

Yasamin Salamat

Mem. ASME
Department of Mechanical
and Industrial Engineering,
Northeastern University,
334 Snell Engineering Center,
360 Huntington Avenue,
Boston, MA 02115
e-mail: s.salamat@neu.edu

Carlos A. Rios Perez

Mem. ASME
Department of Mechanical
and Industrial Engineering,
Northeastern University,
334 Snell Engineering Center,
360 Huntington Ave,
Boston, MA 02115
e-mail: carlos.a.riosperez@gmail.com

Carlos Hidrovo¹

Mem. ASME
Department of Mechanical
and Industrial Engineering,
Northeastern University,
207 Snell Engineering Center,
360 Huntington Avenue,
Boston, MA 02115
e-mail: hidrovo@neu.edu

Performance Improvement of Capacitive Deionization for Water Desalination Using a Multistep Buffered Approach

Due to the increasing demand for clean and potable water stemming from population growth and exacerbated by the scarcity of fresh water resources, more attention has been drawn to innovative methods for water desalination. Capacitive deionization (CDI) is a low maintenance and energy efficient technique for desalinating brackish water, which employs an electrical field to adsorb ions into a high-porous media. After the saturation of the porous electrodes, their adsorption capacity can be restored through a regeneration process. Herein, based on a physical model previously developed, we conjecture that for a given amount of time and volume of water, multiple desalination cycles in a high flow rate regime will outperform desalinating in a single cycle at a low flow rate. Moreover, splitting a CDI unit into two subunits, with the same total length, will lead to higher desalination. Based on these premises, we introduce a new approach aimed at enhancing the overall performance of CDI. An array of CDI cells are sequentially connected to each other with intermediate solutions placed in between them. Desalination tests were conducted to compare the performance of the proposed system, consisting of two CDI units and one intermediate solution buffer, with a two-cascaded-CDI unit system with no intermediate solution. Experimental data demonstrated the improved performance of the buffered system over the nonbuffered system, in terms of desalination percentage and energy consumption. The new proposed method can lead to lower amount of energy consumed per unit volume of the desalinated water. [DOI: 10.1115/1.4035067]

Introduction

Around 2.3 billion people are suffering from clean and fresh water scarcity around the globe, which is expected to increase up to 3.5 billion individuals by the year of 2025 [1,2]. This, along with the fact that over 97% of the earth's water is either brackish water or seawater, has drawn attention to efficient methods to convert aforementioned sources of water into drinkable and portable water [3–8].

Capacitive deionization is a relatively new and novel desalination technique where an electric field is applied to separate ions from a solution stream by adsorbing them onto a high surface area material [4,9]. In CDI, brackish water is pumped through an electrical field using a relatively low DC voltage. Charged contaminants are then pulled toward electrically conductive porous electrodes; positive ions toward the negative electrode, and negative ions toward the positive electrode. Extraction of these ions lowers the output solution concentration until saturation of the electrodes occurs. Electrodesorption of counterions in the electrical double layer (EDL) during desalination stores electrical energy. This energy can be recovered during the porous material regeneration process. Regeneration can happen by short-circuiting or applying a reverse electrical voltage. During the regeneration, the electrodes create an electric current through the discharge of stored ions resulting in recovering part of the energy stored during ion adsorption. The uniqueness of CDI as a desalination method relies on this capability to recover part of the input energy through the regeneration process, making CDI energetically efficient and extremely attractive. Because of its capability to partially recover the input energy, a CDI system would require a significantly low

energy input compared to conventional brackish water desalination methods, such as reverse osmosis [9].

Ultimate performance of CDI depends on various factors, ranging from chemical and physical properties of the porous media, to the solution flow rate and the applied electrical field [10–15]. As it was studied by Mossad and Zou [12] and Dermentziz and Ouzounis [15], increasing the flow rate lessens the desalination performance in a CDI system, due to the fact that in high flow rate regimes, the advection of the ions in the CDI unit surpasses the diffusion toward the porous media. It may seem axiomatic to presume the preference of low flow rates for CDI operation. However, investigating the convective–diffusive regimes in CDI units may not completely advocate this conclusion.

