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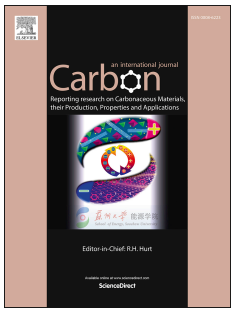

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Qiang Wei: Device design, Experiment methodology, Data analysis, Original draft reparation

Yudi Hu: Experiment methodology

Jian Wang: Device design

Qiang Ru: Discussion on the electrochemical experiments

Xianhou Hou: Primarily developed the present work, Investigation, Data analysis, Paper writing

Lingzhi Zhao: The electrochemical analysis, Paper writing

Denis Y. W. Yu: Reviewing and Editing

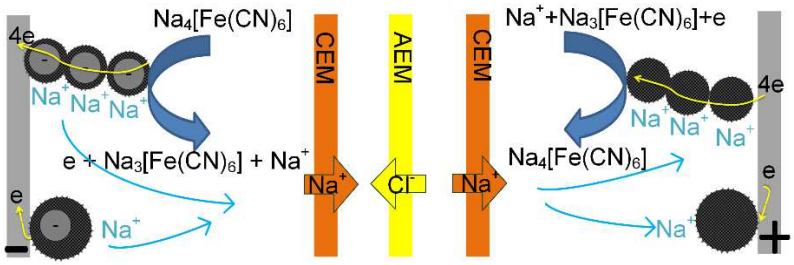
Kwan San Hui: Paper writing and Editing

Dongliang Yan: Discussion and analysis of the supplemented experiments

Kwun Nam Hui: Primarily developed the present work, Data analysis, Paper writing

Fuming Chen: conceptualized the work, Data analysis, wrote the paper, Supervision

By the introduction of redox couples to flow capacitive deionization, the continuous desalination process is achieved with a high salt removal rate and low energy consumption.



Low Energy Consumption Flow Capacitive Deionization with a Combination of Redox Couples and Carbon Slurry

Qiang Wei[1], Yudi Hu[1], Jian Wang[1], Qiang Ru[1], Xianhua Hou[1,2*], Lingzhi Zhao[1,2], Denis Y. W. Yu[3], Kwan San Hui[4], Dongliang Yan[5], Kwun Nam Hui[6*], Fuming Chen[1,2*]

[1] *Guangdong Provincial Key Laboratory of Quantum Engineering and Quantum Materials, Guangdong Engineering Technology Research Center of Efficient Green Energy and Environment Protection Materials, School of Physics and Telecommunication Engineering, South China Normal University, Guangzhou 510006 houxianhua@m.scnu.edu.cn, fmchen@m.scnu.edn.cn*

[2] *SCNU Qingyuan Institute of Science and Technology Innovation Co., Ltd., Qingyuan 511517, China*

[3] *School of Energy and Environment, City University of Hong Kong, Tat Chee Avenue, Kowloon, Hong Kong Special Administrative Region*

[4] *Engineering, Faculty of Science, University of East Anglia, Norwich, UK*

[5] *School of Material Science and Engineering, Guilin University of Electronic Technology, Guilin, 541004, China*

[6] *Joint Key Laboratory of the Ministry of Education, Institute of Applied Physics and Materials Engineering, University of Macau, Avenida da Universidade, Taipa, Macau, PR China bizhui@umac.mo*

Abstract: Flow-electrode capacitive deionization (FCDI) is a new sustainable desalination technology where continuous desalination can be achieved by the electrodialysis coupling method. However, its development is hindered owing to high energy consumption and low salt removal rate. Herein, by combining ferri-/ferrocyanide redox couple with flow activated carbon (AC)/carbon black (CB) slurry, continuous desalination process is achieved with a high salt removal rate of 1.31 μg cm^{-2} s^{-1} and low energy consumption of 102.68 kJ mol^{-1} at the current density 2.38 mA cm^{-2} (50 mA current for a 21 cm^2 active area). The operating voltage plateau can be reduced to 0.69 V when 10 wt% AC/CB (mass ratio of 9:1) is mixed

with 20 mM/20 mM ferri-/ferrocyanide as the flow electrodes, compared with more than 3 V for only carbon flow or redox medium alone. The influences of carbon content and current densities are further investigated to so that the performances can be controlled. This work enables the development of energy-saving desalination systems by coupling FCDI with redox desalination technique.

