189 © 2021 The Authors Water Reuse | 11.2 | 2021

# Advances in efficient desalination technology of capacitive deionization for water recycling

Yuhao Tong, Shuang Zhou, Jun Zhou, Guanteng Zhang, Xiaolin Li, Chunxia Zhao and Pengyan Liu

#### **ABSTRACT**

Available freshwater resources are becoming harder to obtain due to climate change, population growth, industrial development, and water pollution. The main technologies in the field of wastewater desalination include reverse osmosis, electrodialysis, thermal distillation, and adsorption etc. Capacitive deionization technology (CDI) belongs to a novel electrochemical desalination technology with low energy consumption and low environmental impact, simple equipment structure and convenient operation. With the importance of wastewater desalination highlighted, some great technological progress of CDI has been made in electrode materials, reactor structure and the hybrid process. In this paper, the development of CDI technology was expounded from three aspects to achieve the goal of strong adaptability, low cost and strong adsorption capacity by analysis of the latest research papers. Corresponding improved methods of CDI are summarized to solve the main technology bottlenecks such as the inefficient and vulnerable electrode materials, low selectivity and unreasonable unit structure, and limitations of single CDI unit for promoting the continuous development of CDI technology.

**Key words** | capacitive deionization, desalination, electrode materials, reactor structure, water recycling

Yuhao Tong Shuang Zhou Jun Zhou Guanteng Zhang Xiaolin Li Chunxia Zhao (corresponding author) Pengyan Liu College of Chemistry and Environmental Science, Hebei University, Institute of Life Science and Green Development, Hebei University, Key Laboratory of Analytical Science and Technology of Hebei Province,

Baoding 071002, China E-mail: zhaocxhbu@edu.cn

# **HIGHLIGHTS**

- The main technology bottlenecks of CDI were analyzed from three aspects.
- Review the development of CDI based on electrode material, novel process and hybrid process.
- New electrode materials are introduced and analyzed for the future development of CDI.
- The energy consumption of novel FCDI-NF hybrid process is significantly reduced.

# INTRODUCTION

Freshwater resources are essential for the sustainable development of human society. Rapid population growth, urbanization, and climate change are putting tremendous stress on freshwater resources (Fan *et al.* 2020); approximately one-fifth and one-third of the world's population live in areas of water scarcity and in countries with

moderate to high water stress, respectively, according to the report of UN Water (Chen et al. 2020). In the past decades, the usage and treatment of water in China has grown tremendously due to the large population and rapid economic development (Xu et al. 2020). Water recycling can be an effective option for saving water resources, reducing the environmental impacts from the discharge of treated wastewater, and reducing the cost and energy involved in water resource management (Lyu et al. 2016; Takeuchi & Tanaka 2020). The development of novel

This is an Open Access article distributed under the terms of the Creative Commons Attribution Licence (CC BY 4.0), which permits copying, adaptation and redistribution, provided the original work is properly cited (http://creativecommons.org/licenses/by/4.0/).

doi: 10.2166/wrd.2021.101

water regeneration technology is of great significance to alleviate the crisis of freshwater resources.

Traditional nonthermal distillation methodologies, such as reverse osmosis (RO), adsorption/ion exchange, and membrane desalination (MD) (Burakov et al. 2018; Folaranmi et al. 2020; Shi et al. 2020), have wide applications in desalination and water purification, while membrane fouling, adsorbent regeneration and high energy consumption are always problems to be solved (Naushad et al. 2015). Capacitive deionization (CDI) is a promising new technology for water desalination and ions removal compared to other traditional desalination methodologies, due to its low energy consumption (i.e. room temperatures, low pressures and low voltages), low capital and processing costs, easy operation and maintenance processes, and environmentally friendly characteristics (i.e. without chemicals or hazardous substances) (Khalil et al. 2020). CDI has emerged as a promising alternative desalination technology of reverse osmosis, electrodialysis and thermal distillation (Zhang et al. 2018; Tang et al. 2019). The electrosorption/desorption processes of CDI for deionization is based on electric double layer theory. Ions in the feed water will be attracted on the electrode with opposite charge by an external electrical potential (very low voltage, 1.2 V), and desorbed into the concentrate from the electrode by a reversing potential for a continuous desalination process (Faisal et al. 2020).

The technical principle of CDI is shown in Figure 1. Ions are removed by transferring from aqueous solution to

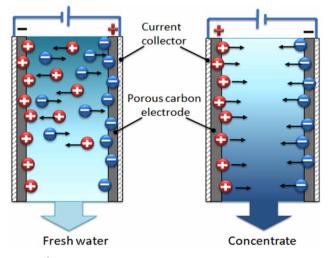


Figure 1 | Schematic diagram of the working and regeneration principle of CDI.

the electrode surface, the charge balance is achieved on the electric double layer, and the effluent is purified (Figure 1(a)). The ions or charged particles adsorbed on the surface of the electrode material are released into solution to realize the electrode regeneration through reverse connection or short-circuit voltage (Figure 1(b)). During the charging and discharging process of the CDI electrodes, the absorption and desorption of ions are completed, respectively. Therefore, the electrode materials with huge surface, ultra-high capacitance and high desalination efficiency are essential to CDI technology for water resources recycling.

Porous carbon electrodes materials are widely used in electrostatic adsorption of ions in the CDI process. Significant efforts have been made in recent decades in the development of customized porous carbon materials for CDI including carbon nanotubes, graphene, MXene, aerogels, xerogels, mesoporous carbon, and activated carbons. Although posing unique benefits, so far these materials have generally failed to provide a significant competitive advantage to traditional adsorption and RO in either capital or operating cost due to low salt adsorption capacity and carbon electrode oxidation (Landon et al. 2019). Nowadays, how to promote the desalination efficiency of CDI device is still a difficult problem for many researchers to develop materials with ultra-high capacitance or new processes with more efficient desalination performance (Zhao et al. 2020).