Theory

Perez et al. developed a physical model analyzing the electroadsorption mechanism for low salinity water in CDI [16]. Advection of the ions in the direction of the flow is in a continuous competition with their diffusion toward the porous media. The sublayer at which the transport of the ions to the porous electrode is more dominant is called the convective–diffusive layer. Analogous to viscous layer in a channel, two regimes of developing and fully developed convective–diffusive layer are defined in the CDI unit.

At low flow rates, the fully developed regime is obtained. Under this condition, the diffusion of the ions toward their oppositely charged electrodes is substantial and also consistent through the whole channel. At high flow rates, on the other hand, due to the significance of ions advection within the CDI unit, the transport of ions toward the electrodes is restrained only to the developing convective–diffusive layer. Figure 1 depicts convective–diffusive layers in a CDI unit. The ionic concentration for the aforementioned regimes can be seen in this figure.

Nernst Planck equation for dilute solutions is usually exploited in studying the mass transport in this layer [16–21]. Under the

¹Corresponding author.

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Nernst–Planck approximation, the net adsorption flux at this layer is defined as [22]

$$j(x) = D_{\text{eff}} \frac{C_o - C_w}{\delta_D(x)} \quad (1)$$

where j is the net molar flux of ions from the bulk solution to the porous media ($\text{mol}/\text{m}^2 \text{ s}$), D_{eff} is the effective diffusive coefficient, which includes both diffusion and electric potential driven motion (m^2/s), C_o is the inlet concentration (mol/m^3), C_w is the concentration at the wall (at the solution–electrode interface) (mol/m^3), δ_D is the convective–diffusive layer thickness (m), and x is the distance from the inlet (m). Based on Eq. (1), the adsorption velocity in a convective–diffusive layer can be calculated as

$$v_{\text{ads}}(x) = \frac{D_{\text{eff}}}{\delta_D(x)} \quad (2)$$

As it can be seen from Fig. 1, half of the channel’s depth can be considered as the convective–diffusive layer thickness, δ_D , in a fully developed regime, resulting in a constant adsorption velocity through the CDI unit. On the other hand, in a developing convective–diffusive regime, the adsorption velocity changes within the unit as the convective–diffusive layer thickness grows through the length of the unit. Since δ_D happens to be lower in a developing regime, the adsorption velocity will be higher, resulting in higher ions adsorption rate, comparing to a fully developed system. Hence, despite the dominance of ion diffusion over advection at low flow rates, under engineered circumstances, high flow rate regimes can lead to better desalination performance, owing to higher adsorption rate in the developing convective–diffusion regime.

For a specified amount of time, instead of desalinating at a low flow rate, desalination can occur in multiple consecutive cycles at a higher flow rate in such a way that the same amount of water is purified. Thanks to the higher adsorption velocity in the developing convective–diffusive regime happening at high flow rates, better performance can be obtained. This was verified by the numerical model developed by Perez et al. [16].

As it is depicted in Fig. 2, for a given CDI unit geometry and inlet concentration, desalinating for 4000 s in a fully developed regime with the low flow rate of 1 ml/min results in a minimum average outlet concentration equal to 81.5% of the initial concentration. However, running four 1000 s desalination cycles, each having the initial concentration equal to the minimum average concentration of the previous cycle, in a developing regime with

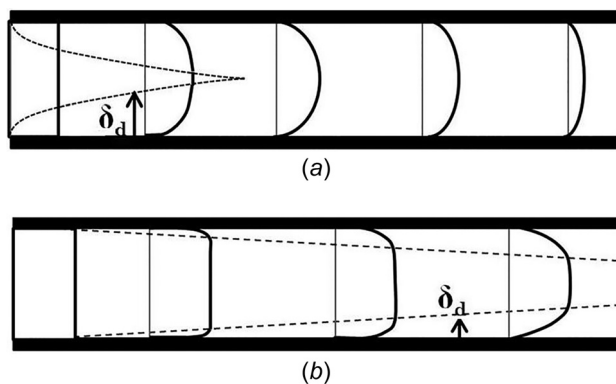


Fig. 1 Schematic of convective–diffusive layers in CDI, and illustration of ionic concentration in two regimes. Fully developed convective–diffusive layer: (a) at low flow rates, diffusion of ions toward the porous walls is more substantial than their advection within the channel. Developing convective–diffusive layer: (b) at high flow rates, due to the dominance of ions advection, the diffusion of the ions toward the porous media is limited to the convective–diffusive layer.

the higher flow rate of 4 ml/min, will lead to a minimum average outlet concentration equal to 52% of the initial concentration. The plots present the normalized outlet concentration up to the point that minimum average concentration is reached.