Keywords: Electrochemical desalination, Flow-electrode capacitive deionization, activated carbon/carbon black, redox desalination

1. **Introduction**

With the increase in population, the scarcity of fresh water is becoming a serious problem[1]. Ocean is a huge water source and it is therefore imperative to obtain freshwater from the sea by some effective desalination methods. Reverse osmosis and distillation are the matured desalination technology. However, the desalination equipment system is very complicated and expensive. Capacitive deionization (CDI) is a water flow desalination technology without the high pressure applied[2-5]. Its advantages include high energy-efficiency, cost-effectiveness, and eco-friendliness[6, 7]. When an electrical field is applied between the carbon electrodes, the ions in the salt feed are adsorbed to purify the water. Traditional CDI utilizes static carbon material as electrodes with only limited salt removal capacity of less than 20 mg g^{-1}, and regeneration has to be carried out after salt absorption saturation [8, 9]. Therefore, flow-electrode capacitive deionization (FCDI) has been introduced and rapidly developed recently [8, 10-13]. Compared with the traditional CDI technology, the electro-active flow with suspended carbon-based materials such as activated carbon, carbon nanotubes, or other conductive agents in slurry form as flow electrodes enhances the electrochemical salt removal capacity in FCDI [14-17]. Specifically, the suspended carbon black (CB) or activated carbon (AC) slurry is allowed to flow in the two electrode streams, and the salt stream passed through the middle compartment. This flow carbon electrode-based CDI exhibits an unlimited salt removal capacity. Other host materials like the redox couples can be introduced into the carbon flow electrode to enhance the charge transport as well as conductivity. Ma et. al. demonstrated the mixture for organic-based redox couple into 1 wt% activated carbon electrode in an aqueous medium which enhances the FCDI desalination performance [18]. The aqueous hydroquinone/benzoquinone was chosen as the redox species for desalination at the constant voltage mode. However, hydroquinone or benzoquinone is dangerously toxic for human. During the re-circulating redox reaction, their derivatives such as halobenoquinones

may be generated in the stream chlorination. The initial salt feed with 2000 ppm was desalted to 1000 ppm at the constant voltage of 1.2 V. The synergistic effect between the carbon and redox couple played a key role to enhance the desalination performance further. The removal rate and energy consumption are two key parameters in the FCDI. In order to achieve low energy consumption and high removal rate, we propose to combine ferri-/ferrocyanide redox couple with AC/CB slurry as FCDI electrodes. The redox couple can enhance the charge transfer between the flowing electrodes and current collector. A low voltage plateau of 0.69 V can be obtained in the combination of redox couples and slurry-type electrode which flow through the positive and negative stream by re-circulation process. As a results, a salt removal rate of 1.31 μg cm^{-2} s^{-1} and energy consumption of 102.68 kJ mol^{-1} can be obtained at the current density 2.38 mA cm^{-2} (50 mA current and 21 cm^2 active area) with the mixture of 10 wt% AC/CB (mass ratio of 9:1) and 20 mM/20 mM ferri-/ferrocyanide as flow electrodes during the continuous desalination process, where the plateau of the operating voltage can be maintained at 0.69 V, compared with more than 3 V with only carbon flow or redox medium alone. The enhanced performance of the FCDI is due to the synergetic effect of carbon flow electrode and redox couple. We also found that the content of carbon electrodes greatly affects the removal rate and energy consumption. This work highlights a route towards fast desalination with the reduced energy consumption via the combination of redox couples and carbon slurry, even under high current conditions.

2. Experimental section:

2.1 Materials and cell design

The activated carbon (specific surface area: 1301.3 m^2 g^{-1}, particle size: ~19 μm) was ordered from Yihuan Carbon Co., Ltd., Fuzhou, China, and carbon black (specific surface area: 190.2