The desalting performance of CDI also depends on the operating parameters including external voltage, inflow velocity, thickness of interlayer between two electrodes and design of flow channel, and quality, species and concentration of solution ions etc. (Chen et al. 2018). Commonly used CDI desalination performance evaluation indicators mainly include electroadsorption capacity, charge efficiency, average desalination rate, and long-term stable desalting belonging to reactor structure optimization (Yasin et al. 2018; Maheshwari et al. 2020; Wang et al. 2020).

In this paper, the three main technology bottlenecks have been analyzed for the desalination performance of CDI device such as the inefficient and vulnerable electrode materials with low selectivity, unreasonable unit structure and limitations of single CDI unit (Figure 2). Furthermore, corresponding improved methods of CDI were

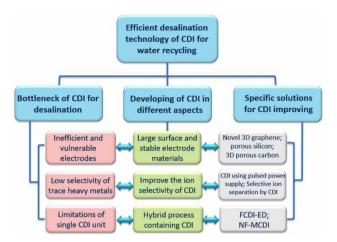


Figure 2 | Overview of the bottlenecks, developments and solutions of CDI for water recycling

summarized for promoting the continuous development of CDI technology based on the electrode material, novel process and hybrid process.

## **NEW CDI ELECTRODE MATERIAL**

Among the various factors that affect the desalination performance of CDI, the most important one is the selection of electrode materials. The pore size distribution, specific surface area, hydrophilicity, stability, specific capacitance and resistance of the electrode material all have the most direct impact on the capacitance adsorption effect (Shi et al. 2020; Zhao et al. 2020). This section focuses on solving the problems of instability and low performance of traditional CDI electrodes. Based on the research of the past five years, researchers have developed new electrode materials with a large specific surface area and stable electrochemical performance. Next, three representative new electrode materials are introduced and analyzed, the advantages and disadvantages of each new electrode material are explored, and the future development of CDI electrode materials is expected.

#### Electrospun network electrode

Conventional CDI electrode materials include activated carbon, graphite and carbon nanofibers (CNFs), but these materials have the problems of high manufacturing cost and low adsorption capacity, which limits the large-scale application of CDI. Researchers find that the high surface area and porosity of the electrode material are very important to enhance the specific capacitance and cycling stability of the capacitor to achieve high performance (Jiang & School of Petrochemical Engineering 2020). Electrospinning, as a simple yet effective technique to fabricate continuous CNFs, has attracted much attention in the CDI field (Feng et al. 2019). The combination of free-standing e-CNFs and PCP with hierarchical pore structure should be a promising electrode material for CDI application (Liu et al. 2015). A new CNFs reinforced 3D PCP network (e-CNF-PCP) was fabricated through electrospinning followed by thermal treatment and used as electrode materials for CDI with an EC of 16.98 mg/g in 500 mg/L NaCl solution (Figure 3), which shows great improvement compared to those of PCP and e-CNFs (Liu et al. 2016). The CDI performances and manufacturing difficulties of e-CNF-PCP were compared with those of other carbon electrode materials reported in the literature, as shown in Table 1.

However, due to the complex processing technology and high cost of CNF materials, it is difficult to realize large-scale industrial application. Therefore, in the future development of this technology, researchers should focus on the realization of low-cost electrospinning and thermal treatment process to reduce the manufacturing cost of e-CNF-PCP, so that e-CNF-PCP can be widely used as a CDI electrode (Wang et al. 2020).

# Novel 3D graphene electrode

In order to achieve optimal desalination during CDI, CDI electrodes should possess high electrical conductivity. large surface area, good wettability to water, narrow pore size distribution and efficient pathways for ion and electron transportation (Shi et al. 2016). Graphene is one of the thinnest materials in the world, and the theoretical thickness value, band length, and bond angle are 0.335 nm, 0.142 nm, and 120°, respectively. Compared with 2D graphene, 3D graphene is an ideal electrode material for CDI because of its large surface area, good mechanical strength and flexibility (Yu et al. 2016). The researchers fabricated a novel CDI electrode based on a three-dimensional graphene (3DG) architecture

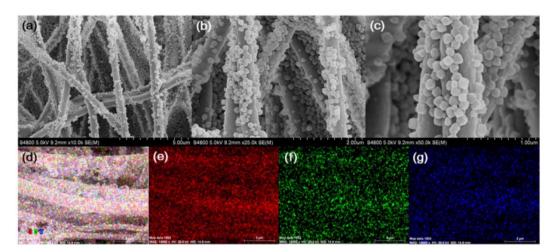


Figure 3 (a-c) FESEM images of e-CNF-PCP at different magnifications and elemental mapping images of (d) e-CNF-PCP, (e) C element, (f) N element and (g) O element (Liu et al. 2016).