According to Eq. (2), in developing regime, the adsorption velocity decreases along the cell. Therefore, in order to get the full benefit of the high concentration gradient and adsorption rate at the beginning of the convective–diffusive layer, a CDI unit with the length of L can be split into two CDI units, each having the length of $L/2$. Hereby, higher adsorption flux of ions toward the porous media can be achieved with the same total length. Validated by the physical model, Fig. 3 shows the positive effect of splitting the CDI cell on the overall performance of the desalination system in a developing convective–diffusive regime at the flow rate of 5 ml/min. For this numerical analysis, the length of L was set to 150 mm. Again, the normalized outlet concentration in both systems is plotted until the minimum average concentration is achieved.

However, as the electrical voltage is applied on each cell, in a desalination system consisting of multiple CDI units consecutively connected to each other, desalination takes place, and, consequently, the outlet concentration of each cell will decrease. Hence, all the CDI units, besides the first one, will experience a variable initial concentration at the inlet. In an attempt to provide a constant inlet concentration for all the CDI cells, we propose the buffer system. The proposed architecture consists of an array of CDI units connected in series with solution buffers in between

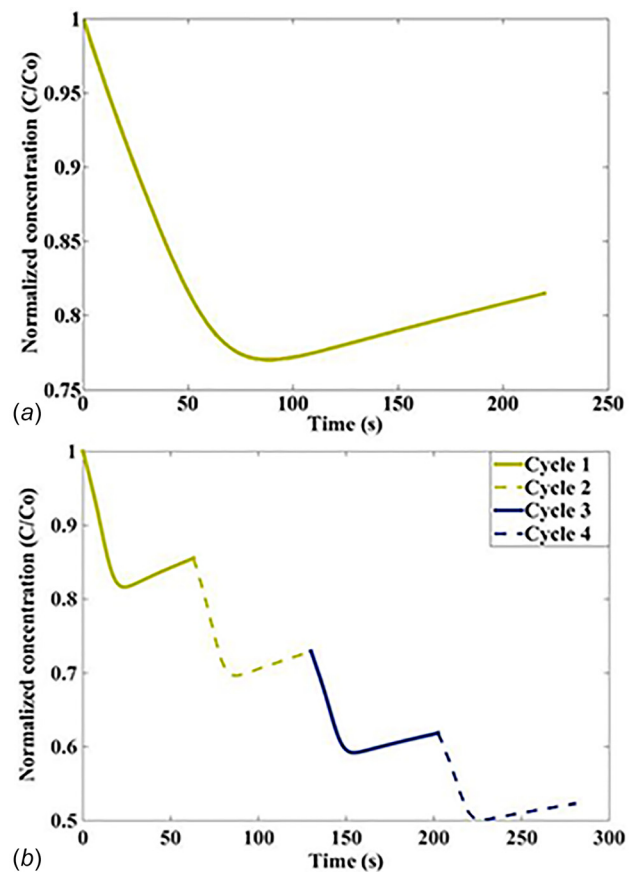


Fig. 2 Normalized outlet concentration over time for two desalination systems, with the same initial concentration. (a) One cycle of desalination in the fully developed convective–diffusive regime with low flow rate of 1 ml/min. (b) Four consecutive cycles of desalination in the developing convective–diffusive regime at high flow rate of 4 ml/min. Each cycle has the inlet concentration equal to the minimum average concentration of the previous cycle (except for the first cycle).