$m^2\ g^{-1}$, particle size: 12 μm) from Alfa Aesar. $K_3Fe(CN)_6$ (99%) was purchased from Energy Chemical, and $K_4Fe(CN)_6$ (⩾99.5%) from Macklin. NaCl (99.5%) was brought from Aldrich. The cell of redox flow FCDI was fabricated according to the following architecture as shown in Figure 1a and Figure S1: graphite paper (JING LONG TE TAN, Beijing, 0.2 mm thickness) as current collector | redox couples/carbon flow chamber || desalinated stream || brine stream || redox couples/carbon flow chamber | graphite paper as the current collector. The symbols of | and || detonate the compartment separation and membrane, respectively. The carbon flow chambers were made of acrylic plates (130 mm × 130 mm × 2 mm) with a snake flow path (7 cm length, 2 mm width and 2 mm depth). Each carbon flow chamber has 15 flow paths. The active area is 21 cm^2 in totally. Two pieces of cation exchange membrane (CEM, Tokuyama, Japan) are placed between the salt stream and redox carbon flow chamber at both sides. One piece of anion exchange membrane (AEM, Tokuyama, Japan) is located between brine and desalination stream.

2.2 Electrochemical measurement and desalination tests

The electrochemical properties of redox couples was measured with a three-electrode cell by cyclic voltammetry (CV) using an electrochemical workstation (CHI760E, CH Instruments, Ins, USA) in 0.5 M NaCl electrolyte containing 20 mM/20 mM of $K_3Fe(CN)_6$ and $K_4Fe(CN)_6$. The working electrode was a glassy carbon electrode with an exposed area with a diameter of 3 mm, polished by the suspended Al_2O_3 in deionized water. Pt grid and standard Ag/AgCl are used as the counter and reference electrode, respectively. The applied scan rate was controlled at 5 $mV \cdot s^{-1}$ with the working window range from −0.1 to 0.5 V vs. Ag/AgCl.

The flow electrode material contains water with 10 wt % AC/CB (mass ratio of 9:1) carbon slurry and 20 mM/20 mM of $K_3Fe(CN)_6$ and $K_4Fe(CN)_6$. 9000 ppm of NaCl was added to the above solution to enhance the conductivity, and the total volume of solution is 160 ml. The

whole mixture is allowed to stir for two days prior to the experiment. The desalination and concentrate chambers (streams A and B) contain the same concentration of NaCl with 9000 ppm in 80 mL initially. The desalination current was provided by the battery analyzer (Neware, Shenzhen, China) with a current of 10 mA, 30 mA, 50 mA, 70 mA or 90 mA at ambient temperature. The system was also tested with flow electrode with amount of different carbon slurry (i.e. 0, 1.11, 3.33, 5.56, 7.78 and 10 wt % AC/CB with mass ratio of 9:1). All the water flow was pumped at the rate of 8.1 ml min^{-1}. Before desalination, the solutions were kept under the flow condition for two hours without any applied current. The variation of salt concentrations in streams A and B were recorded by conductivity meters (EPU357, EDAQ, Australia).

2.3 Calculation Procedures

The salt removal rate (ASRR, μg cm^{-2} s^{-1}), which describes the desalination rate, can be calculated by the equation 1 [19, 20].

$$\mathrm{ASRR} = \frac{\left(\frac{\Delta c}{\Delta t} \times V\right)}{A_{cell}} \quad (1)$$

Where Δc/Δt is the salt concentration change per second (Δppm s^{-1}), V is the volume of the salt stream (80 mL) and A_{cell} is the cross-sectional area of the cell (21 cm^2) in this device.

Charge efficiency (Λ, %) is defined as the percentage ratio of salt removed to the applied electrons, which can be determined by the following formula:

$$\Lambda = \frac{\mathrm{ASRR} \times F}{60 \times 10^3 \times i \times M} \quad (2)$$

Where *i* is the current density (in mA cm^{-2}) and M is the molar mass of NaCl (58.44 g mol^{-1}). F is Faraday constant.

Energy consumption (E_c, kJ mole^{-1}) can be calculated as follows:

$$\overline{E} = \frac{3.6\Delta E}{\left[\left(c_0 - c_f\right) \times \frac{V}{10^6 \times M}\right]} \quad (3)$$

Where ΔE is the total energy consumption (Wh) during charging, c_0 and c_f are the initial and final NaCl concentrations in ppm, respectively.

3. Results and discussion

3.1 The combination desalination mechanism of redox couples and carbon slurry

The schematic representation of the FCDI device is shown in Figure 1a. All the compartments are made of acrylic plates. The two end plates with 8 mm thickness are fastened with screws to compactly support the device to avoid the leakage. The thicknesses of the other flow ccompartments are 2 mm. The flow slurry electrodes are re-circulated using peristaltic pumps. The flow electrode and the salt feed are recirculated independently. A proposed combination mechanism is schematically demonstrated in Figure 1b-d for flow electrode with carbon only, redox couples only and their synergetic combination.