Table 1 Comparison of CDI performances and manufacturing difficulties among various carbon electrode materials from the literature (Liu et al. 2016)

Electrode material	CDI performances NaCl concentration (mg/L)	Electrosorption capacity (mg/g)	Manufacturing difficulties
CAs	~500	2.9	Supercritical drying, binder
AC	500 1,000 1,500 2,000	9.72 10.80 11.00 11.76	Coating process, binder
CNTs	500 1,000 1,500 2,000	2.57 3.71 4.76 5.24	Chemical vapor deposition, binder
e-CNF- PCP	100 300 500	10.03 12.56 16.98	-

constructing interconnected graphene sheets with in-plane nanopores (NP-3DG). As compared to 3DG, NP-3DG features a larger specific surface area and therefore higher specific capacitance (Figure 4). The NP-3DG electrode can be used as a promising electrode material for highperformance CDI applications. The results of NP-3DG exhibit ultrahigh CDI performance with a superior electrosorption capacity of 22.09 mg/g, high adsorption rate, good deionization cyclic stability and high salt removal percentage. Furthermore, due to its excellent electrosorption performance, it is expected that NP-3DG will also

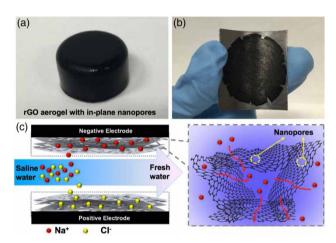


Figure 4 | Photographs of (a) as-prepared rGO hydrogel with in-plane nanopores (NP-3DG) and (b) CDI electrode, (c) Schematic diagram of the CDI process (Shi et al. 2016).

play an important role in the removal of charged hazardous species and other heavy metal ions (Khan et al. 2018).

As a new type of 3D carbon nanomaterial, NP-3DG has incomparable advantages for use in CDI electrodes. Because it is still in the initial stage of development, there are some problems such as high cost, imperfect theoretical system and unsatisfactory treatment effect (Zhang et al. 2018). In recent years, although some progress has been made in the design and preparation of graphene CDI electrode materials, further research on the construction of graphene pore structure and the comprehensive performance of CDI graphene electrode materials is needed to promote the large-scale practical development and application of graphene electrode materials in CDI.

# High efficiency particle free porous silicon network electrode

An energy saving water desalination process using low-cost materials is a key step for the sustainable demand of clean water resources in the future. Here, porous silicon has been proven to be a kind of material with rich content, low cost and strong biocompatibility (Gu et al. 2016). With appropriate surface passivation of the porous silicon material to prevent surface corrosion in aqueous environments, the results showed that porous silicon templates can enable salt removal of 0.36 and 0.52% in brackish water (10 mM, or 0.06% salinity) and ocean water (500 mM, or ~2.9% salinity) by CDI, respectively. The use of porous silicon for CDI enables new routes to directly couple water desalination technology with microfluidic systems or photovoltaics cells (Figure 5), while mitigating adverse effects of water contamination occurring from nanoparticulate-based CDI electrodes (Metke et al. 2016).

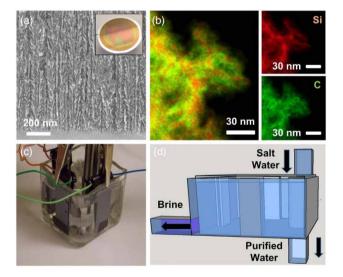


Figure 5 | Porous silicon-based CDI electrodes: (a) Cross-sectional SEM image of a carbon passivated porous silicon electrode for water desalination; (b) Analytical elemental TEM analysis a carbon passivated porous silicon material showing uniform carbon passivation on the material; (c) Photograph of the CDI testing cell combining four pairs of silicon-based electrodes. and (d) illustration of a CDI module in the context of the testing apparatus for continuous flowthrough CDI water desalination using porous silicon (Metke et al. 2016).

According to Figure 6, the porous silicon network electrode described in this section has a specific salt removal rate of 3.8-5.2 mg/g, ranging from fresh water to seawater salt concentration, and only 1.45 Wh/L of energy input is needed to convert seawater into drinking water (Yu et al. 2017; Zhao et al. 2019a, 2019b).

In summary, compared with the traditional carbonbased electrode, the main advantage of a porous silicon network is that the silicon content is huge, resulting in lower cost, and it has a tunable pore morphology and structure that can optimize CDI performance in different flow architectures and mitigate the effect of fouling. Therefore, a porous silicon network can be used in developing countries on a large scale, thus becoming a low-cost and easy to operate basic seawater purification equipment.

## **NEW CDI PROCESS**

Although CDI has made great breakthroughs in electrode materials, architecture and system integration, it has no advantages over traditional desalination technologies in terms of energy efficiency, water recovery rate, desalination concentration range and capital cost. However, CDI has illustrated great potential for selective removal of target ions because of its tailorable electrode materials and ability to couple with selective ion exchange membranes (Zhang et al. 2020; Park et al. 2021). In this section, two novel CDI processes were introduced for their higher ability of selective ions removal, and the application feasibility was explored.

#### Novel CDI using a pulsed power supply

In recent years, capacitive deionization technology (CDI) has been widely used in many fields due to its environmental protection, low energy consumption, simple operation, and safety (Cho et al. 2019). For the desalination of low-concentration brine (50 mg/L), electrodialysis, reverse osmosis and ion exchange are usually used. However, the above technologies have high energy consumption and high additional costs. Therefore, the CDI desalination was proposed to provide new ideas for the preparation of ultrapure water and the advanced treatment of wastewater (Yang et al. 2017).

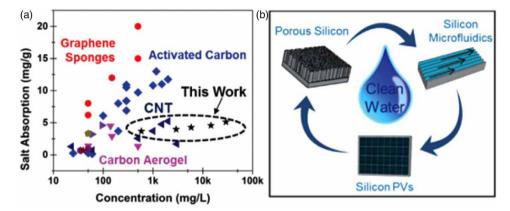


Figure 6 | High performance of porous silicon CDI electrodes and integration vision: (a) Comparison of average specific salt removal capacity for porous silicon electrodes compared to other notable forms of carbon; (b) Scheme emphasizing the vision of porous silicon integration with microfluidics and/or solar cells for technological water desalination platforms (Metke et al. 2016).