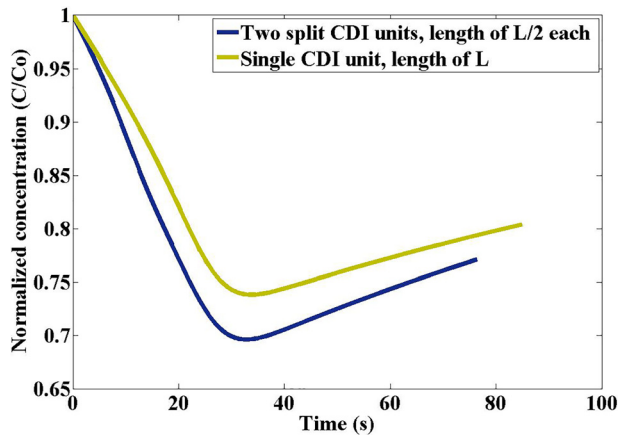


Fig. 3 Normalized outlet concentration over time for two architectures, with the same flow rate and initial concentration. These numerical results demonstrate effective impact of splitting the CDI cell on the ultimate desalination performance of the system.

them. Each buffer between two cells homogenizes the outlet solution of the preceding cell and provides a continuous reservoir for the following cell.

Herein, proof-of-concept of the proposed arrangement was accomplished by desalination of each unit at the time, with different initial concentrations. For this paper, the performance of the proposed system with two CDI units and one solution buffer in between versus a two-cascaded-CDI unit system without solution buffer was evaluated. Figure 4 illustrates the desalination performance of the proposed buffered system versus the two-cascaded-CDI unit system up to their minimum average concentration, obtained from the numerical model developed by Perez et al. For this set of simulations, the initial concentration and the flow rate for both systems were assigned as 0.05 mg/ml and 5 ml/min, respectively, and each porous electrode had the length of 75 mm. In the cascaded system, CDI units are consecutively connected to each other with no intermediate solution. For the proposed buffered system, first a desalination test is performed with a single CDI unit. Afterward, the CDI unit goes under another desalination test with the initial concentration equal to the minimum average concentration of the previous test.

Although, the cascaded system reached a lower outlet concentration, the buffered system had a lower minimum average concentration; 24% of the initial concentration, comparing to 22% of the initial concentration for the cascaded system. This also can be inferred from the normalized concentration plot for the buffered system that happens to have a higher minimum but higher surface area, which is correlated to lower minimum average concentration. Obtaining a lower minimum concentration may seem to imply a better performance. However, since CDI is an unsteady process, having a lower average concentration at the outlet is a better indication of a superior desalination system.

Experimental tests were also conducted to evaluate the desalination performance of the proposed buffered CDI system versus the cascaded system.

Methodology

Experimental Setup. All the experiments were executed in Multiscale Thermal Fluids Laboratory, at the Northeastern University, Boston, MA. For the sake of consistency, one CDI cell was employed for all the tests. The CDI casing consisted of one pair of 300 mm × 75 mm acrylic plates. The main flow channel in the cell had dimensions of 267 mm × 25.4 mm. Highly conductive and corrosion resistant titanium sheets served as the current collectors and 75 mm × 20 mm × 0.250 mm activated carbon sheets

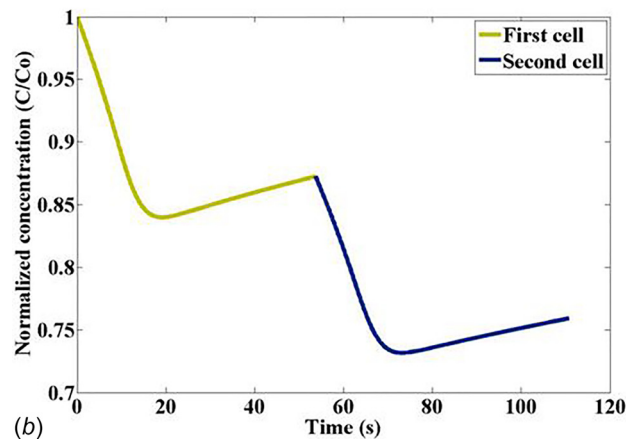
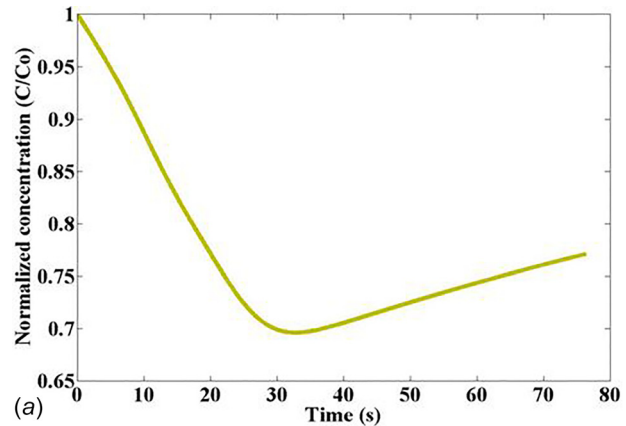