Figure 1b shows the desalination mechanism with carbon absorption. In the anode carbon stream, the negative charged carbon releases an electron to the current collector and a sodium ion to the neighbor salt stream through CEM, in which the sodium ion are obtained from the cathode stream. This process is exactly the same as in the conventional FCDI or CDI. For the case with only redox couple material in the flow electrode, as displayed in Figure 1c, the desalination mechanism is based on the coupling electrolysis owing to the redox reaction of redox couples. On the surface of the anode current collector, the oxidation reaction of ferrocyanide occurs with the release of an electron to the outer circuit while the sodium ion is

transported to the neighbor salt stream through the CEM. At the cathode stream, ferricyanide is reduced to ferrocyanide with the acceptance of an electron from the outer circuit and sodium ion moves to cathode from the neighbor salt stream.

With the combination of redox couples and carbon material in Figure 1d, the release of electron and sodium ion will be greatly enhanced owing to the large active area from the carbon and more reactive sites from the redox couples on the surface of carbon material, especially with a high concentration of carbon slurry. During the charge process, the slurry in anode chamber is oxidized with the release of electron and sodium ion as shown in equation 4-5.

$$Negative\ carbon\ (Na^+) \rightarrow Carbon + Na^+ + e^- \quad (4)$$

$$Na_4Fe(CN)_6 \rightarrow Na_3Fe(CN)_6 + Na^+ + e^- \quad (5)$$

At the cathode side, the mixture electrode was reduced by electrons from the outer circuit and sodium ion is accepted via CEM as shown in equation 6-7.

$$Carbon + Na^+ + e^- \rightarrow Negative\ carbon\ (Na^+) \quad (6)$$

$$Na_3Fe(CN)_6 + Na^+ + e \rightarrow Na_4Fe(CN)_6 \quad (7)$$

The ion movement in salt stream can be presented as follows:

$$Na^+(Stream\ A) \xrightarrow{CEM} Positive\ stream \quad (8)$$

$$Cl^-(Stream\ B) \xrightarrow{AEM} Stream\ B \quad (9)$$

$$Na^+(Negative\ stream) \xrightarrow{CEM} Stream\ B \quad (10)$$

The three-electrode CV of ferricyanide/ferrocyanide is displayed in Figure S2. The electrolyte was prepared with 20 mM/20 mM of ferri-/ferrocyanide with the addition of NaCl. A pair of

redox peaks are observed, corresponding to the electron transfer owing to the reduction and oxidation reaction. The reduction peak at 0.2 V corresponds to $[Fe(CN_6)]^{3-}$ to $[Fe(CN_6)]^{4-}$ and the peak at 0.26 V was assigned to oxidation reaction. The redox potential interval of 60 mV indicates the excellent redox behavior, which will be beneficial to lower down the energy consumption during electrochemical desalination[21-23].