Pulsed electric field is a new technology to treat liquid and semi-solid objects with high electric field intensity (10-50 kV/cm), short pulse width (0-100 us) and high pulse frequency (0-2000 Hz) (Sistat et al. 2015) Under the pulsed electric field, ions transfer from solution to the electrode or the electro-dialysis membranes (Kim et al. 2019). The CDI cell, including the reactor and the activated carbon coated electrode, was designed. The pulsed power supply was applied in CDI for the electric field. In Figure 7, the removal rate of direct current-CDI (DC-CDI) and pulsedcapacitive deionization (pulsed-CDI) are compared (Jin et al. 2019).

In all, compared with the traditional direct current-CDI, novel pulsed-CDI has better ion removal rate and lower energy consumption, which proves that pulsed CDI can be an excellent alternative to traditional DC-CDI. However, due to the high cost and complex operation of a pulsed electric field, and the diversity of capacitive deionization electrode materials, repeated testing is needed to find the best voltage condition. Therefore, in the future, researchers should focus on enhancing the compatibility of the pulsed electric field so that it can better adapt to various CDI electrode materials, so as to realize large-scale industrial application.

# Selective ion separation by CDI based technologies

Selective ion extraction from aqueous solution is of great significance for water purification as well as resource recovery (Kavitha et al. 2019). However, despite the great

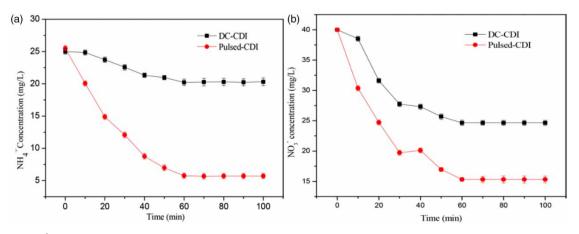


Figure 7 Performance comparison of DC-CDI and pulsed-CDI (a frequency of 100 Hz and a duty ratio of 50%) in desalination. (a) NH<sup>‡</sup><sub>4</sub>, (b) NO<sup>-</sup><sub>3</sub> (Jin et al. 2019).

development in the material, architecture, and system integration in the past 15 years, CDI was reported to have no advantages on energy efficiency, water recovery, salinity range, salt rejection, and capital cost compared with reverse osmosis (RO) (Jain et al. 2018). Nevertheless, due to its tailorable electrode materials and ability to couple selective ion exchange membranes/coatings on the electrode interface, the great potential of CDI has been shown for selective ions removal, which cannot be effectively solved by other desalination technologies including RO, electrodialysis or thermal evaporation (Zhang et al. 2020). Therefore, how to improve the ion selectivity of CDI has become an important development goal of CDI.

Hence, asymmetric activated carbon (AC) electrodes have been designed and grafted by sulfonic groups and amine groups on the surface of AC through covalent bonds as a cation-selective electrode and an anion-selective electrode respectively for enhanced capacitive deionization (Choi et al. 2019; Cen et al. 2020). Ten pairs of asymmetric electrodes are assembled with sulfonic AC as positive electrodes and aminated AC as negative electrodes. The scheme of co-ion minimization is illustrated in Figure 8. The electrosorption capacity and charge efficiency of asymmetric AC electrodes have been significantly improved because of not only preventing co-ion expulsion effect but also promoting the wettability and accelerating salt solution infiltration. These results will be beneficial to solve the technical bottlenecks and accelerate the practical engineering application of CDI. These electrodes have higher charge effi-(0.84) and higher electrosorption capacity ciency (23.2 mg·g<sup>-1</sup>) compared to pristine AC electrodes (0.54 of charge efficiency and 11.0 mg·g<sup>-1</sup> of electrosorption capacity) due to the wettability and hydrophilicity effectively enhanced by grafting ion-selective functional groups (Yan et al. 2018).

Asymmetric AC electrodes show better cycling stability (Zuo et al. 2018). The modification of the ion-selective functional groups on the surfaces of AC could reduce the co-ion effects effectively and improve the salt removal efficiency from the solution. This will provide a new opportunity for the practical development of capacitive deionization technology.

In all, compared with the traditional symmetrical electrode CDI, CDI using the asymmetric activated carbon electrode has higher charge efficiency and higher electrosorption capacity. However, many factors, including the oxidation of carbon-based materials, decomposition of intercalation structures, blockage of electrode pores caused by fouling and scaling, and membrane fouling resulting from heavy metals, would lead to the decline of desalination performance and also affect the stability of selective separation. Therefore, long-term operation stability should be evaluated, and protection measurements (such as water pretreatment, developing new materials, etc.) can also be adopted to enhance the system stability in the future (Zhang et al. 2020).

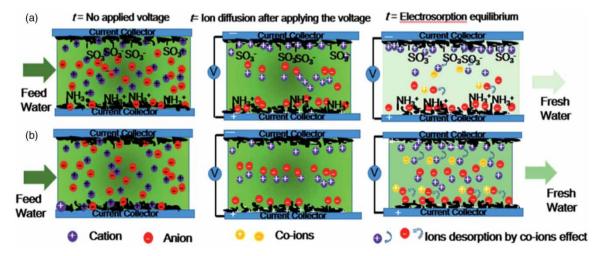


Figure 8 | Scheme illustration of the time-dependent CDI model of (a) asymmetric AC electrodes and (b) symmetric activated carbon electrodes (Yan et al. 2018).