Fig. 4 Normalized concentration over time for the two-cascaded-CDI unit system (a) and the proposed buffered system (b). In the cascaded system, two CDI units are consecutively connected to each other, with no intermediate solution reservoirs. In the buffered system, first a CDI unit performs a desalination test, and then, the obtained minimum average concentration is assigned as the initial concentration of the second desalination test.

(Material Methods LLC, Irvine, CA) were utilized as the porous electrodes for the purpose of ions adsorption. In order to prevent any electrical contact between the electrodes, three 0.19 mm thick polyester meshes were placed between them, providing the total distance of 0.57 mm between the electrodes. Figure 5 depicts an exploded view of the CDI cell used in the experiments.

A Harvard apparatus PHD-2000 syringe pump drove the solution through the CDI unit. An LS32-1500 Sensirion flow meter and an ET908 eDAQ Flow-Thru Conductivity Electrode were, respectively, used to measure the flow rate and outlet conductivity. An Agilent E3647A power supply was utilized to apply the constant voltage and measure the electrical current consumed by the system, as well.

Experimental Procedure. A 0.05 mg/ml NaCl solution with the flow rate of 5.0 ml/min was used to evaluate the desalination performance of buffered and cascaded systems. Constant voltage of 1 V was applied for all the tests. Setting the voltage below the minimum required voltage for water electrolysis at 298 K (1.23 V) prevents electrode decomposition of water molecules during the desalination [23–26]. Proof-of-concept for the buffered system was implemented by evaluating the overall performance of two desalination experiments with two different initial concentrations. First, a steady long-term desalination test was conducted with the initial concentration of 0.05 mg/ml, with one pair of electrodes. Afterward, another desalination test was performed with the same

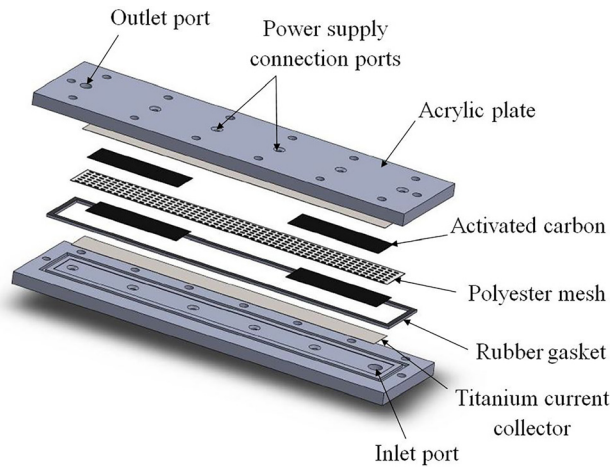


Fig. 5 Exploded view of the CDI unit. Titanium sheets serve as current collectors. Activated carbon pairs are separated by polyester mesh, and the rubber gasket is placed in the rectangular groove to seal the unit. The inlet and outlet fittings and power supply contact screws are not shown in this figure.

CDI unit but with a new pair of electrodes, at the same flow rate. The minimum average concentration obtained at the outlet of the first test was set as the initial concentration of the second desalination experiment (the buffer concentration). Average outlet concentration at each instant can be calculated as

$$C_{ave} = \frac{1}{\tau} \int_0^{\tau} C(t) dt \quad (3)$$

where C is the outlet concentration (mg/ml), and τ is the time at which the measurement takes place (s). For the cascaded system, two pairs of electrodes and current collectors, with 35 mm space in between, were used in the CDI unit. As it was mentioned before, the inlet concentration and the solution flow rate were 0.05 mg/ml and 5 ml/min, respectively. Figure 6 depicts the schematic of the experimental setup for both desalination systems.