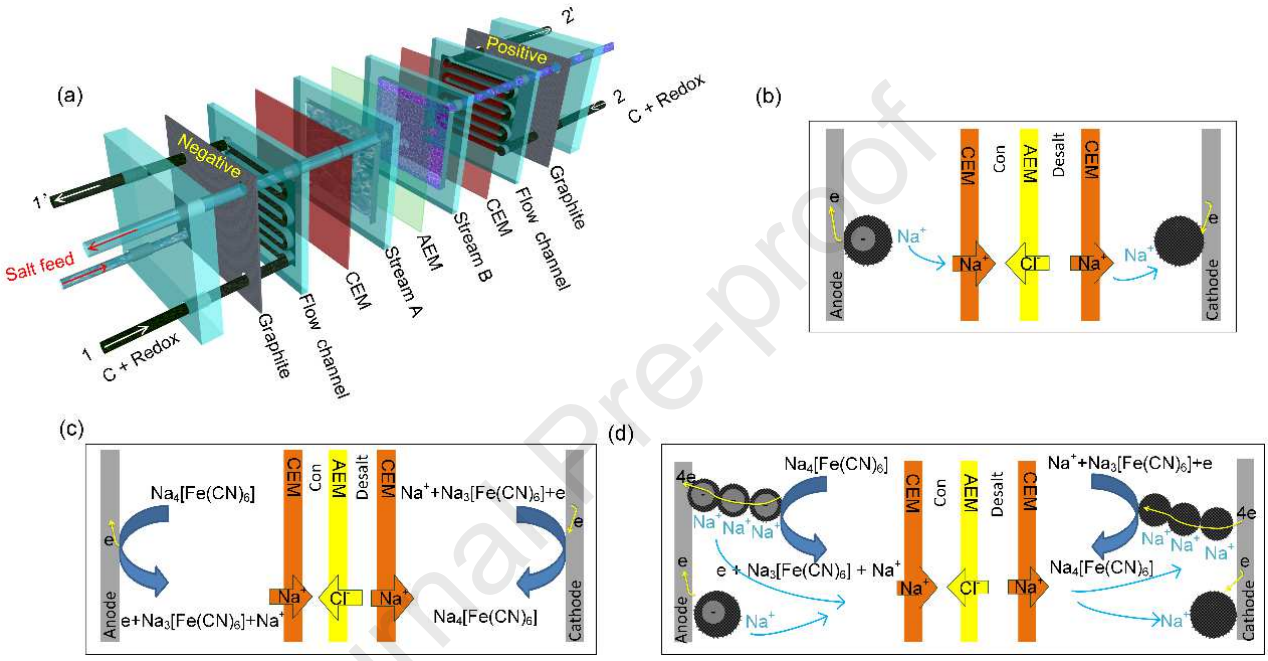


Figure 1. (a) Schematic representations of the FCDI cell; The proposed mechanism of FCDI with carbon material (b), the redox couple ferri-/ferrocyanide (c), and the synergetic combination of carbon and redox couples.

3.2 The enhanced desalination performance with redox couple and carbon slurry

To investigate the enhancement of desalination performance by combining redox couples and carbon slurry, we performed the comparison desalination tests using the redox couples or carbon slurry, separately. The results are demonstrated in Figure 2. The same current density of 2.38 mA cm^{-2} is applied. The 9 wt% AC and 1 wt% CB are dispersed in 20 mM/20 mM

ferri-/ferrocyanide as the flow electrode material. For AC/CB or redox couples alone, the operating voltage of more than 3 V can be observed. However, the operating voltage is lower down to 0.69 V with the combination of carbon slurry and redox couple, corresponding to the lowest energy consumption among the tested samples. As shown in Figure 2b, the energy consumption for the system with carbon slurry or redox couple is 410.26 kJ mol^{-1} or 356.18 kJ mol^{-1} individually. However, the energy consumption drops to 102.68 kJ mol^{-1} for the system with a mixture of carbon slurry and redox couple. This is attributed to the more reactive sites of redox couple on the surface of carbon slurry. The released electron from ferrocyanide at the anode can be easily transferred to the outer circuit, and accepted by the reduction of ferricyanide at cathode chamber owing to the large active surface area of the used AC/CB. To compare the electrochemical properties of carbon slurry, redox couples, or their combination, we supplemented the tests of linear sweep voltammetry (LSV). As shown in Figure S3, only ohmic behavior is observed in the carbon flow electrode. In the redox flow streams, the limiting current can be observed from the redox reaction of ferro/ferricyanide species. However, the limiting current is enhanced with the combination of carbon slurry and redox flow electrode compared with bare redox couples or carbon slurry only. This may be due to the more reaction sites of redox couples in the surface of carbon material. Thus, the LSV electrochemical tests indicate the mixed significance of carbon slurry and redox couples in the flow electrode chambers. Figure S4 shows the electrochemical impedance spectroscopy (EIS) of the flow electrodes of carbon slurry, redox couples, or their combination. The surface resistance (Rs) between the electrode and electrolyte in only redox couples is up to 113.7 ohms owing to the limited contact area on the surface of graphite paper. In the carbon slurry, the Rs value drops to 57.1 ohms. However, with the combination of carbon slurry and redox couples, the Rs value is down to 48.4 ohms, indicating the improved charge transfer kinetics. The EIS results are consistent with the LSV data in Figure S3. In the current tests, the salt

removal rate in the redox couples or the mixture of redox couple and carbon slurry is slightly higher than the one in carbon even at the same current density. This may be due to the high charge efficiency in redox couples owing to the effective electron and ion transfer. In the long-term tests of the current system as shown in Figure S5, the batch cycling experiment was carried out by topping up the salt feed without the replacement of electrode materials and membranes. At the current density of 2.38 mA cm^{-2}, 9000 ppm feed salt was desalted to approximate 650 ppm level during the four cycles. The long-term tests demonstrate the excellent stability of the current desalination system, which will be applicable for multi-cycle desalination.