## **HYBRID CDI PROCESS**

Although CDI has been developed for many years, there are still some problems that are difficult to solve, such as difficult regeneration and low treatment efficiency. These problems are usually attributed to the limitations of the device itself, which cannot be fundamentally solved by conventional device adjustment (Campione et al. 2020; Wu et al. 2020). The purpose of this section is to introduce the coupling application of CDI with other water treatment technologies. Through the advantages of other devices, the shortcomings of traditional CDI unit are supplemented. This method effectively frees CDI from its own limitations, solves the problems in traditional CDI and makes the industrial development of CDI have practical significance.

# Electrodialysis and FCDI coupling technology in continuous desalination

Membrane capacitive deionization (MCDI) and capacitive deionization (CDI) both adopt the fixed electrode mode, so the adsorption capacity is low and needs frequent regeneration, which does not fundamentally solve the problems of continuous desalination and low water efficiency caused by the small adsorption capacity of the fixed electrode. Researchers put forward the concept of flow capacitive deionization. Activated carbon (AC) was used as the mobile electrode. The scale of the project was realized by increasing the amount of the flow electrode (Jeon et al. 2019). By using electrodialysis (ED) as the regeneration unit of mobile electrode, and using flow capacitance deionization (FCDI) for desalination, the continuous and stable desalination process of brine can be realized (Cho et al. 2019). FCDI plays a role in the salt concentration of medium and low concentration brine (to obtain concentrated solution and desalination water), while ED carries out intermittent high-efficiency and rapid desalination of concentrated solution with high salt concentration. The FCDI-ED coupling process can realize industrial application of low energy consumption, high water efficiency and continuous treatment of brine (Hassanvand et al. 2017; Chen & Hou 2019; Gao et al. 2020). The processing route of FCDI-ED is shown in Figure 9.

However, the disadvantages of FCDI-ED technology are complex process, high cost and easy failure, so it is difficult to carry out large-scale industrial application at this stage. Therefore, researchers should focus on improving the stability of the process and using renewable materials, so as to make the FCDI-ED coupling process better developed.

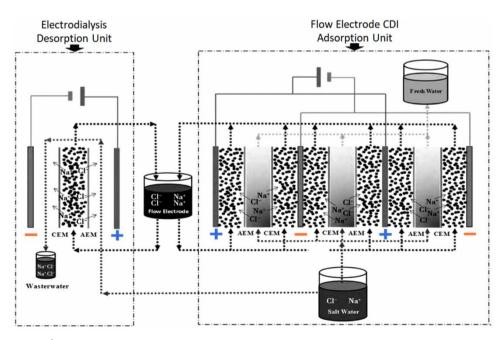


Figure 9 | Schematic diagram of deionization principle of FCDI-ED process (Mo et al. 2017).

# Novel nanofiltration and membrane capacitive deionization (NF-MCDI) hybrid system

Among various desalination processes, CDI has advantages in terms of cost and energy efficiencies and ease of operation and maintenance, as it can remove salts by adsorbing the ions on the surface of an electrode with a relatively low cell voltage of 1-1.4 V and without high pressure pumps or heat sources (Suss et al. 2015). However, single CDI has demonstrated low removal efficiency with a high-concentration (TDS > 3,000 mg/L) because of the ion saturation on the electrode (Choi et al. 2019). Using the combined RO-MCDI system, TDS of seawater was decreased from 35,000 to 200 mg/L (Abdelkader et al. 2018). However, the operation cost of this process is high, so it is difficult to use in a large-scale seawater desalination plant.

In recent years, a novel hybrid process that combines CDI with nanofiltration (NF) has received growing attention for implementation in energy-efficient desalination systems. Researchers proposed an MCDI unit with flowelectrodes followed by NF, abbreviated as FCDI-NF (Choi et al. 2017; Oatley-Radcliffe et al. 2017; Sami et al. 2018). The energy consumption of FCDI-NF (70% water recovery with 0.460 kWh m<sup>-3</sup>) was lower by 16% when treating water in comparison with the practical minimum energy consumption of the RO (0.550 kWh m<sup>-3</sup>) unit for 10,000 ppm NaCl solution desalination to drinking water range. Moreover, the FCDI-NF system attained the drinking water TDS standard (≤500 mg/L) in the final effluent. The schematic of nanofiltration and membrane capacitive deionization (NF-MCDI) hybrid system configuration is shown in Figure 10. The location and sequence of NF unit and CDI unit should be comprehensively considered by the influent quality, requirements of effluent and energy consumption.

After study, researchers found that the standalone NF system was insufficient for salty water desalination due to its comparatively low removal rate of <60% (NaCl). However, it was improved through combination with the MCDI unit to adsorb the remaining salts in the NF-treated water, thereby achieving a total salt removal rate of up to 95% (Jeon et al. 2019). Therefore, compared with the single NF or FCDI process, the desalination rate of FCDI-NF is significantly improved, and the energy consumption of the process is also significantly reduced.

However, this hybrid system may require a higher capital cost than the typical RO membrane system because of the price of the electrodes and ion-exchange membranes (Marchetti et al. 2017). Also, the NF-MCDI system performance is influenced by the feed concentration, MCDI flow rate and the NF recovery rate. Therefore, the complexity of the system will result in a significant increase in the capital cost and maintenance complexity of the specific equipment used to couple the MCDI system with the NF system. In future work, how to overcome the limitations of industrial applications brought about by the high cost and high complexity of FCDI-NF devices is very important.