In order to calculate the volume of the water treated and amount of energy consumed during the desalination process, the time at which the minimum average concentration was obtained was assigned as the operation time. The aforementioned values were then computed as below

$$V_w = \frac{\tau \cdot Q}{60} \quad (4)$$

$$E = \int_0^{\tau} V \cdot I \cdot dt \quad (5)$$

where V_w is the volume of the desalinated water (ml), Q is the flow rate (ml/min), E is the input energy of the system (J), V is the applied electrical voltage, and I is the electrical current. The desalination percentage was calculated as

$$p = \frac{C_o - C_{ave,min}}{C_o} \times 100 \quad (6)$$

where C_o and $C_{ave,min}$ are the initial and minimum average concentration of the outlet, respectively.

Results and Discussion

Figure 7 presents the normalized outlet concentration changes of the buffered and cascaded systems over time before reaching the minimum average outlet concentration. The results obtained from the experimental data are also shown in Table 1. All the

tabulated values are the recorded data up to the minimum average outlet concentration of each system. The desalination percentage achieved in the buffered system was higher than the cascaded system. Additionally, less energy was consumed to reach the minimum average concentration in the buffered system, comparing to the cascaded system. Nevertheless, in the buffered system, the total amount of treated water happened to be lower. In order to compare the efficiency of two systems, the amount of salt adsorbed per unit of consumed energy, up to the point with the minimum average concentration, was calculated. According to the obtained values, the efficiency of the buffered system in terms of the proposed metric is higher than the cascaded, nonbuffered system.

Comparing to the numerical values obtained for the minimum average concentration in both systems, in the theory section, although the experimental values appear to be lower, the terms of minimum average outlet concentration still hold. The aforementioned difference between the simulation and experimental values might be due to simplifications and assumptions made for developing the physical model and inherent differences between the experimental setup used for this work and the one employed by Perez et al.

Aiming at improving the performance of the buffered system, in terms of energy consumed per unit volume of treated water, the number of the CDI units in the buffered system can be increased. Furthermore, after each solution buffer, two or more CDI cells can desalinate in a parallel architecture, decreasing the amount of energy consumed per unit volume of the desalinated water. Also, dimensions of the CDI unit and the activated carbon sheets can be enlarged, boosting the overall performance of the buffered system.

It is worth mentioning that although the properties of the porous media and the electrical field applied during the desalination directly affect the ultimate performance of the CDI unit, all the theories discussed in this publication do not depend on the electrode's material and the applied voltage. As a result, all the theoretical statements regarding the desalination performance of one-fully developed-cycle versus multiple-developing-cycles CDI, CDI unit

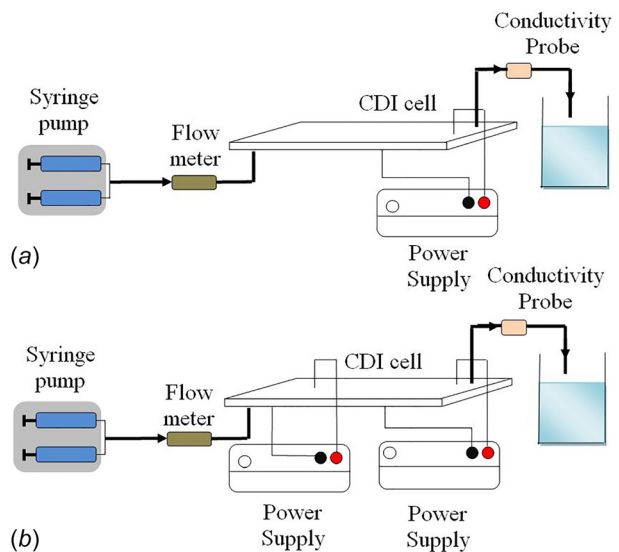


Fig. 6 Schematic of the experimental setup for two CDI systems. The buffered system: (a) a steady desalination test was conducted with a single CDI unit. The minimum average concentration obtained was used as the inlet concentration for the second steady desalination test, with the same CDI unit. The cascaded system: (b) one steady desalination test was performed with a CDI cell consisting of two pairs of current collectors and activated carbon electrodes, located at a distance from each other.