The comparison of energy consumption and salt removal rate were tabulated in Table S1, together with results obtained from the literatures. The current study displays one of the best performances with a high salt removal rate and a low energy consumption compare to other works. This high performance may be due to the improved charge transfer kinetics and the reduced interface resistance of the flow electrode to accelerate the transfer of ion and electron in FCDI.

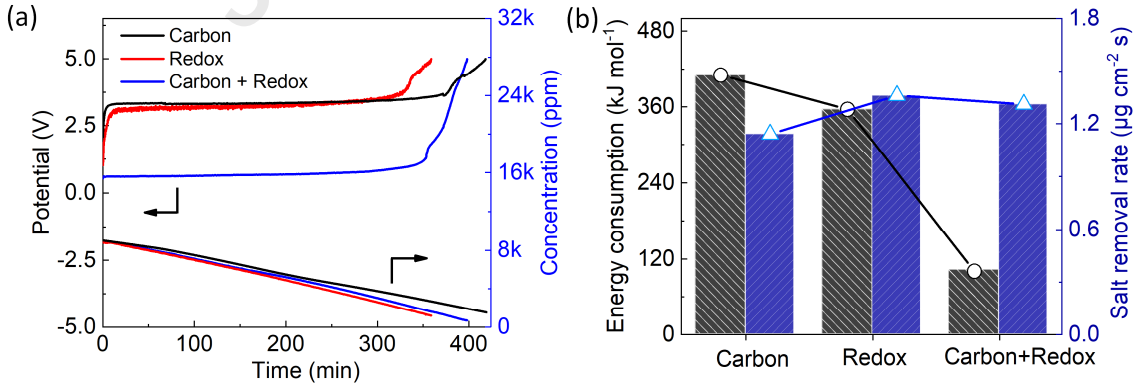


Figure 2. (a) The curves of charge voltage and the corresponding variation of salt concentration in carbon slurry, redox couples, and their mixture; (b) the corresponding energy consumption and salt removal rate. The current density: 2.38 mA cm^{-2}.

3.3 The influence of desalination performance from carbon content

As discussed above, the carbon content in flow electrode can greatly affect the operating voltage plateau. To investigate the influence of carbon content, the comparisons were conducted with the various amount of AC/CB such as 0, 1.11, 3.33, 5.56, 7.78 and 10 wt %. The ratio of AC:CB = 9:1 is kept in all the tested samples with the addition of the 20 mM/20 mM ferri-/ferrocyanide. The current density is 2.38 mA cm^{-2}. When the carbon mass fraction is larger than 10 %, the flow ability of the electrode material is restricted, thus the maximum carbon concentration is fixed at 10 % in this study. Figure 3a shows the voltage change during desalination at various carbon content. The detailed voltage and concentration change in diluted and concentrated streams are displayed in Figure S6. With the increase of carbon content in electrode streams, the voltage declines, which further confirmed the explanation, i.e. more reactive sites or contact opportunity between redox couples and carbon surface. At the last stage, the voltage rises quite fast owing to the ion depletion in the diluted channel. In all the experiments, the voltage was cut off at a maximum value of 5 V. The corresponding energy consumption and salt removal rate are displayed in Figure 3b. When a carbon content of 10 wt % is applied, the energy consumption is as low as 102.68 kJ mol^{-1} and the salt removal is 1.31 μg cm^{-2} s^{-1}. With the increase of carbon content, the salt removal rate increases firstly and then decreases. Among the tested samples, the best removal rate can be obtained at 5.56 % carbon content as shown in Figure 3b. The key point of FCDI device is the smooth flow carbon slurry between the positive and negative electrodes. When we disassembled the device after desalination completion, some minor carbon slurry still precipitates inside the path, which will cause the potential variation. In the future study, we will further optimize the design of snake path and make the carbon slurry flow smoothly.