# **CONCLUSION AND PROSPECTS**

In less than 20 years CDI technology has developed into the most attractive research field and it is expected that in the

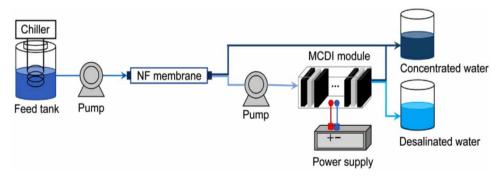


Figure 10 | Schematic of nanofiltration and membrane capacitive deionization (NF-MCDI) hybrid system configuration (Jeon et al. 2019).

next few years CDI will further explosive growth in a more mature and advanced direction. Nowadays, CDI has begun to adopt a variety of new technologies and electrode materials, which have unique functions and characteristics, and have made significant progress.

Through the systematic review of the current research it is found that improvement of the structure and composition of electrode materials can improve the desalination effect of CDI technology. However, further research is needed to realize the large-scale industrialization of CDI technology in the following aspects. (1) It is necessary to expand the research on the mechanism of influencing factors of adsorption performance and the kinetics of electro adsorption reaction, so as to provide theoretical basis for the study of electrode materials. (2) Improving the ability of CDI to remove ions selectively can develop CDI into a desalination device that can specifically remove specific ions, thus making up for the deficiency that traditional desalination technology can not specifically remove ions. (3) The development of electrode materials with strong adaptability, low cost and strong adsorption capacity can provide convenience for future research and development, and further promote the application of CDI technology.

The exciting development in the field of CDI is the emergence of FCDI batteries. The selection and synthesis of carbon electrode materials in FCDI have revolutionary changes because there is no need for polymer binders which have side effects on ion adsorption. The FCDI system not only has strong salt adsorption capacity to treat high salinity water, but also can continuously produce fresh water and regenerate electrodes by using non-charged carbon-based materials to continuously supplement the electroactive region. However, FCDI is still in the initial development stage, and there are many possibilities for its future development. One of the main current challenges for FCDI is the low efficiency of charge transfer between collector and flow electrode, and much work needs to be done to solve this problem.

In conclusion, despite the challenges, CDI is still a promising water treatment technology and exciting research field. With the continuous solution to these challenges and the emergence of new CDI units, CDI may be regarded as the key to solve the global water shortage and water pollution in the future.

## **ACKNOWLEDGEMENTS**

This work was supported by financial support from the Natural Science Foundation of (51308179); the Natural Science Foundation of Hebei Province (E2020201036, B2018201224); the research fund of Hebei Education Department (KCJSZ2020008); and the Laboratory Opening Project of Hebei University (sy202054).

## **DATA AVAILABILITY STATEMENT**

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

#### REFERENCES

- Abdelkader, B. A., Antar, M. A. & Khan, Z. 2018 Nanofiltration as a pretreatment step in seawater desalination: a review. Arabian Journal for Science and Engineering 43 (9),
- Burakov, A. E., Galunin, E. V., Burakova, I. V., Kucherova, A. E., Agarwal, S., Tkachev, A. G. & Gupta, V. K. 2018 Adsorption of heavy metals on conventional and nanostructured materials for wastewater treatment purposes: a review. Ecotoxicology and Environmental Safety 148, 702-712.
- Campione, A., Cipollina, A., Toet, E., Gurreri, L., Bogle, I. D. L. & Micale, G. 2020 Water desalination by capacitive electrodialysis: experiments and modelling. Desalination **473**, 114150.
- Cen, B., Li, K., Lv, C. & Yang, R. 2020 A novel asymmetric activated carbon electrode doped with metal-organic frameworks for high desalination performance. Journal of Solid State Electrochemistry 24 (3), 687-697.
- Chen, T. & Hou, C. 2019 Low energy desalination using a novel capacitive deionization-electrodialysis process. Abstracts of Papers of the American Chemical Society 257, 313.
- Chen, Z., Zhang, H., Wu, C., Luo, L., Wang, C., Huang, S. & Xu, H. 2018 A study of the effect of carbon characteristics on capacitive deionization (CDI) performance. Desalination **433**, 68-74.
- Chen, Z., Wu, G., Wu, Y., Wu, Q., Shi, Q., Ngo, H. H., Vargas Saucedo, O. A. & Hu, H. 2020 Water Eco-Nexus cycle system (WaterEcoNet) as a key solution for water shortage and water environment problems in urban areas. Water Cycle 1, 71 - 77.
- Cho, Y., Yoo, C. Y., Lee, S. W., Yoon, H., Lee, K. S., Yang, S. & Kim, D. K. 2019 Flow-electrode capacitive deionization with highly enhanced salt removal performance utilizing high-