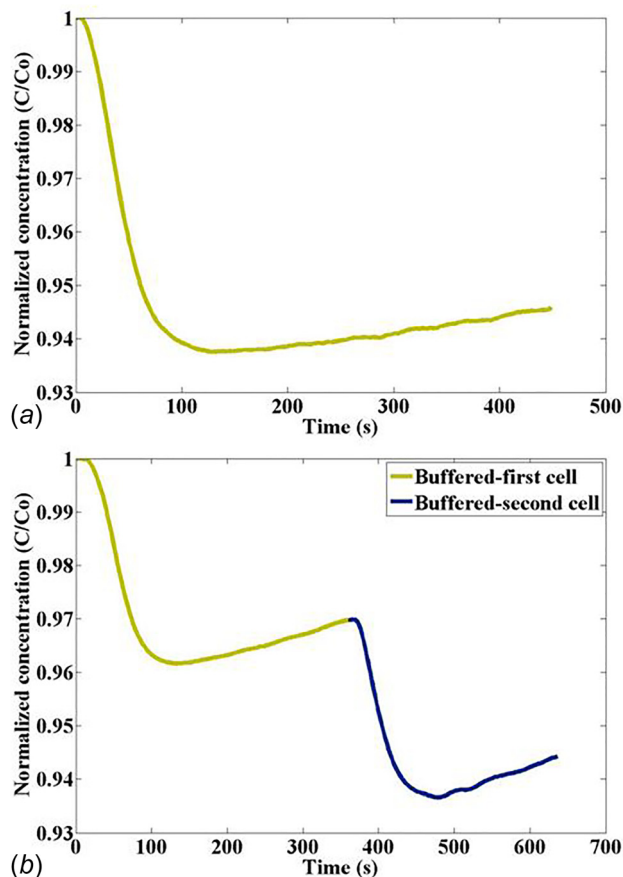


Fig. 7 Experimental data for normalized outlet concentration over time for the cascaded (a) and the buffered (b) CDI systems

with total length of L versus two split CDI cells, each having the length of $L/2$, and buffered versus cascaded CDI systems are valid for other porous electrodes and electrical voltages, as well. The values of the calculated metrics (desalination percentage, energy consumption, volume of the treated water, and the total amount of salt adsorbed per unit of the energy consumed) will change based on the electrodes' physical and chemical properties and the applied voltage. However, the overall superiority of one system over the other does not depend on these factors.

Conclusion

Based on the previously developed physical model, a new technique, called the buffered system, for CDI water desalination was introduced. As the first step for proof-of-concept, performance of a buffered system consisting of two CDI units and one solution buffer was compared with two-cascaded-CDI unit desalination system without any intermediate solution. Experimental data demonstrated the superiority of buffered system in terms of desalination percentage, energy consumption, and the total amount of salt adsorbed per unit of consumed energy. Broadening the scope of the new proposed method, increasing the number of solution

Table 1 Summary of the experimental data for two CDI desalination systems: buffered and cascaded

	Desalination percentage (%)	Energy consumed (J)	Purified water (ml)	Salt adsorbed per unit of energy ($\text{g/J} \times 10^4$)
Buffered	8.3	0.39	22.07	1.77
Cascaded	5.4	0.91	37.20	1.06

buffers in the system, and employing multiple CDI units, connected in parallel, after each intermediate solution will improve the efficiency of the process in terms of energy input per volume of treated water.

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Nomenclature

- C_o = inlet concentration, mg/ml
 C_{ave} = average outlet concentration, mg/ml
 D_{eff} = effective diffusion coefficient, m^2/s
 E = energy input to the system, J
 I = electrical current, A
 L = length of the CDI cell, mm
 p = desalination percentage, %
 Q = solution flow rate within the cell, ml/min
 t = instant of the measurement, s
 V = applied voltage, V
 V_w = volume of the purified water, ml

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