1	27.5	32.1	44.3	65.5

Table 3	. Effect	of voltage	on the salt	removal	efficiency,	$FR_w =$	= 3 mL	/min
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V_{apply} (V)	1	1.2	1.5	1.6	1.73	2	2.6	3.9
$\Delta V ({ m V})$	0.25	0.3	0.37	0.4	0.43	0.48	0.6	0.9
E%	11.9	14.4	18.5	19.7	21.5	25.6	35	47

Table 4. Effect of voltage on the salt removal efficiency, $FR_w = 5 \text{ mL/min.}$

V_{apply} (V)	1	1.2	1.6	1.74	2	2.69	3.8	
$\Delta V \; ({ m V})$	0.24	0.3	0.4	0.43	0.5	0.69	1	
<i>E</i> %	8.6	10.1	13.3	14.5	16.7	23.4	28.7	



Figure 3. E and normalized pH in terms of the applied voltage.

mode). The value of applied voltage differed from the measured voltage of the cell due to the electric resistance of the cell. In the following, the relation between $\Delta V = V_{apply} V_{cell}$ and salt concentration of feed water is investigated. The results showed that the effect of FR_w on ΔV was negligible for $FR_w > 1$ mL/min. The highest voltage in each table corresponds to the condition that bubbles were observed in the output tube of the FCDI set.

The value of pH in the outlet salt water was also measured. The water flow rate and concentration of feed water were fixed at $FR_w = 2 \text{ mL/min}$ and $C_{in} = 35$ g/L, respectively. The porosity of the spacer was 0.44 (different from the previous part). Figure 3 shows the values of salt removal efficiency and pH in terms of an applied voltage. The values of pH are normalized by dividing it on the initial pH about 6.2 (to display in a single chart with E simultaneously). It must be noted that in this figure, E (0 < E < 1)is used instead of E%. Figure 3 shows that the normalized pH was approximately uniform at different applied voltages lower than 1.7 $V(V_{cell} = 1.35 \text{ V})$ but it decreased suddenly at applied voltages higher than 1.8 V ($V_{cell} = 1.42$ V). It means that some reactions occurred at applied voltages higher than 1.7 V, resulting in a decrease of PH.

The pH of the desalted water remained constant at low voltages, which is an indication of symmetric removal of anion and cation from the salt water. However, at higher voltages, not only water electrolysis in salt solution, but also redox reactions such as oxidization over carbon electrode (e.g. carbon oxidation, chloride oxidation) could occur. These faradaic effects could cause a significant pH change in the



Figure 4. $\Delta V = V_{apply} - V_{cell}$ for different inlet salt concentrations (C_{in}) at various applied voltages.

system. Therefore, one may conclude that increasing the applied voltage leads to an increase in the salt removal efficiency but decreases the quality of water (as indicated by its pH). It must be noted that a decrease in pH is a result of an increase in ionic conductivity which means that the desalting efficiency may be slightly overestimated for high applied voltages. It is postulated that the difference between the applied voltage and cell voltage can be related to the salt concentration in the feed water.

To confirm such a relation, a series of salt removal experiments were performed using feed water with different salt concentrations. Three different voltages were applied: 1, 1.2, and 1.5 V. After perfect desalting of feed water in the same FCDI cell (when the conductivity meter showed a constant value), V_{cell} was measured. Figure 4 shows ΔV for different inlet salt concentrations (C_{in}) at various applied voltages. From Figure 4, for $C_{in} = 1$ g/L, ΔV is about zero. The results showed that ΔV increased with increasing the concentration of salt in the feed water. The voltage difference increased until a certain concentration was reached and then gradually became constant. The increased applied voltage contributed to the increase in the voltage difference, as well. From Figure 4, it can be observed that voltage drop relates to the salt concentration because electric current is larger in high salt concentration. Kirchhoff law relates the applied voltage and current as:

$$V_{cell} = V_{apply} - r_{cell} \times i, \tag{2}$$

where *i* is the current (A), and r_{cell} is the cell and external resistance. For $V_{apply} = 1.2$ V and salt concentration of more than 25 g/L, r_{cell} was about 0.56 Ω as measured by the Ohmmeter.

Another important result obtained from Figure 4 is that when a batch mode, i.e. a closed loop for salt water, is performed for sea water desalination by the FCDI, a higher applied voltage can be used for increasing the salt removal efficiency but as the salt concentration decreases it is necessary to decrease the applied voltage to avoid electrolysis reaction because for low salt concentrations, V_{apply} is close to V_{cell} . For example in Figure 4, the applied voltage must be gradually decreased from 1.5 V to 1.2 V along with decreasing the salt concentration to less than 20 g/Lit. For salt concentrations less than 10 g/L, voltages lower than 1.2 V are allowed to be applied.

4. Conclusions

The effects of electric potential in a Flow-electrode Capacitive Deionization (FCDI) set are experimentally investigated. The experiments were performed at different applied voltages from 0.6 to 3.9 V. The results show that:

- Salt removal efficiency increases by increasing the voltage;
- The difference between the applied voltage and the cell voltage, $\Delta V = V_{apply} V_{cell}$, varies as the salt concentrations of the feed water vary;
- According to the experimental results, ΔV increases by increasing the inlet salt concentration (C_{in}) at different V_{apply} up to C_{in} = 25 g/L after which it nearly attains a constant value;
- To investigate the electrolysis threshold of the set, PH measurements were performed. The optimum point was $V_{apply} = 1.7$ V ($V_{cell} = 1.35$ V) where at higher voltage (> 3.5 V) bubbles are observed due to electrolysis;
- Higher voltages $(V_{apply} > 1.2 \text{ V})$ can be used for the water with high salt content (e.g. sea water) leading to increased salt removal efficiency but for salt concentrations less than 10 g/Lit, voltages less than 1.2 V are allowed to be applied to avoid electrolysis reaction.

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KoYeon Choo started studies regarding to Ni-MH battery, hydrogen storage, and purification using hydrogen storage alloys. She proceeded to a PhD degree by study of the electrochemical characteristics of capacitive flow-electrode cell in a static state. Currently, she is performing various researches in the fields of extraction, distillation, CDI, and redox flow battery.

Ali Akbar Alemrajabi was employed as a Teaching Assistant at the Isfahan University of Technology (IUT) in 1978. Later he received his MSc and PhD in Mechanical Engineering from the University of Birmingham and continued his collaboration with the IUT. He is still working with the IUT as an Emeritus Professor since he retired in 2017. He has been supervisor of more than 70 MSc and PhD research projects. His research interests include the application of renewable energies especially solar energy.