- aspect ratio functionalized carbon nanotubes. Water Research 151, 252-259.
- Choi, S., Chang, B., Kang, J. H., Diallo, M. S. & Choi, J. W. 2017 Energy-efficient hybrid FCDI-NF desalination process with tunable salt rejection and high water recovery. Journal of Membrane Science 541, 580-586.
- Choi, J., Dorji, P., Shon, H. K. & Hong, S. 2019 Applications of capacitive deionization: desalination, softening, selective removal, and energy efficiency. Desalination 449, 118-130.
- Faisal, A. A. H., Al-Wakel, S. F. A., Assi, H. A., Naji, L. A. & Naushad, M. 2020 Waterworks sludge-filter sand permeable reactive barrier for removal of toxic lead ions from contaminated groundwater. Journal of Water Process Engineering 33, 101112.
- Fan, W., Yang, X., Wang, Y. & Huo, M. 2020 Loopholes in the current reclaimed water quality standards for clogging control during aquifer storage and recovery in China. Water Cycle 1, 13-18.
- Feng, J., Xiong, S. & Wang, Y. 2019 Atomic layer deposition of TiO<sub>2</sub> on carbon-nanotube membranes for enhanced capacitive deionization. Separation and Purification Technology 213, 70-77.
- Folaranmi, G., Bechelany, M., Sistat, P., Cretin, M. & Zaviska, F. 2020 Towards electrochemical water desalination techniques: a review on capacitive deionization, membrane capacitive deionization and flow capacitive deionization. Membranes (Basel) 10 (5), 96.
- Gao, F., Wang, L., Wang, J., Zhang, H. & Lin, S. 2020 Nutrient recovery from treated wastewater by a hybrid electrochemical sequence integrating bipolar membrane electrodialysis and membrane capacitive deionization. Environmental Science -Water Research & Technology 6 (2), 383-391.
- Gu, H., Rahardianto, A., Gao, L. X., Christofides, P. D. & Cohen, Y. 2016 Ultrafiltration with self-generated RO concentrate pulse backwash in a novel integrated seawater desalination UF-RO system. Journal of Membrane Science 520, 111-119.
- Hassanvand, A., Wei, K., Talebi, S., Chen, G. Q. & Kentish, S. E. 2017 The role of ion exchange membranes in membrane capacitive deionization. Membranes 7 (3), 54.
- Jain, A., Kim, J., Zuo, K., Li, Q. & Verduzco, R. 2018 Ion-selective and high capacity electrodes for membrane capacitive deionization. Abstracts of Papers of the American Chemical Society 255, 469.
- Jeon, S., Lee, J., Jo, K., Kim, C., Lee, C. & Yoon, J. 2019 Novel reuse strategy in flow-electrode capacitive deionization with switch cycle operation to enhance desalination performance. Environmental Science & Technology Letters 6 (12), 739-744.
- Jiang, X. & School of Petrochemical Engineering L. U. O. T. 2020 Electrospinning preparation of Fe<sub>3</sub>O<sub>4</sub>/porous carbon nanofibres for use as supercapacitor electrode materials. International Journal of Electrochemical Science 15 (5), 4602-4618
- Jin, Z., Hu, X., Sun, T. & Zhang, W. 2019 Capacitive deionization using a pulsed power supply for water treatment. Desalination and Water Treatment 160, 23-31.

- Kavitha, J., Rajalakshmi, M., Phani, A. R. & Padaki, M. 2019 Pretreatment processes for seawater reverse osmosis desalination systems - a review. Journal of Water Process Engineering 32, 100926.
- Khalil, K. A., Barakat, N. A. M., Motlak, M. & Al-Mubaddel, F. S. 2020 A novel graphene oxide-based ceramic composite as an efficient electrode for capacitive deionization. Scientific Reports 10 (1), 9676.
- Khan, Z. U., Yan, T., Shi, L. & Zhang, D. 2018 Improved capacitive deionization by using 3D intercalated graphene sheet-sphere nanocomposite architectures. Environmental Science Nano **5** (4), 980–991.
- Kim, Y. H., Tang, K., Chang, J., Sharma, K., Yiacoumi, S., Mayes, R. T., Bilheux, H. Z., Santodonato, L. J. & Tsouris, C. 2019 Potential limits of capacitive deionization and membrane capacitive deionization for water electrolysis. Separation Science and Technology 54 (13), 2112-2125.
- Landon, J., Gao, X., Omosebi, A. & Liu, K. 2019 Progress and outlook for capacitive deionization technology. Current *Opinion in Chemical Engineering* **25**, 1–8.
- Liu, Y., Xu, X., Wang, M., Lu, T., Sun, Z. & Pan, L. 2015 Metalorganic framework-derived porous carbon polyhedra for highly efficient capacitive deionization. Chemical Communications 51 (60), 12020-12023.
- Liu, Y., Ma, J., Lu, T. & Pan, L. 2016 Electrospun carbon nanofibers reinforced 3D porous carbon polyhedra network derived from metal-organic frameworks for capacitive deionization. Scientific Reports 6, 32784.
- Lyu, S., Chen, W., Zhang, W., Fan, Y. & Jiao, W. 2016 Wastewater reclamation and reuse in China: opportunities and challenges. Journal of Environmental Science (China) 39, 86-96.
- Maheshwari, K. & Agrawal, M. 2020 Advances in capacitive deionization as an effective technique for reverse osmosis reject stream treatment. Journal of Environmental Chemical Engineering 8 (6), 104413.
- Marchetti, P., Peeva, L. & Livingston, A. 2017 The selectivity challenge in organic solvent nanofiltration: membrane and process solutions. Annual Review of Chemical and Biomolecular Engineering 8, 473-497.
- Metke, T., Westover, A. S., Carter, R., Oakes, L., Douglas, A. & Pint, C. L. 2016 Particulate-free porous silicon networks for efficient capacitive deionization water desalination. Scientific Reports 6, 24680.
- Mo, H., Tang, Y., Chen, Y. & Wan, P. 2017 Technical research in continuous desalting by coupling flow capacitive deionization and electrodialysis. Modem Chemical Industry **39** (5), 91–95. (in Chinese).
- Naushad, M., Mittal, A., Rathore, M. & Gupta, V. 2015 Ionexchange kinetic studies for Cd(II), Co(II), Cu(II), and Pb(II) metal ions over a composite cation exchanger. Desalination and Water Treatment 54 (10), 2883-2890.
- Oatley-Radcliffe, D. L., Walters, M., Ainscough, T. J., Williams, P. M., Mohammad, A. W. & Hilal, N. 2017 Nanofiltration membranes and processes: a review of research trends over the past decade. *Journal of Water Process Engineering* **19**, 164–171.