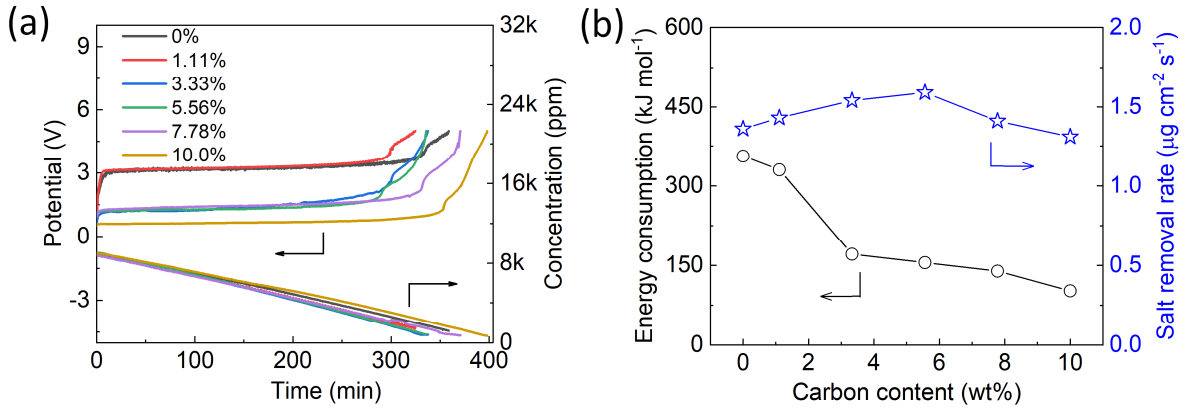


Figure 3. (a) The voltage curves with different carbon content (0, 1.11, 3.33, 5.56, 7.78 and 10 wt %) and the corresponding variation of salt concertation; (b) the corresponding energy consumption and the salt removal rate. The current density: 2.38 mA cm^{-2}.

3.4 Desalination performance at various current densities

Further exploration was conducted on the influence of current densities on the desalination process, as shown in Figure 4. The mixture of 10 wt% AC/CB (mass ratio of 9:1) and 20 mM/20 mM ferri-/ferrocyanide was used as the flow electrodes. The current densities of 0.47, 1.42, 2.38, 3.33 and 4.28 $mAcm^{-2}$ were applied. Figure 4(a) demonstrates the variations of voltage and salt content at current densities from 0.47 to 4.28 mA cm^{-2}. With the rise of current density, the voltage plateau increases as displayed in Figure 4b and Figure S7 due to the polarization. The salt removal rate depends on the applied current density in Figure 4c. The high current will drive the fast ion separation movement in both salt feeds. The removal rates in diluted channel is approximately equal to the capture rate in the concentrated reservoir. The energy consumption can be calculated from the measured the potential, current density, and removal rate. The energy consumption rises gradually with current density as shown in Figure 4d. The major reason is the increase in operating voltage with applied current. As shown in Figure 4d, in the 10 wt % carbon electrodes, the energy consumption is 52.22, 98.07,

135.39, 160.21 kJ mol^{-1} at the corresponding current densities of 1.42, 2.38, 3.33 and 4.28 $mAcm^{-2}$ respectively.

A comparison of literature on FCDI energy consumptions and removal rates is shown in Table S1 and Figure S8. For FCDI with 10 wt% DARCO and Noriactivated charcoal, the energy consumption is 162.61 kJ mol^{-1} at a current of 1.818 mA cm^{-2} [20]. Reducing the current to 0.909 mA cm^{-2} can lower the energy consumption down to 35.79 kJ mol^{-1}. On the other work, the energy consumption of 126.4 kJ mol^{-1} can be achieved using 13 wt % YP-50F activated carbon at the current of 3.0 mA cm^{-2} in FCDI [24]. In the current work, the energy consumption can be as low as 102.68 kJ mol^{-1} with 20 mM/20 mM ferri-/ferrocyanide and 10 wt% AC/CB (mass ratio of 9:1) at a current density of 2.38 mA cm^{-2} as shown in Figure 2. The synergetic effect from the combination of redox couple and carbon slurry enables low energy consumption and fast ion removal rate, as proved in LSV (Figure S3) and EIS (Figure S4). The results in this work demonstrate a feasible method to reduce the energy consumption by the introduction of redox species. Currently we are looking for other redox couples or carbon material with the better desalination performance to replace the current ones.