- Park, G., Hong, S. P., Lee, C., Lee, J. & Yoon, J. 2021 Selective fluoride removal in capacitive deionization by reduced graphene oxide/hydroxyapatite composite electrode. Journal of Colloid and Interface Science 581 (A), 396-402.
- Sami, S. K., Seo, J. Y., Hyeon, S., Shershah, M. S. A., Yoo, P. & Chung, C. 2018 Enhanced capacitive deionization performance by an rGO-SnO2 nanocomposite modified carbon felt electrode. RSC Advances 8 (8), 4182-4190.
- Shi, W., Li, H., Cao, X., Leong, Z. Y., Zhang, J., Chen, T., Zhang, H. & Yang, H. Y. 2016 Ultrahigh performance of novel capacitive deionization electrodes based on a threedimensional graphene architecture with nanopores. Scientific Reports 6, 18966.
- Shi, L., Xiao, L., Hu, Z. & Zhan, X. 2020 Nutrient recovery from animal manure using bipolar membrane electrodialysis: study on product purity and energy efficiency. Water Cycle 1, 54-62.
- Sistat, P., Huguet, P., Ruiz, B., Pourcelly, G., Mareev, S. A. & Nikonenko, V. V. 2015 Effect of pulsed electric field on electrodialysis of a NaCl solution in sub-limiting current regime. Electrochimica Acta 164, 267-280.
- Suss, M. E., Porada, S., Sun, X., Biesheuvel, P. M., Yoon, J. & Presser, V. 2015 Water desalination via capacitive deionization: what is it and what can we expect from it? Energy & Environmental Science 8 (8), 2296-2319.
- Takeuchi, H. & Tanaka, H. 2020 Water reuse and recycling in Japan - history, current situation, and future perspectives. Water Cycle 1, 1-12.
- Tang, W., Liang, J., He, D., Gong, J., Tang, L., Liu, Z., Wang, D. & Zeng, G. 2019 Various cell architectures of capacitive deionization: recent advances and future trends. Water Research 150, 225-251.
- Wang, S., Wang, S., Wang, G., Che, X., Li, D., Li, C. & Qiu, J. 2020 Ion removal performance and enhanced cyclic stability of sno2/CNT composite electrode in hybrid capacitive deionization. Materials Today Communications 23, 100904.
- Wu, G., Yin, Q., Guo, F. & Hu, Z. 2020 Slow growers possess a high pollutant removal potential through granule formation for wastewater treatment. Water Cycle 1, 63-69.
- Xu, A., Wu, Y., Chen, Z., Wu, G., Wu, Q., Ling, F., Huang, W. E. & Hu, H. 2020 Towards the new era of wastewater treatment of China: development history, current status, and future directions. Water Cycle 1, 80-87.

- Yan, T., Xu, B., Zhang, J., Shi, L. & Zhang, D. 2018 Ion-selective asymmetric carbon electrodes for enhanced capacitive deionization. RSC Advances 8 (5), 2490-2497.
- Yang, S., Kim, H., Jeon, S., Choi, J., Yeo, J., Park, H., Jin, J. & Kim, D. K. 2017 Analysis of the desalting performance of flowelectrode capacitive deionization under short-circuited closed cycle operation. Desalination 424, 110-121.
- Yasin, A. S., Mohamed, I. M. A., Mousa, H. M., Park, C. H. & Kim, C. S. 2018 Facile synthesis of TiO<sub>2</sub>/ZrO<sub>2</sub> nanofibers/nitrogen co-doped activated carbon to enhance the desalination and bacterial inactivation via capacitive deionization. Scientific Reports 8 (1), 541.
- Yu, F., Wang, C. & Ma, J. 2016 Applications of graphene-modified electrodes in microbial fuel cells. Materials (Basel) 9 (10), 807.
- Yu, Z., Yu, Z., Zheng, D., Zheng, D., Zhang, K., Zhang, K., Yang, T., Yang, T., Chen, Y., Chen, Y., Li, X. & Li, X. 2017 Optimally catalyzed porous-silicon electrode of self-breathing micro fuel cells. Microsystem Technologies 23 (8), 3257-3262.
- Zhang, C., He, D., Ma, J., Tang, W. & Waite, T. D. 2018 Faradaic reactions in capacitive deionization (CDI) - problems and possibilities: a review. Water Research 128, 314-330.
- Zhang, X., Zuo, K., Zhang, X., Zhang, C. & Liang, P. 2020 Selective ion separation by capacitive deionization (CDI) based technologies: a state-of-the-art review. Environmental Science Water Research & Technology 6 (2), 243-257.
- Zhao, C., Zhang, L., Ge, R., Zhang, A., Zhang, C. & Chen, X. 2019a Treatment of low-level Cu(II) wastewater and regeneration through a novel capacitive deionization-electrodeionization (CDI-EDI) technology. Chemosphere 217, 763-772.
- Zhao, C., Ge, R., Zhen, Y., Wang, Y., Li, Z., Shi, Y. & Chen, X. 2019b A hybrid process of coprecipitation-induced crystallization-capacitive deionization-ion exchange process for heavy metals removal from hypersaline ternary precursor wastewater. Chemical Engineering Journal (Lausanne, Switzerland: 1996) 378, 122136.
- Zhao, X., Wei, H., Zhao, H., Wang, Y. & Tang, N. 2020 Electrode materials for capacitive deionization: a review. Journal of Electroanalytical Chemistry 873, 114416.
- Zuo, K., Kim, J., Jain, A., Wang, T., Verduzco, R. & Li, Q. 2018 Resin-modified capacitive deionization for selective uptake and removal of sulfate. Abstracts of Papers of the American Chemical Society 256, 419.

First received 31 October 2020; accepted in revised form 18 January 2021. Available online 25 February 2021