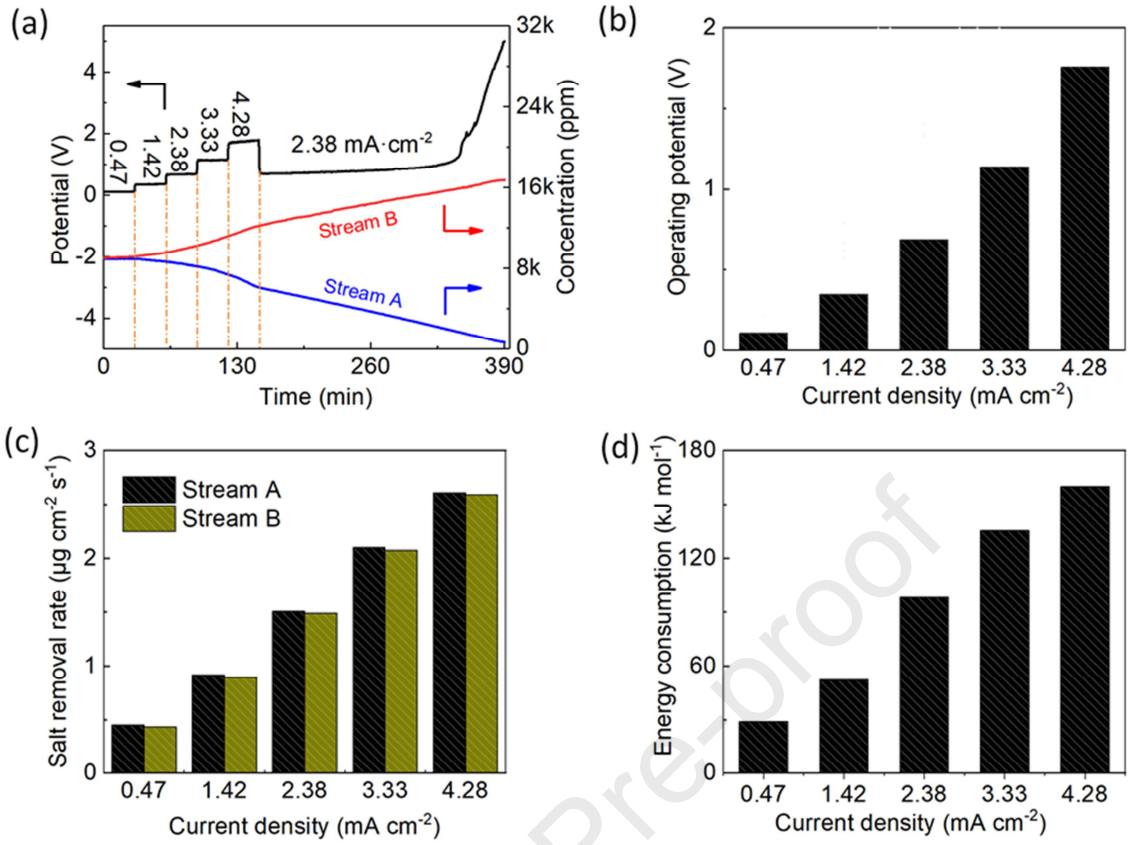


Figure 4. (a) Desalination performance, (b) operating voltage, (c) ASRR of brine and desalinated streams and (d) energy consumption at various current densities for 10 wt% carbon content.

Conclusion

In summary, the combination of ferri-/ferrocyanide redox couple and AC/CB slurry was demonstrated as FCDI electrode to achieve a low energy consumption and high removal rate. The salt removal rate of 1.31 μg cm^{-2} s^{-1} can be obtained with 10 wt% carbon slurry and 20 mM/20 mM of ferri-/ferrocyanide while the energy consumption is only 102.68 kJ mol^{-1}. The desalination performance of the electrode with combination of redox couple and carbon slurry is much better than that of the individual components alone. The excellent desalination performance is due to the synergetic effect from the combination of redox couple and carbon

slurry, which enables fast electron transfer and ion movement as proved by LSV and EIS. In addition, the desalination performance is affected by carbon content and current densities. Increasing the concentration of carbon slurry enhances salt removal. The new system with the combination of redox couple and carbon slurry presented in this work will propel the development of energy-saving FCDI system that can solve fresh water shortage.

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.

Acknowledgments

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Declaration of interests

☒ 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.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: