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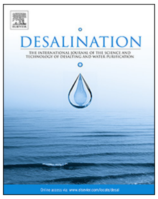

Versatile applications of capacitive deionization (CDI)-based technologies

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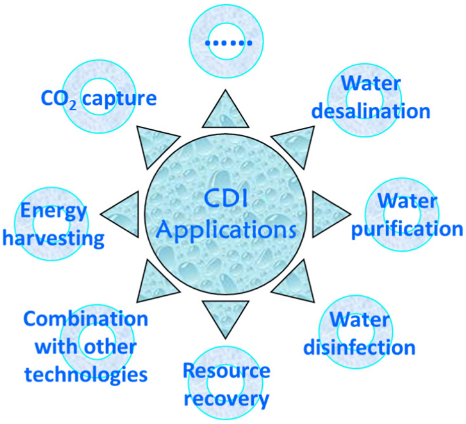
GRAPHICAL ABSTRACT



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ABSTRACT

Water pollution and freshwater scarcity are two of the most important environmental problems faced by human around the globe in the 21st century. Capacitive deionization (CDI), as a promising electrochemical water treatment technology, has attracted large attention over the past decade for the facile removal of ions from water with the advantages of environmental friendliness, cost effectiveness, low energy consumption, and convenient electrode regeneration. Enormous progress has been made in the CDI research field and now CDI encompasses various cell architectures assembled with either capacitive electrodes or battery electrodes. These scientific advances are accompanied by a diverse application of CDI-based technologies. This work is intended to summarize the versatile applications of CDI and highlight the representative achievements in each of the applications primarily covering water desalination, water purification, water disinfection, resource recovery and synergistic combination with other technologies. Emerging application areas of CDI such as energy harvesting and CO$_2$ capture are also presented. Lastly, the challenges and future outlook for CDI practical applications are discussed. This work should be of value in promoting CDI-based technologies to develop into a competitive option for coping with multiple water or energy-related issues.

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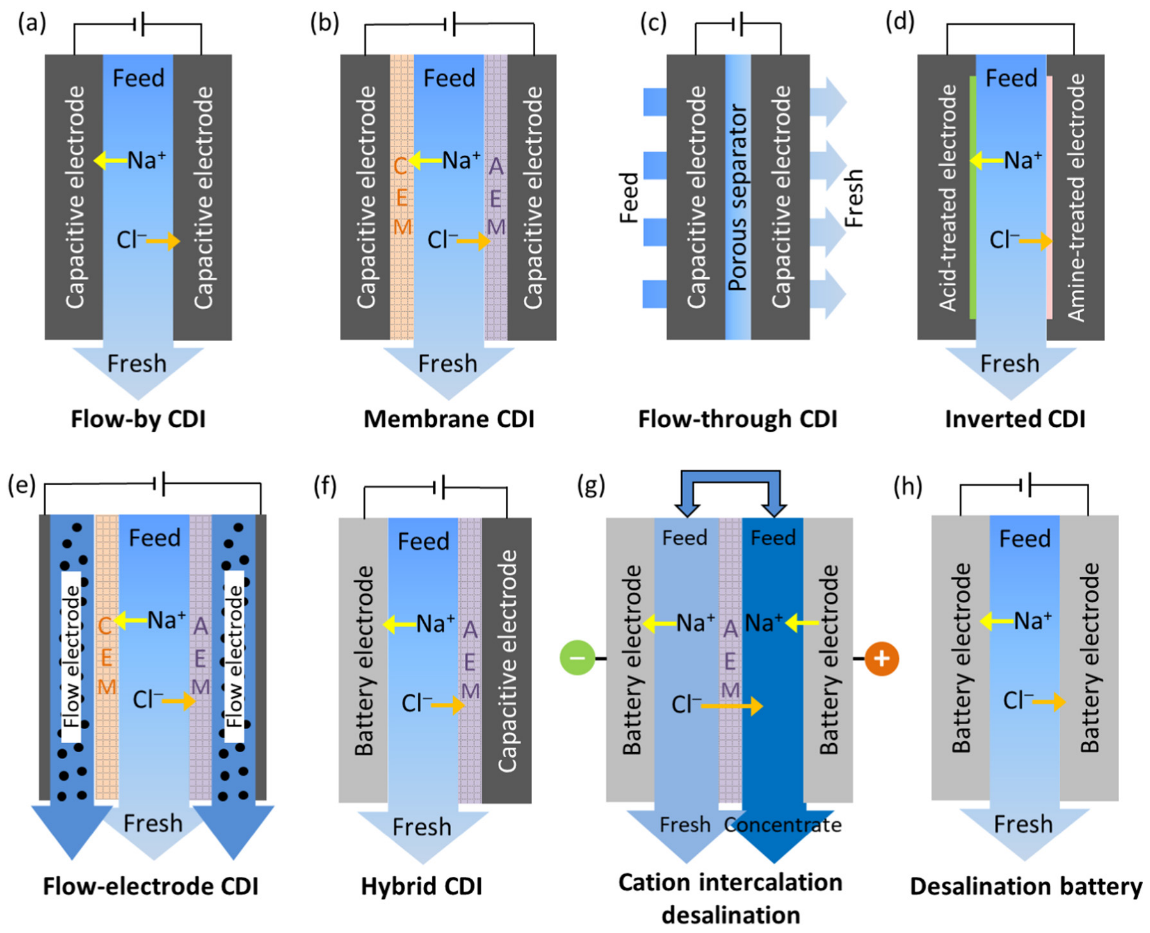

Fig. 1. Typical cell architectures of various CDI: (a) flow-by CDI, (b) Membrane CDI, (c) flow-through CDI, (d) inverted CDI, (e) flow-electrode CDI, (f) Hybrid CDI, (g) cation intercalation desalination, and (h) desalination battery.
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1. Introduction

With the rapid industrial development, increasing human population and active agricultural activities in modern society, water pollution and freshwater scarcity are becoming two of the key global environmental challenges of the 21st century [1–5]. With primary advantages of environmental friendliness, cost effectiveness, low energy consumption, and convenient electrode regeneration, capacitive deionization (CDI) has emerged as a promising method to tackle the above two issues because it can achieve the facile removal of undesired ion species from various water sources [3,5–15]. CDI operates by adsorbing ions in the electric double layers (EDLs) of capacitive electrodes via electrostatic interaction or in the crystal structures of battery electrodes via Faradaic intercalation through the application of a limited voltage (usually $\leq$1.23 V avoiding water electrolysis) [12,16]. The most conventional CDI cell configuration, also called flow-by CDI, consists of a pair of oppositely charged porous carbon electrodes (a positive electrode and a negative electrode) enabling capacitive ion sorption and a separator in-between preventing short circuit and allowing the solution to flow (see Fig. 1a) [6]. The applied potential difference generates an electric field that causes ions to migrate towards the electrodes and then be retained in the EDLs, thereby producing a diluted stream. Saturated electrodes can be regenerated by desorbing ions either by applying reverse voltage or short-circuiting the electrodes with the release of ions producing a concentrated stream. In parallel with ion desorption, the charge invested for ion removal would be also released, and there is a possibility of energy recovery. When feed water flows straight through the electrodes rather than between the electrodes, this cell configuration is named flow-through CDI (see Fig. 1c).

Though first proposed in the 1960s, CDI has gone through a slow initial development and, until about ten years ago, began to attract extensive attention with currently > 200 publications annually reporting on the topic [17]. This recent rapid growth has been motivated by the substantial innovations in the electrode and membrane materials, the employment of new operational modes, the development of theoretical models enabling parameter optimization and performance prediction, a deepened understanding of the complicated processes related to ion removal and interfacial electrochemistry, and, particularly, a fast proliferation of novel CDI cell architectures [18–26]. Nowadays, in addition to flow-by CDI and flow-through CDI, CDI encompasses membrane CDI (MCDI), inverted CDI, flow-electrode CDI (FCDI), hybrid CDI (HCDI), desalination battery and cation intercalation desalination (CID). With similar configuration to flow-by CDI, MCDI introduces ion exchange membranes (IEMs) between the electrodes and spacer channel to avoid the co-ion effect and to improve the charge efficiency (see Fig. 1b). Through chemical modification of flow-by CDI electrodes, the novel CDI (also called i-CDI) composed of a positive electrode with net negative surface charge and a negative electrode with net positive surface charge shows opposite adsorption-desorption behavior that ions are desorbed from the electrode EDLs during charging while ions are electrosorbed during discharging (Fig. 1d) [27,28]. A significant novel improvement based on MCDI is the FCDI using a flowable carbon suspension, rather than a solid porous carbon electrode (Fig. 1e). The FCDI cell where the flow electrode can be pumped through electrode compartments and flows between the IEMs and the corresponding current collectors is capable of providing

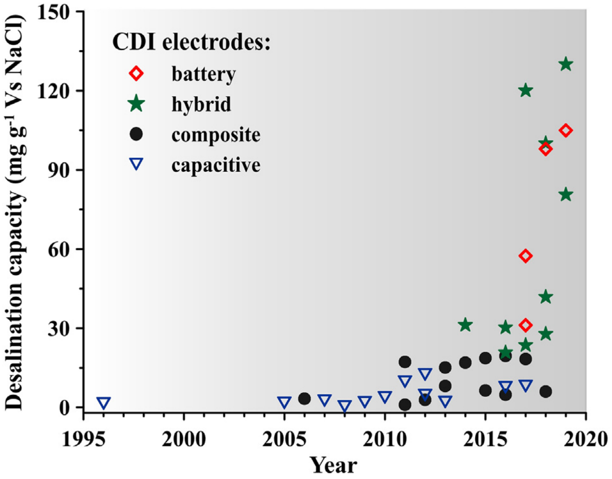

Fig. 2. Historical evolution of maximum desalination capacity for capacitive, composite, hybrid and battery CDI electrode [5,32,54,64,72–106].

continuous removal of high-salinity feed water with no need for a typical discharging step [29–31]. Cells of flow-by CDI, MCDI, flow-through CDI, i-CDI and FCDI remove ions via the mechanism of non-Faradaic process (i.e., electrostatic and capacitive effects) since they consist of capacitive electrodes [17]. A great innovation is introducing the battery electrodes to CDI to remove ions via the mechanism of Faradaic process (i.e., reversible redox reactions), which gives rise to the cells of HCDI, desalination battery and CID. The HCDI cell is normally composed of a battery electrode for cation intercalation/deintercalation, a capacitive electrode for anion adsorption/desorption and an anion exchange membrane (AEM) adjacent to the capacitive electrode [32] (Fig. 1f). The desalination battery cell consists of two kinds of battery electrodes, one for cation intercalation/deintercalation reactions and the other for anion intercalation/deintercalation reactions [33] (Fig. 1h). The CID cell is composed of symmetric or asymmetric Faradaic cation intercalation electrodes with the electrodes separated by an AEM [34] (Fig. 1g).

These aforementioned various CDI cell architectures and their corresponding scientific advances are accompanied by a diverse application of CDI-based technologies, including water desalination, water purification, resource recovery, water disinfection, synergistic combination with other technologies, energy harvesting, CO_2 capture, etc.. A number of reviews regarding CDI electrode materials and CDI cell architectures have been already published [17,35,36]. However, a state-of-the-art review that summarizes the versatile applications of CDI is still missing in the literature. Therefore, in this work, we provide a comprehensive overview of CDI applications for various objectives, from its initial applications through to the present day. The representative achievements in each of the applications are highlighted, and the challenges and future prospect for CDI practical applications are discussed.

2. Water desalination

2.1. Brackish water desalination

Plenty of saline water on earth provides a feasible solution to the problem of freshwater scarcity with the development of desalination technologies. Reverse osmosis and thermal distillation are currently the dominant commercial technologies for water desalination. Nevertheless, such technologies are not suitable for treatment of low salinity streams such as brackish water (roughly 0.6–3 g L^{-1} TDS) due to their relatively high energy cost and requirement of subsequent remineralization for human consumption [3,6,12]. For brackish water desalination, it can be advantageous to use CDI technology which has the potential to be energy-efficient as CDI focuses on the removal of the minority salt ions from the feed water rather than extracting the majority water molecules from the salt solution, and moreover, enables salt removal at room temperatures, low pressures and low voltages with additional possibility of energy recovery [3,6]. Although CDI is attractive for the desalination of brackish water which, in the lab, could be prepared with deionized water (HHitech, China), there is great room for improvement. Extensive studies have been conducted to enhance the brackish water desalination performance of CDI and maintain the performance stability since the 1990s when the interest in CDI revived with research efforts centering around three main aspects, i.e., synthesis of superior electrode materials, development of new operational modes, and design of novel cell configurations.

Synthesis of new electrode materials and their chemical modification for higher adsorption capacity and improved longevity has always been a major CDI research area. It is estimated that 60% of efforts devoted to CDI field have been put into the development of superior electrode materials [37]. Many kinds of single carbon capacitive materials like carbon aerogels [38], activated carbon [39], activated carbon cloth [40], carbon nanofibers [41], carbide-derived carbons [42], mesoporous carbon [43,44], carbon nanotubes [45], graphenes [46], and metal-organic framework-derived carbons [47,48] have been investigated, and the CDI electrodes composed of these materials exhibited relatively low salt adsorption capacity (SAC) with the maximum SAC below 15 mg g^{-1}, as shown in Fig. 2. Tuning the surface properties of carbon materials such as heteroatom (e.g., nitrogen, sulfur, phosphorus, and boron) doping and functional group grafting are feasible methods for the capacity enhancement by providing good wettability/conductivity and/or high specific capacitance [17,49–52]. The next-generation electrode materials are carbon-based composites (carbon-carbon composites, carbon-metal oxide composites, carbon-polymer composites and carbon-polymer-metal oxide composites) [53,54]. By taking advantage of the merits of carbon and other constituent materials, carbon-based composites allow for significantly higher SAC than single carbon capacitive materials [55]. The additional components in the composite can potentially enhance the desalination performance of pristine carbon materials via adjusting the intrinsic pore structure, inhibiting the aggregation tendency, improving the surface chemistry, increasing the wettability, adding the pseudocapacitance or altering the zeta potential [36,56]. Typical metal oxides incorporated into carbon materials include TiO_2 [5,57], MnO_2 [58,59], ZnO [60], CeO_2 [5], SnO_2 [61], ZrO_2 [53], Fe_2O_3 [5], and Fe_3O_4 [62] while typical polymers incorporated into carbon materials include polyaniline, polypyrrole, chitosan and ion-exchange resin [54,63]. The recent utilization of Faradaic ion intercalation materials (or say, battery materials) in CDI electrodes allows for boosts in SAC to over 30 mg g^{-1}. Battery electrodes offer a higher SAC than capacitive electrodes because ions are intercalated into their crystal structures via chemical bonds, analogous to the higher energy density achieved by battery electrodes than electric double-layer capacitor electrodes in energy storage systems [16,32]. A carbon-coated battery material as a composite electrode can address the drawbacks of pure battery materials (low electroconductivity and slow ion diffusion rates), thereby enabling a further higher SAC [64]. Instead of pursuing a higher SAC, several groups have focused efforts on improving the long-term performance of electrode materials. One route is to chemically treat the electrodes making the anode carry net negative surface charge and the cathode carry net positive surface charge (i.e., i-CDI) [28]. In this case, the i-CDI exhibited excellent operation longevity despite the key disadvantage of a low SAC. A second strategy is to introduce a thin layer of ion-exchange polymers onto the carbon electrodes to mitigate anode corrosion [65,66]. The SAC and charge efficiency can be also increased because the thin layer of ion-exchange polymers acts similar to the IEMs. Here, it is worth pointing out that, while the SAC and lifetime at given operating conditions might be critical parameters in evaluating the performance of an electrode, their implication on practical CDI operations is limited. Other performance metrics have emerged to describe the desalination process, including average salt adsorption rate, charge and coulombic efficiencies, water recovery, concentration reduction, specific energy consumption [67–71].

Unlike electrode materials mainly for a higher SAC, development of new operational modes is mainly aimed at improving other performance metrics such as water recovery, energy consumption and concentration reduction [17]. Customarily, there are two kinds of designs with regard to fluid flow (batch mode and single-pass mode), two kinds of charging modes for the adsorption step (constant voltage and constant current), and three kinds of discharging modes for the desorption step (short circuit, reverse constant-voltage and reverse constant-current). Benefiting from the creative ideas of many researchers, now there are a variety of new operational methods. For example, in the aspect of fluid flow, a brine stream was utilized that is more concentrated than the feed solution to regenerate the CDI unit during desorption and found that this significantly increased the energy efficiency and/or water recovery [107,108]. A new operating scheme based on a variable flowrate (flowing at a nominal flowrate during charging followed by a decrease in flowrate during discharging) was introduced and the results showing that water recovery can be readily increased to very high values at minimal additional performance reductions compared to conventional constant flowrate operation [26]. In the aspect of electricity, Kim et al. described a new discharging modes for the desorption step, that is, applying a discharging voltage smaller than the charging voltage but with the same polarity, whereby the charge efficiency of a flow-by CDI cycle could be increased to nearly 100% with only a small loss of SAC [109].Time-varying voltage/current waveforms (sinusoidal voltage/current) were developed for flow-by CDI operation and this strategy could simultaneously achieve high salt removal and strong energy performance, which is impossible for the conventional CDI operating methods [110].

Designing novel cell configurations is the third main research effort towards improving the brackish water desalination performance of CDI. Considering that planar CDI electrodes exhibit poor energetic performance due to the resistance associated with salt depletion and tortuous diffusion in the macroporous structure, Bhat et al. etched macroporous patterns along the length of activated carbon porous electrodes in a flow-by CDI cell and demonstrated that the use of bi-tortuous electrodes is a novel and effective approach of reducing impedance to ionic flux in CDI which translates to enhanced salt adsorption capacity and rate (Fig. 3) [111]. When feed water flows straight through the electrodes rather than between the electrodes, flow-through CDI could be established. The main advantage of flow-through CDI over flow-by CDI is that, since the separator does not act as a major channel for fluid flow, the separator thickness in the flow-through CDI may be minimized on the premise that no short circuit occurs, thereby allowing for more compact cells, improved desalination rates and a higher reduction in salt concentration of the feed per unit of charge. However, it should be also noted that flow-through CDI suffers from a greater feed pressure and a faster oxidation of the positive electrodes than flow-by CDI. Incorporation of IEMs in the flow-by CDI to form MCDI is recognized to be an effective method to improve the flow-by cell's salt removal efficiency and charge efficiency. A further enhancement of the desalination performance is achievable via packing the MCDI cell's flow chamber with ion-exchange resin granules, granular activated carbons or activated carbon fibers [112]. It is worth mentioning that, recently, a very novel multichannel MCDI (MC-MCDI) system was fabricated with two high-molar flow streams (e.g., 1000 mM) to control the environment around the electrodes and a middle channel for brackish water (e.g., 5 mM) desalination, as shown in Fig. 4 [113]. This MC-MCDI cell configuration exhibited an excellent SAC up to 56 mg g^{-1} in the middle channel concurrently with a high charge efficiency close to unity and a low energy consumption. Other great novel cell architectures such as FCDI, HCDI, CID and desalination battery enable unprecedented SAC and will be described in detail in the next section of seawater desalination.

2.2. *Seawater desalination*

While conventional CDI such as flow-by CDI and MCDI is highly efficient for brackish water desalination, it struggles with feed water of higher salinity such as seawater because of its limited ion adsorption capacity and the higher energy requirement than that of reverse osmosis for treating highly salty water. The recent advent of FCDI, HCDI, desalination battery and CID cells makes it possible for the application area of CDI to extend to seawater desalination.

The FCDI cell arises from the modification of MCDI cell through the use of flowing electrodes made of carbon suspension instead of the stationary electrodes between current collectors and IEMs (Fig. 1e), and was first proposed by Jeon et al. in 2013 [114]. FCDI is able to perform the desalination of high-salinity feed water because of the infinite ion adsorption capacity of the flow-electrode achieved via the continuous inflow of fresh uncharged carbon electrodes [115,116]. Meanwhile, FCDI enables a continuous salt removal and a steady production of desalinated water with electrode regeneration completed downstream of the cell by simply mixing the positive and negative carbon slurries followed by carbon separation from water via settling [29,117]. Despite the great prospect of FCDI in the application of seawater desalination, there is long way to go to realize FCDI's true commercialization and a lot of efforts are required to further improve FCDI's desalination

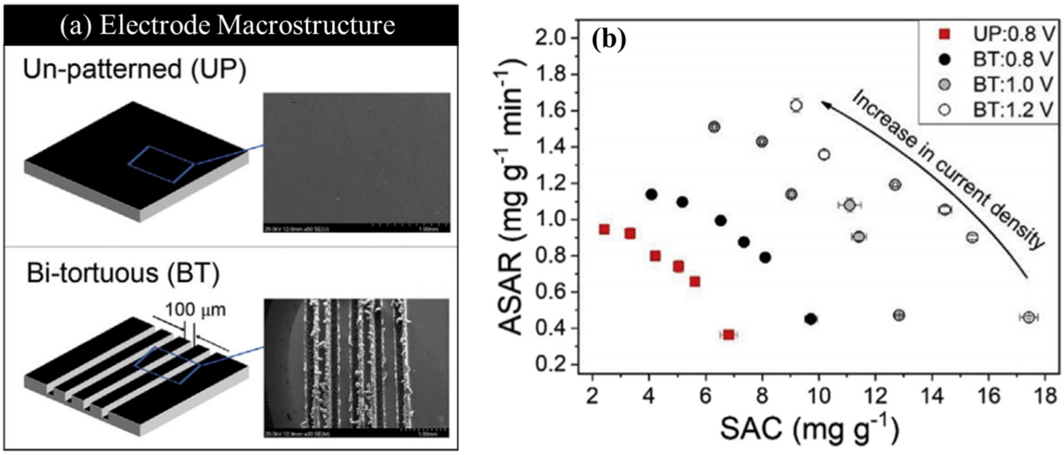

Fig. 3. (a) Graphical and SEM images of view of the un-patterned (UP) and bi-tortuous (BT) electrode; (b) Kim-Yoon plot for un-patterned electrodes at a voltage cut-off of 0.8 V and bi-tortuous electrodes at voltage cut-offs of 0.8, 1.0 and 1.2 V.
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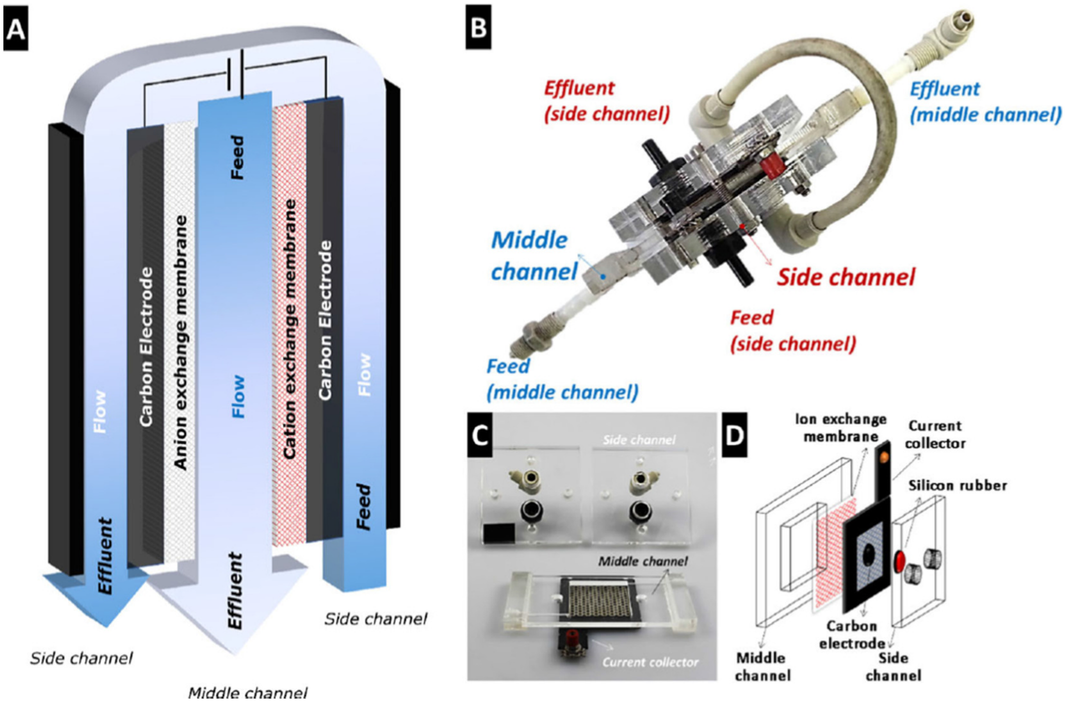

Fig. 4. Multichannel membrane CDI (MC-MCDI) cell using channels and membranes: (A) scheme, (B) top view, (C) components, and (D) exploded view of the cell. Reproduced with permission from Ref. [113]. Copyright 2017 Wiley Online Library.

performance [118]. In view of the inefficient charge transfer between the current collectors and flow electrodes as a result of the relatively low conductivity of the flow electrodes compared to that of the solid electrodes, researchers have suggested several methods to solve this limitation, including appropriately increasing the carbon mass loading or electrolyte concentration in the slurries to enhance the flow-electrode conductivity, performing chemical surface treatment of carbon particles to lower the viscosity of carbon suspensions, leveraging up-flow fluidized bed electrodes to raise the maximum allowable carbon loading to 35 wt%, utilizing activated carbon (AC)/MnO_2 composite suspension as the anode and AC suspension as the cathode to increase the operation voltage to 1.8 V, and introducing additives such as carbon black or redox couples to reduce the internal resistance of the electrode chambers, and using a titanium mesh-membrane assembly as the current collector to greatly reduce the charge transport distance [17,30,119,120]. A most recent innovation indicates that a significant increase in salt removal performance without sacrificing the flowability could be achieved by adding a very small amount of functionalized carbon nanotubes (FCNTs) into AC-based flow electrodes as it leads to the formation of conducting bridges between dispersed AC particles thereby facilitating electron transport, as shown in Fig. 5 [121]. Scaling-up FCDI systems with lower costs and simpler process is also the key to promote the application of FCDI in seawater desalination [122]. Cho et al. realized a highly compact and scalable three-dimensional (3D) FCDI desalination system by utilizing honeycomb-shaped porous lattice scaffolds and by coating an IEM (AEM or CEM solution) and a graphene layer within appropriate channels, as schematically shown in Fig. 6 [123]. Since it consists of a simple and repetitive unit cell with no need for a water-channel spacer layer and free-standing IEMs, the novel 3D FCDI system offers distinct advantages of high space utilization and low construction investment. Additionally, the unit cell can be readily scaled-up in three dimensions and the desalination efficiency and capacity significantly increase as the cell size is expanded (Fig. 6d). While its desalination performance is somewhat lower than intensively studied conventional FCDI systems for now, the 3D FCDI cell opens a new door in the field of desalination research and there are plenty of opportunities for further improvement making it more suitable for seawater desalination application.

HCDI, desalination battery and CID can be categorized as battery-based desalination techniques as one or two electrodes of these cells consist of battery materials [124,125]. Battery electrodes remove ions via the mechanism of Faradaic process (i.e., reversible redox reactions), and can provide higher capacities for desalination than capacitive electrodes due to their abilities to store salt ions within the structures in crystallographic sites or between atomic planes, and they can reduce energy consumption and avoid parasitic reactions through the use of a smaller voltage window [16,32]. HCDI is composed of one capacitive electrode and one battery electrode. Lee et al. first introduced the concept of HCDI by using a $Na_4Mn_9O_{18}$ cathode for sodium-ion Faradaic insertion/release and an activated carbon anode for chloride-ion capacitive adsorption/desorption, and obtained a desalination capacity of 31.2 mg g^{-1} [32]. Subsequently, many other Na + Faradaic electrode materials, such as $Na_3V_2(PO_4)_3$ [126], $Na_2FeP_2O_7$ [73], $Na_{0.7}MnO_2$ [127], α-MnO_2 [128], RuO_2 [129], Co_3O_4 [130], V_2O_5 [74], $Na_4Ti_9O_{20}$ [64,76], $NaTi_2(PO_4)_3$ [131], Prussian blue [75,106], MOF ($K_{0.03}Cu[Fe(CN)_6]_{0.65}\cdot 0.43H_2O$) [132], TiS_2 [133], $Li_4Ti_5O_{12}$ [134], $FePO_4$ [77], MoS_2 [135], and a few Cl^- Faradaic electrode materials such as Ag/AgCl [33] and Bi/BiOCl [80,136] have been applied in desalination and have achieved good results with a desalination capacity as high as 167.4 mg g^{-1} reported [137]. Here it should be noted that most of these reported battery electrode materials need to be coated by carbon materials (e.g., activated carbon, carbon nanofibers, mesoporous carbon, carbon nanotubes, graphene) to form composite electrode because pure Faradaic electrode materials have shortcomings of low electronic conductivity, poor ion diffusion coefficient and relatively high capital cost. The typical carbon coating method is to form a core-shell heterostructure. One route is coating carbon materials onto the surface of Faradaic electrode materials. Zhao et al. successfully utilized the solvothermal method to develop a quasi-one-dimensional wire structure of $C@Na_3V_2(PO_4)_3$ realizing an ion conductor network in electrodes and achieving superior desalination performance, rate capability, and cycle capability (Fig. 7) [81]. Another route is coating Faradaic electrode materials onto the surface of carbon materials. Shi et al. fabricated an excellent electrode structure using atomic layer

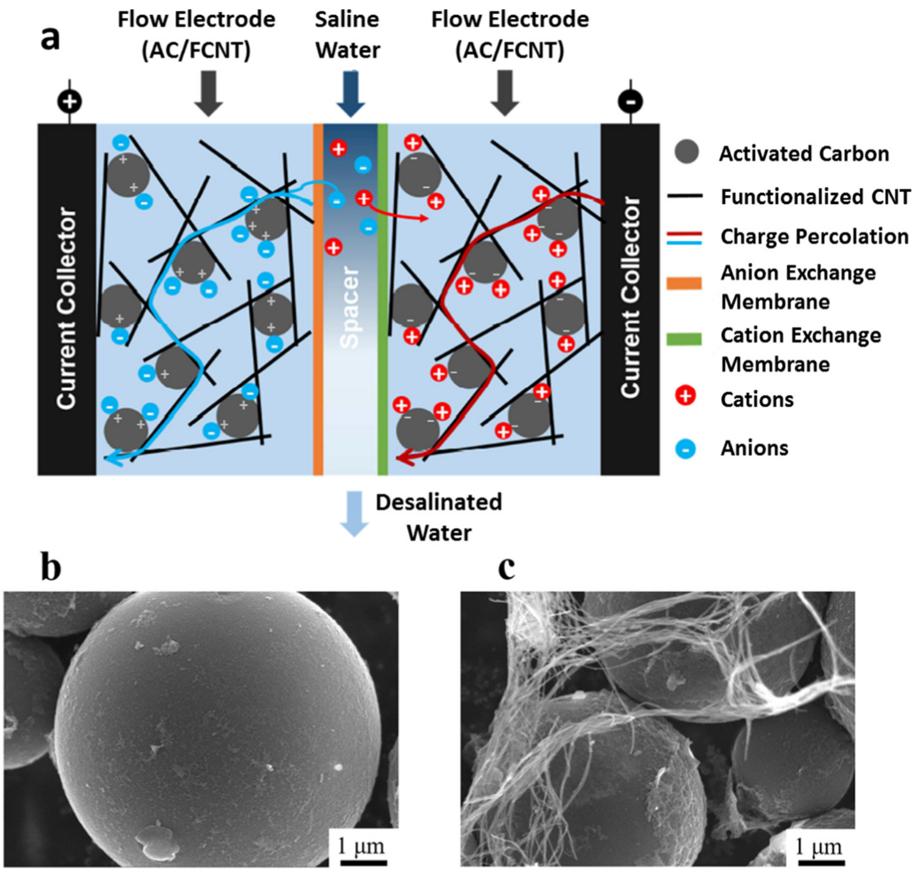

Fig. 5. (a) Schematic of flow-electrode capacitive deionization (FCDI) process using activated carbon (AC) with functionalized carbon nanotube (FCNTs). Field emission scanning electron microscopy (FE-SEM) images of (b) AC and (c) AC with FCNTs with a high aspect ratio (~50,000).
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deposition (ALD) to coat thin films of MnO_x onto vertically aligned carbon nanotubes (Fig. 8) [138]. The inherently hierarchical, anisotropic, 3D macroporous structure of vertically aligned carbon nanotubes as well as the tunable coating endowed the electrode with outstanding specific capacitance with respect to both mass and geometric area. Desalination battery comprises two different battery electrodes, one for cation Faradaic intercalation/de-intercalation and the other for anion Faradaic intercalation/de-intercalation. The first reported desalination battery cell is composed of a Ag negative electrode and a $Na_2Mn_5O_{10}$ positive electrode [33], and the following research about desalination battery is primarily focused on seeking novel combinations of Na^+ Faradaic electrode materials and Cl^- Faradaic electrode materials [79]. It turned out that desalination battery cells could also obtained excellent desalination capacity.

CID cells are fabricated through the use of Na^+ Faradaic electrode materials for both electrodes with the electrodes separated by an AEM. Either symmetric Na^+ intercalation electrodes (i.e., electrodes with identical chemical composition but with a different degree of intercalation) or asymmetric Na^+ intercalation electrodes (i.e., electrodes with different chemical composition) could be utilized. CID system enables a continuous production of desalinated solutions and also possesses a high desalination capacity with efficient energy consumption and good stability [139]. Anion intercalation desalination (AID), representing an opposite version of CID, has seldom studied because

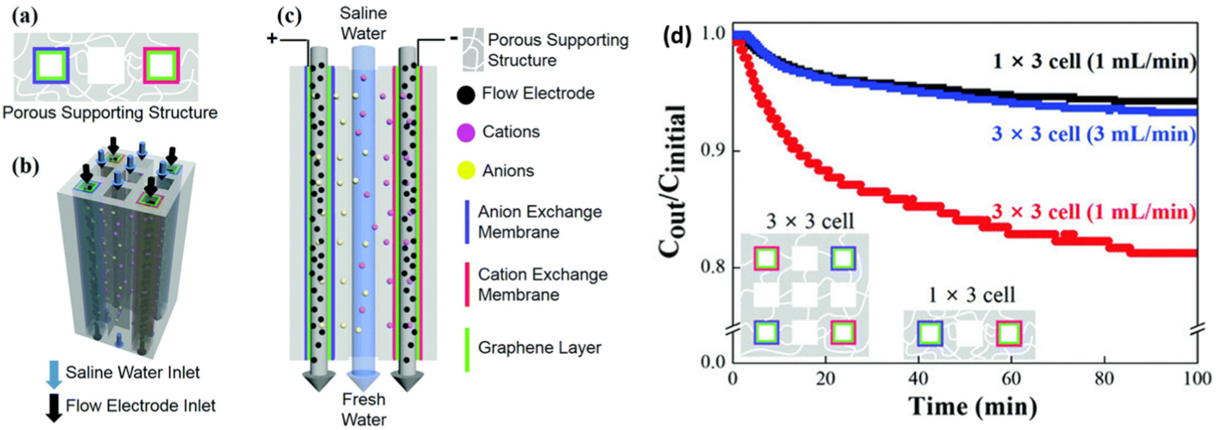

Fig. 6. Schematic diagram of the highly compact and scalable three-dimensional (3D) FCDI system based on honeycomb-shaped porous lattice scaffolds made out of cordierite: (a) top-view (1 × 3-channel cell), (b) 3 × 3-channel cell and (c) desalination process. (d) Variation in salt concentration of 1 × 3- and 3 × 3-channel cells during desalination operated in the batch mode.
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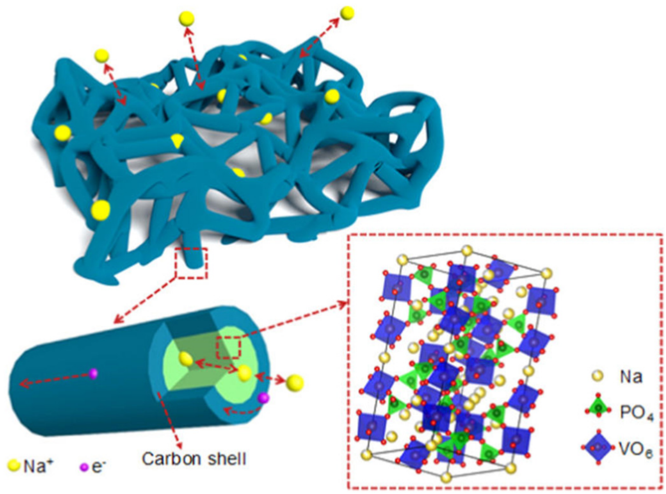

Fig. 7. Schematics of the C@$Na_3V_2(PO_4)_3$ wire ion conductor network and $Na_3V_2(PO_4)_3$ atomic structure.
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very few options are available for Cl^- Faradaic electrode materials. We believe that future development of HCDI, desalination battery, CID and AID devices will benefit from on-going research in the synthesis of new Na^+ or Cl^- Faradaic electrode materials, and the current outstanding desalination performance demonstrates their promising application for seawater desalination in the future.

3. Water purification

3.1. Removal of heavy metals

Heavy metal pollution is becoming a severe environmental issue with the rapid development of metallurgical and chemical manufacturing, mining and batteries. Due to the high toxicity, potential carcinogenicity and bioaccumulation of heavy metals, scientists have been seeking for effective methods to treat the water contaminated with heavy metals [140]. The successful utilization of CDI-based technologies in water desalination has attracted many researchers to explore their potential applications in the removal of heavy metals from polluted water bodies.

Until now, CDI-based technologies have been investigated for removal of various heavy metal ions including Zn(II) [141], Cu(II) [38,142,143], Fe(II) [142,144], Fe(III) [144,145], Pb(II) [45,146,147], and Cd(II) [144,148], Ni(II) [149], Pd(II) [150], V(V) [151], Cr(III) [148], Cr(VI) [39,152–155], Li(I) [156,157], As(III) [158,159], As(V) [155,160] and U(VI) [161,162]. Removal of heavy metals by CDI mainly relies on the mechanism of electrostatic interaction, whereas physicochemical adsorption and electrochemical reduction might also play important roles [163]. For example, Tang et al. found that, at a charging voltage of 1.2 V, 13% of the total adsorbed Zn^{2+} were retained via physi- or chemisorption while 87% of the total adsorbed Zn^{2+} were held via electrostatic interactions [141]. Among the aforementioned heavy metals, removal of the neutral species As(III) and U (VI) by CDI deserves attention. CDI is capable of serving as a technique for the removal of uncharged As(III) species owing to the combined effects of electrooxidation and electrosorption. As(III) can be anodically oxidized to less toxic As(V) followed by electrosorption of the in-situ formed As(V) onto the surface of the anode [12]. Generally, groundwaters enriched in uranium have circumneutral to alkaline pH values and high carbonate concentrations. Such conditions favor the formation

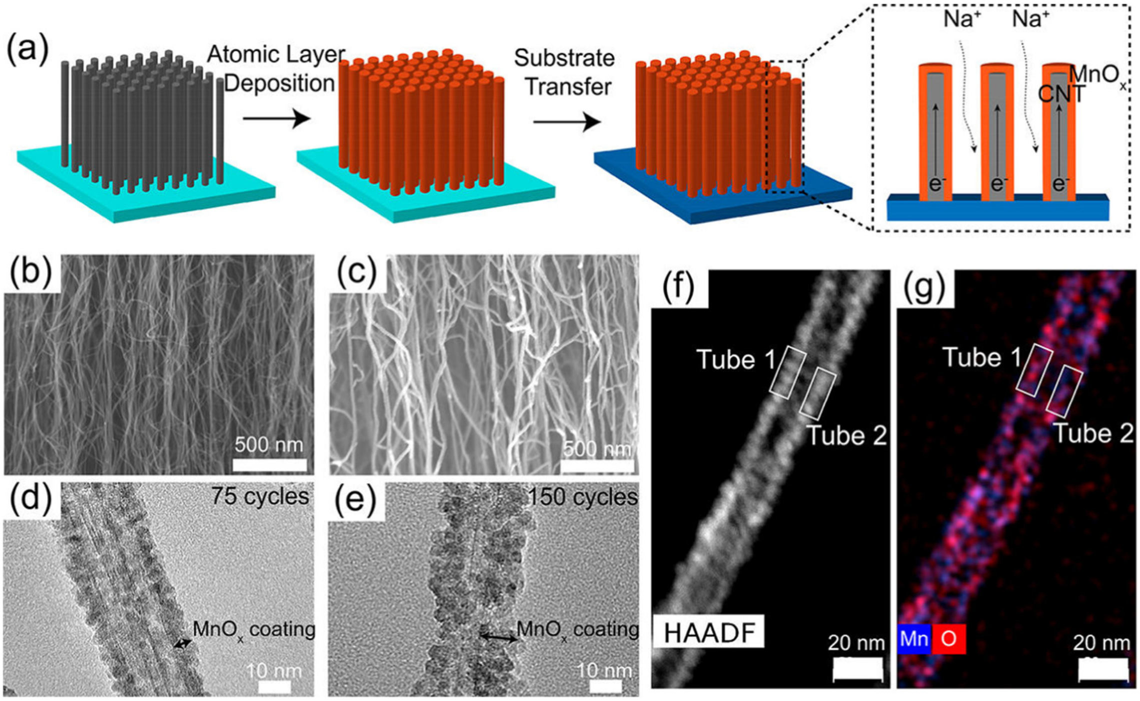

Fig. 8. (a) Schematic illustration of the MnO_x coating strategy on vertically aligned carbon nanotubes (VACNTs) via ALD and proposed model of electrosorption of ions on the coated VACNTs. (b) Cross-sectional SEM image demonstrating the alignment of pristine VACNTs. (c) Increase in VACNT diameter after 150 ALD cycles. (d and e) TEM images demonstrating the tunable MnO_x coating by changing the ALD cycle number at 75 and 150 cycles, respectively. (f) STEM image (high-angle annular dark-field imaging) of VACNT-MnO_x with 75 ALD cycles and (g) corresponding energy-dispersive X-ray spectroscopy elemental mapping confirming the presence of the MnO_x coating.
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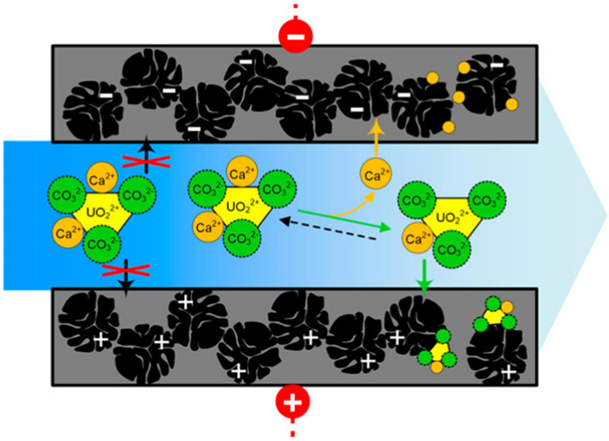

Fig. 9. Schematic showing removal of the uncharged $Ca_2UO_2(CO_3)_3{}^0$ ternary complex from brackish potable groundwater by FCDI.
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of thermodynamically stable ternary aqueous U(VI) complexes (e.g., $Ca_2UO_2(CO_3)_3{}^0$) that are neutrally charged. Ma et al. demonstrated that FCDI is versatile in reducing uranium concentrations to < 10 μg L^{-1} with low electrical consumption [161]. The workability of FCDI largely depends on the preferential removal of $Ca(UO_2)(CO_3)_3{}^{2-}$in the electrical field resulting in the rapid re-equilibration between the uncharged and negatively charged species ensuring ongoing removal of uranium, as shown in Fig. 9. Additionally, it is worthwhile to point out that arsenic and chromium ionic species are amphoteric ions which are a strong function of electrolyte pH values. As the electrolyte pH values might fluctuate significantly during charging, removal processes of these ions are relatively intricate. Moreover, in typical environment scenarios, it is common for heavy metal ions to coexist with other electrolytes such as NaCl and natural organic matter, and the electrosorption capacity of heavy metal ions would be usually reduced in the presence of such coexisting substances due to the competitive electrosorption and/or fouling of carbon electrodes.

3.2. *Removal of inorganic contaminants*

Consumption of water with extreme hardness may be linked to an increased incidence of chronic kidney disease [164]. Additionally, treating hard water is crucial in reducing domestic and industrial problems such as pipe clogging, membrane fouling, lime scaling in heaters, and decrease in cleaning efficiency [165]. Water hardness is ascribed most often to the existence of calcium ion (Ca^{2+}) and magnesium ion (Mg^{2+}), and can be reduced by chemical precipitation, ion exchange and reverse osmosis (RO). However, these conventional technologies require either additional chemicals or consume intensive energy, and are, hence, expensive [165,166]. Considering that CDI is a chemical-free and cost-effective technology enabling facile removal of ions, studies have been conducted to explore its prospect for water softening. Both He et al. and Shi and Lin demonstrated that the selective removal of Ca^{2+} over Na^+ can simply be achieved using MCDI and FCDI technologies primarily due to the presence of cation exchange membranes (CEMs) regulating the kinetics of ion transport [164,166]. Worthy of note is that, in MCDI, deposition of Ca^{2+} via the formation of $Ca(OH)_2$ on the carbon surface would quickly result in a decrease in available adsorption sites, leading to deterioration of the CDI performance [167]. FCDI operated in short-circuited closed-cycle (SCC) mode could alleviate calcium precipitation on the carbon surface as the continuous mixing of the carbon particles significantly inhibited pH excursion in the flowing electrodes, thereby making it an effective method for water softening. Yoon et al. tested calcium-alginate coated cathode for harness control, and found that flow-by CDI with the Ca-alginate coated-electrode showed superior calcium ion removal performance (44% more) than that of the conventional flow-by CDI, and MCDI with the Ca-alginate coated-electrode exhibited comparable performance to the conventional MCDI [165]. Herein, Ca-alginate acts as an appropriate alternative to the commercial CEM. In another study, Kim et al. coated a calcium selective nanocomposite (CSN, i.e., nano-sized calcium chelating resins with aminophosphonic groups in a sulfonated polyvinyl alcohol hydrogel matrix) layer on the cathode and achieved a Ca^{2+} over Na^+ selectivity of 3.5 to 5.4 at Na^+: Ca^{2+} equivalent concentration ratio from 10:1 to 1:1, 94–184% higher compared with the uncoated electrode [168]. Moreover, the CSN coated electrode exhibited excellent performance over long-term operation with highly stable SAC, Ca^{2+} selectivity and regenerability.

Apart from the removal of hardness ions, CDI-based technologies have also been applied for removal of other inorganic contaminants including sulfate [11], nitrate [10], fluoride [169], perchlorate [170], bromine [171], thiocyanate [172], ammonium [173], phosphate [174], and boron [175]. As real water and wastewater contain many background ions whose concentrations are usually significantly higher than those of target contaminants, selective separation remain one of the most important processes for water purification. Many studies were performed under the condition of multiple solute solutions to simulate the practical environmental scenarios, for example, investigating the removal of specific contaminants of concern in the presence of NaCl. Assuming that the initial molar concentrations of anion *i* and anion *j* are equal in the treated water, the intrinsic electrosorption selectivity of pristine carbon electrodes is attributed to the electrode functional groups as well as the different ion diffusion coefficient and ion charge number. The interaction between hydrated ionic radius and carbon nanoporous size also seems to play an important role as described in detail in Hawks et al.'s work [176]. Coating the surface of carbon electrodes with a polymer material which serves as an ion selective barrier, or synthesizing a composite electrode by mixing carbon particles with a material with high selectivity towards the specific ion of concern is beneficial to increasing the extent of preferential ion removal. As a key component in CDI cells assembled with IEMs, membrane might also play a significant role in the selective ion removal. With the technological advancements, recently, Kim et al. proposed a new approach, i.e., using two copper hexacyanoferrate (CuHCF) battery electrodes separated by an AEM to selectively remove the ions of ammonium from wastewaters [177]. The CuHCF battery electrodes removed $NH_4{}^+$ with a selectivity > 5 when operated at 0.1 V, despite the much higher initial Na^+ concentration (20 mM) than $NH_4{}^+$ (5 mM). In contrast, only negligible selective removal of $NH_4{}^+$ over Na^+ was observed when using nonselective electrodes or AEM. Furthermore, the carbon electrode coated with a thin film containing quaternary amine functionalized poly (vinyl alcohol) (QPVA) and $SO_4{}^{2-}$ selective resin was fabricated for selective removal of $SO_4{}^{2-}$ [178]. While a much higher selectivity of $SO_4{}^{2-}$ over Cl^- was achieved than that using commercial CDI device, the selectivity was much less than that the “solution-diffusion” model predicted. Thus, the specific transport mechanism in the QPVA and ion selective resin should be further studied. The phosphate (P) includes three ionic species: $PO_4{}^{3-}$, $HPO_4{}^{2-}$ and $H_2PO_4{}^-$, and in typical scenarios, the pH of the source water is circumneutral with P in this pH range mainly present in the form of $HPO_4{}^{2-}$ and $H_2PO_4{}^-$. The selective removal of P species over other competing ions such as chloride depends heavily on solution pH due to the complexity and variability of P species with pH [174,179,180]. Boron in the form of neutral boric acid cannot be removed by a regular CDI process, but boric acid could react with OH^- produced near the negative electrode to form borate anions which can be then electrosorbed to the positive electrode in CDI cells thereby achieving boron removal [175]. For water purification, concentrated streams containing a relatively high level of target contaminants would be inevitably produced during electrode regeneration. How to dispose of the

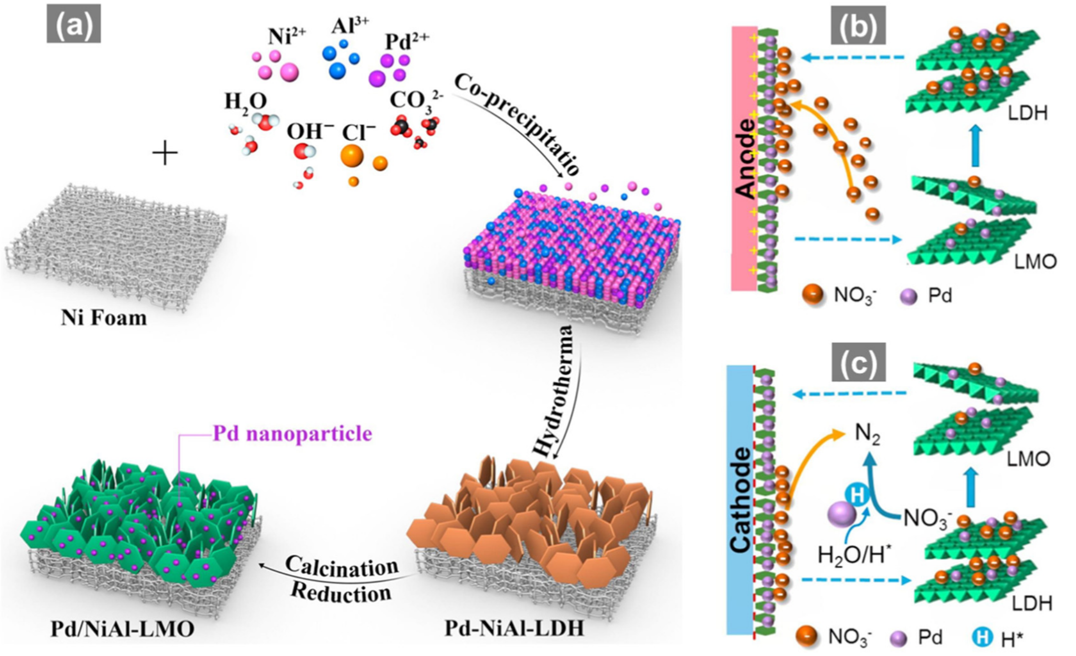

Fig. 10. (a) Schematic illustration of the fabrication of Pd/NiAl-LMO film electrode. Schematic diagram of (b) nitrate electro-sorption and (c) nitrate electro-reduction by Pd/NiAl-LMO electrode.
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concentrated streams appropriately is a challenge confronting CDI's applications. For instance, CDI has been demonstrated to be effective in removing nitrate from water. However, during desorption, nitrate would be discharged as a concentrated solution, which needs further complex treatment normally by denitrification to convert nitrate into harmless nitrogen gas before entering the environment. As a great innovation, Hu et al. successfully prepared a Pd/NiAl-layered metal oxide (Pd/NiAl-LMO) film electrode and used it to achieve an environmentally friendly outcome, that is, nitrate is electro-adsorbed by the Pd/NiAl-LMO electrode with concomitant reconstruction of the original hydrotalcite structure by the intercalation of NO_3^-, then the adsorbed nitrate is electro-reduced to N_2 by atomic H∗ in the electrode regeneration process, as shown in Fig. 10 [181].

3.3. Removal of organic contaminants

Beyond the removal of heavy metals and inorganic contaminants, CDI-based technologies have also been utilized for removal of organic contaminants such as ionizable antibiotics and dyes as well as carboxylate, sulfonate and phosphonate organic compounds. The representative works investigating the removal of these organic pollutants were selected and presented below.

(1) Ionizable antibiotics removal. Antibiotics have emerged as contaminants of global concern owing to their potential effects on human health and natural ecosystems once released into the environment, and many of the antibiotics currently used are ionizable at environmentally relevant pH values [182]. Wang et al. reported electro-assisted adsorption of three commonly used antibiotics, sulfadimethoxine, ciprofloxacin and clarithromycin, on activated carbon fibers in the continuous-flow mode and demonstrated the potential of electro-assisted adsorption in treatment of real antibiotic-contaminated waters [41]. Benefiting from electrostatic interactions between the contaminants and the adsorbent, the adsorption capacities for the target antibiotics increased by ~5 times under an applied voltage of 1.0 V compared to those of open circuit adsorption. The activated carbon fiber could be in-situ regenerated by reversing the polarities of the electrodes and remained a high recovery efficiency (~96%) after 5 cycles.

(2) Anionic or cationic dyes removal. Shi and Zhitomirsky fabricated N-doped activated carbon nanofibers (N-AC-NF) by carbonization and chemical activation of polypyrrole nanofibers and utilized the N-AC-NF electrodes for capacitive removal of triphenylmethane family of anionic dyes (pyrocatechol violet, eriochrome cyanine R) and chromotropic acid family of anionic dyes (chromotropic acid disodium salt, 2-(phenylazo) chromotropic acid disodium salt, 2-(1-naphthyldiazenyl) chromotropic acid disodium salt, 2,7-Bis(2-sulfophenylazo) chromotropic acid tetrasodium salt) from aqueous solutions [183]. The capacitive removal method offers advantages for the dyes of chromotropic acid family, e.g., 9.2 mass % eriochrome cyanine R and 18.3 mass % chromotropic acid disodium salt were removed during charging. For dyes of the same family, the higher charge to mass ratio and lower size of the molecules led to higher capacitance, lower impedance and higher power-energy characteristics. Chen et al. prepared novel mesoporous amorphous B–N–O–H nanofoams with high conductivity and large specific surface area [184]. The resulting B–N–O–H nanofoam electrodes exhibited a high adsorption capacity for cationic dyes, especially methylene blue (MB), with easy regeneration and well-controlled recycling behavior. The maximum MB adsorption capacity of the nanofoam obtained from the Langmuir isotherm equation was 3333 mg g^{-1}. Furthermore, the nanofoam also exhibited good selective separation of MB over other dyes.

(3) Removal of carboxylate, sulfonate and phosphonate organic compounds, etc. It is a great challenge for conventional adsorption or fixed-bed ion-exchange methods to treat complex organic ions that are highly toxic and present in the micromolar range, including pesticides, endocrine disruptors and pharmaceuticals, in the presence of excess competing salts. Redox-based electrochemical separation processes offers a platform for addressing these water purification and wastewater treatment needs through specifically designing chemical selectivity at the electrode interface. Su et al. have shown that redox electrodes functionalized with organometallic polymers, particularly PVF, can serve as an high-performance

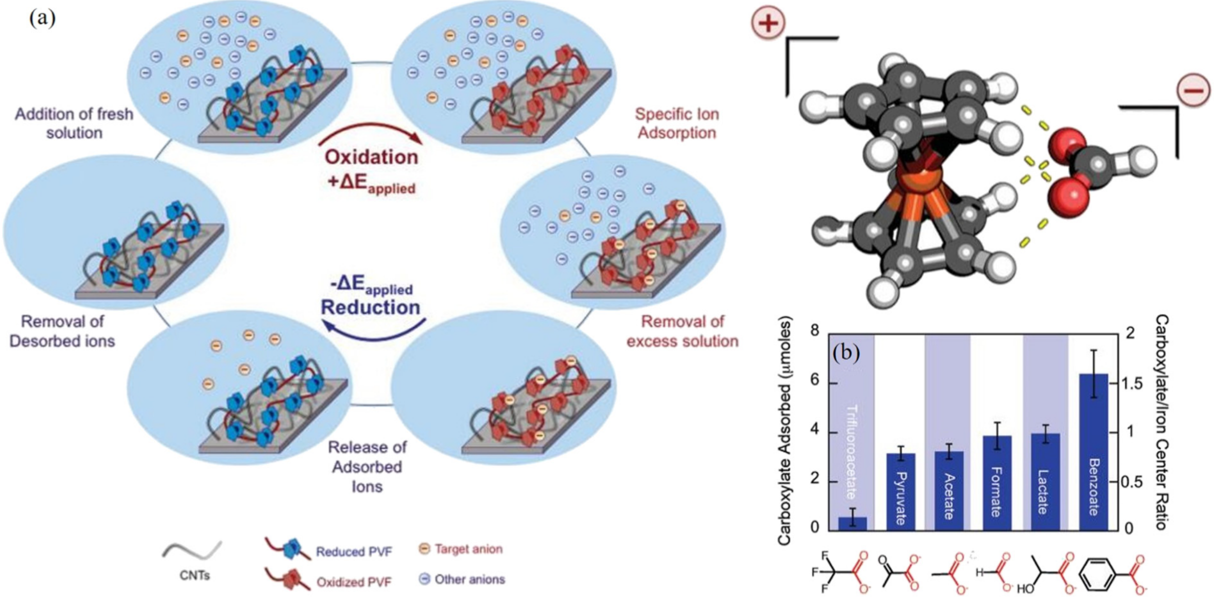

Fig. 11. (a) Specific anion interaction based on redox-species oxidation through electrochemical control for selective electrosorption. (b) An example of using specific chemical interactions to achieve selective electrosorption of carboxylate anions over size and charge, 3.4×10^{-3} M carboxylate anions with 100×10^{-3} M $LiClO_4$ as the supporting electrolyte.
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approach allowing for selective separation of dilute carboxylates (~3 mM) present in strong inorganic electrolyte solutions (100 mM of perchlorate or hexafluorophosphate) with a separation factor > 140 and subsequent reversible release of the adsorbed ions to a stripping phase solely through control of the electrochemical potential (see Fig. 11) [185]. The molecular selectivity was found to be driven by a structure-specific chemical interaction (i.e., hydrogen bonding between carboxylate anions and the cyclopentadienyl rings on the ferrocenium moiety) superimposed on the inherent electrostatic attraction. The PVF-functionalized redox electrodes were also shown to have strong specific interaction with sulfonates and phosphonates, thereby being useful for the separation of diverse classes of organic contaminants of concern [185,186]. Following this study, Su et al. demonstrated the viability and superior performance of asymmetric electrochemical cells with redox-functionalization of both cathode and anode in the enhancement of ion separation with suppressed parasitic reactions, particularly targeting organic micropollutants [187]. Su and coworkers also reported novel redox electrodes based on hemin-functionalized carbon nanotubes showing high electrocatalytic activity for nitrosamine reduction at low potentials (−0.27 V vs the standard hydrogen electrode) [188]. The underlying mechanism involved a proton-mediated conversion of the nitroso group to hydrazines and secondary amines.

4. Water disinfection

More than one billion people in the world have no or little access to safe drinking water and millions of people die every year from diseases related to water-borne pathogenic microorganisms [189]. Therefore, disinfection for drinking water has been an indispensable process to the reduction in global mortality and morbidity. Many current chemical disinfectants are effective to annihilate pathogenic microorganisms in drinking water, however, they are generally toxic themselves or may produce disinfection by-products that are harmful to human health [190]. Water disinfection based on physical methods like ultraviolet treatment suffers from high cost due to the high power consumption. Electrochemical disinfection has emerged as one of the most feasible techniques with good prospects.

During electrochemical process, generally, water disinfection can be achieved via direct and indirect pathways. For CDI devices assembled with unmodified activated carbon electrodes, the disinfection properties are mainly attributed to the electrosorption of those bacterial cells carrying negative charges induced by the presence of phosphates and lipopolysaccharides onto the positive electrodes without killing them. The high concentration of anions in the vicinity at the anode surface creates a hypertonic environment, leading to dehydration of bacterial cells and inhibiting bacterial growth [191]. The activated carbon electrodes can be surface-modified to enhance disinfection performance, say, functionalized with contact-active antibacterial materials that can efficiently kill microbes on contact by physical disruption of the anionic microbe cytoplasmic membrane. For example, activated carbon electrodes coated with cationic nanohybrids of graphene oxide-graft-quaternized chitosan [190], activated carbon electrodes coated with quaternized poly(4-vinylpyridine) [189] and activated graphene electrodes coated with quaternary ammonium cellulose. El-Deen et al. have been reported to be used for capacitive deionization disinfection [46]. These systems exhibited ultrahigh disinfection performance of > 99% killing of *Escherichia coli* (*E. coli*). Moreover, the contact-active antibacterial materials were securely attached on the carbon electrode surface thereby avoiding second contamination to the water, and these systems showed a high cyclic stability. When the applied voltage is higher than 2 V, microbes can be inactivated via indirect oxidation pathway resulting from by-products generated in situ (e.g., chlorine, hydrogen peroxide, oxidizing radicals) through the occurrence of electrochemical reactions in CDI cells, with the species and concentration of these generated oxidants depending primarily on the composition of electrolyte and electrode potential [192–194]. Most recently, Liu et al. utilized carbon fiber felt to construct two flow-through electrode systems with sequential reduction-oxidation (cathode-anode) or oxidation-reduction (anode-cathode) processes to systematically compare their disinfection performance towards *E. coli* pathogen [194].The flow-through electrode system with sequential reduction-oxidation process showed much better disinfection performance and energy efficiency than that with sequential oxidation-reduction process (see Fig. 12), and application of 3.0 V contributed to

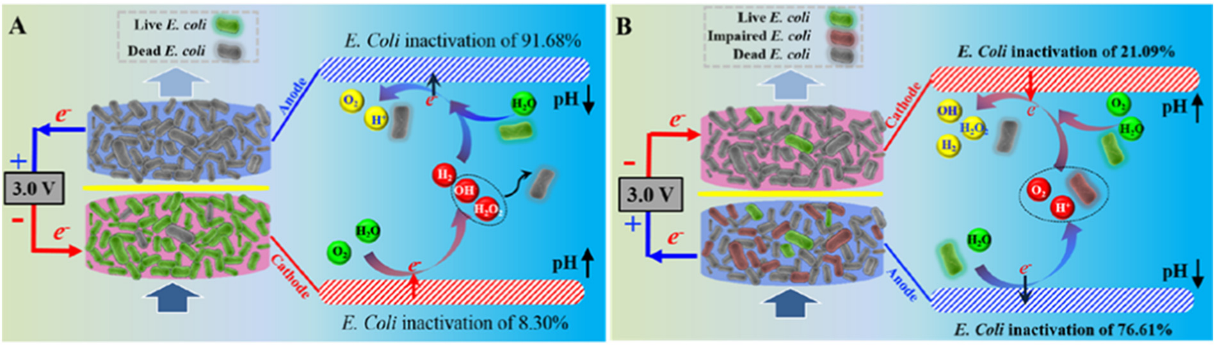

Fig. 12. Depiction of electrochemical inactivation of *E. coli* cells in reduction-oxidation (A) and oxidation-reduction (B) systems at applied voltage of 3.0 V and flow rate of 75 mL min^{-1} by OH^- and/or H_2O_2 induced indirect inactivation on cathode and direct oxidation on anode.
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~6.5 log *E. coli* inactivation at a flow rate of 50 mL min^{-1}. These encouraging results revealed the application potentials of electrochemical cells as point-of-use drinking water purification devices for pathogen inactivation, if designed and operated appropriately.

5. Resource recovery

Another potential and promising application of CDI-based technologies is to use such techniques to selectively harvest and recover valuable resources, such as lithium, vanadium, palladium, heavy metals, nitrogen, phosphorus and bromide, from wastewaters or saline water sources as an economical and environmentally friendly route for tackling environmental issues.

Lee et al. reported a modified MCDI system with a lithium manganese oxide electrode (LMO) for lithium ion capturing and a carbon electrode for anion capturing for selective lithium recovery from multication mixture solutions [157]. The adsorption capacity of lithium ions was approximately 0.35 μmol g^{-1}, 7 times higher than that from the physisorption process under the same experimental conditions. Furthermore, the LMO electrode also exhibited preferential selectivity of lithium ions over other cations. Recovery of lithium from water is of economic importance for the construction of batteries and other portable high-tech electronic devices. The separation and recovery of pentavalent vanadium (V(V)) from complex vanadium-bearing solution were also explored in a flow-by CDI unit assembled with resin-activated carbon (RAC) composite electrode [151]. Most impurity ions (Al, P and Si) are adsorbed in the EDLs of the RAC electrode while V(V) is dominantly adsorbed by the resins in the electrode. The adsorbed V(V) could be effectively separated and recovered by eluting with NaOH solution after the impurity ions were removed with diluted H_2SO_4, and then the vanadium-bearing NaOH eluent could be recycled to recover and enrich vanadium. Kim et al. verified the suitability of MCDI for palladium recovery from metal plating wastewater [150]. Palladium could be effectively enriched through multiple adsorption-desorption cycles of MCDI operation by accumulatively desorbing the ions onto the palladium concentrate. In such cases, the conventional electrowinning is feasible to serve as a post-treatment process to improve palladium recovery. Liu et al. first presented functional three-dimensional (3D) graphene-based asymmetric electrodes by grafting ethylenediamine triacetic acid (EDTA) and 3-aminopropyltriethoxysilane on the surface of 3D graphene (3DGR) respectively acting as the cathode (3DEGR) and the anode (3DNGR) for achieving separation and recovery of heavy metals (Pb^{2+}) and salt ions (Na^+) from wastewater [147]. Because of the different adsorption mechanisms (Pb^{2+} adsorbed by EDTA through chelation reaction and Na^+ adsorbed into the 3D graphene pores by electrosorption), Pb^{2+} and Na^+ can be successfully separated and recovered in the desorption process with high desorption efficiency in two steps, as shown in Fig. 13. Most recently, Liu et al. successfully utilized alternating current electrodeposition with graphene-oxide-modified carbon felt electrode to recover high-concentration heavy metals (Cu, Cd, and Pb) from industrial wastewater [195]. The graphene oxide provided high-density surface functional groups anchoring the heavy-metal ions to facilitate the nucleation. The alternating current electrodeposition could recover > 99.9% heavy-metal ions, and moreover, by tuning the alternating current frequency and voltage, the electrodeposition method was capable of further selectively recovering Cu, Cd and Pb separately.

Recovering nutrients from wastewater can alleviate challenges of both environmental systems and fertilizer demands while improving water quality. Considering the advantages and properties of CDI, studies have been conducted to explore its prospect for nutrient recovery. Wang et al. proposed a method of ion-exchange resin combined with MCDI for ammonium recovery, which achieved recovery of 65% NH_4^+ in a resin regeneration cycle after three MCDI cycles [196]. Zhang et al. developed capacitive membrane stripping for simultaneous salt removal and ammonia recovery from dilute wastewaters that involves a FCDI unit combined with a hydrophobic gas-permeable hollow fiber membrane contactor (see Fig. 14a) [197]. In the system, ammonia migrates across a CEM and selectively accumulates in the cathode chamber of a flow electrode followed by transformation to dissolved NH_3 with subsequent stripping via a membrane contactor and recovery in an acidic solution as ammonium sulfate. An ammonia removal efficiency of ~90% and a recovery efficiency of ~60% with relatively low energy consumption could be achieved. To optimize this first-generation system (e.g., simplification of the configuration, minimization of energy requirement), following this study, Zhang et al. further proposed an integrated cell with a flat-sheet hydrophobic gas-permeable membrane for ammonia recovery from both dilute and concentrated wastewaters (see Fig. 14b) [198]. The integrated cell provided a more compact and cost-effective means of ammonia recovery, exhibiting excellent ammonia removal and recovery efficiencies (up to ~90% and ~80% respectively) and comparable energy consumptions to those of conventional alternatives. Most recently, Bian et al. demonstrated the feasibility of a FCDI cell with separated anode and cathode for concurrent salinity, ammonium, nitrate and phosphates removal and recovery from water [199]. When a reverse potential was applied, > 80% of the removed nutrient ions were recovered in the concentrate during discharging operation. The connected flow operation enabled further enhanced nutrients removal by freeing up more adsorption sites via external flow anode-cathode mixing.

Also of note is that Cohen et al. introduced a novel approach to achieve the selective removal and recovery of Br^- from a mixed solution of Br^- and Cl^- by utilizing an asymmetric CDI cell to selectively enable electro-oxidation or electro-reduction of Br^- while removing Cl^- via electrostatic interactions [171]. This asymmetric CDI cell functioned via asymmetric polarization of the electrodes, whereby the applied potential was divided between the positive and negative electrodes asymmetrically. Upon electro-oxidation of Br^-, Br_2 molecules

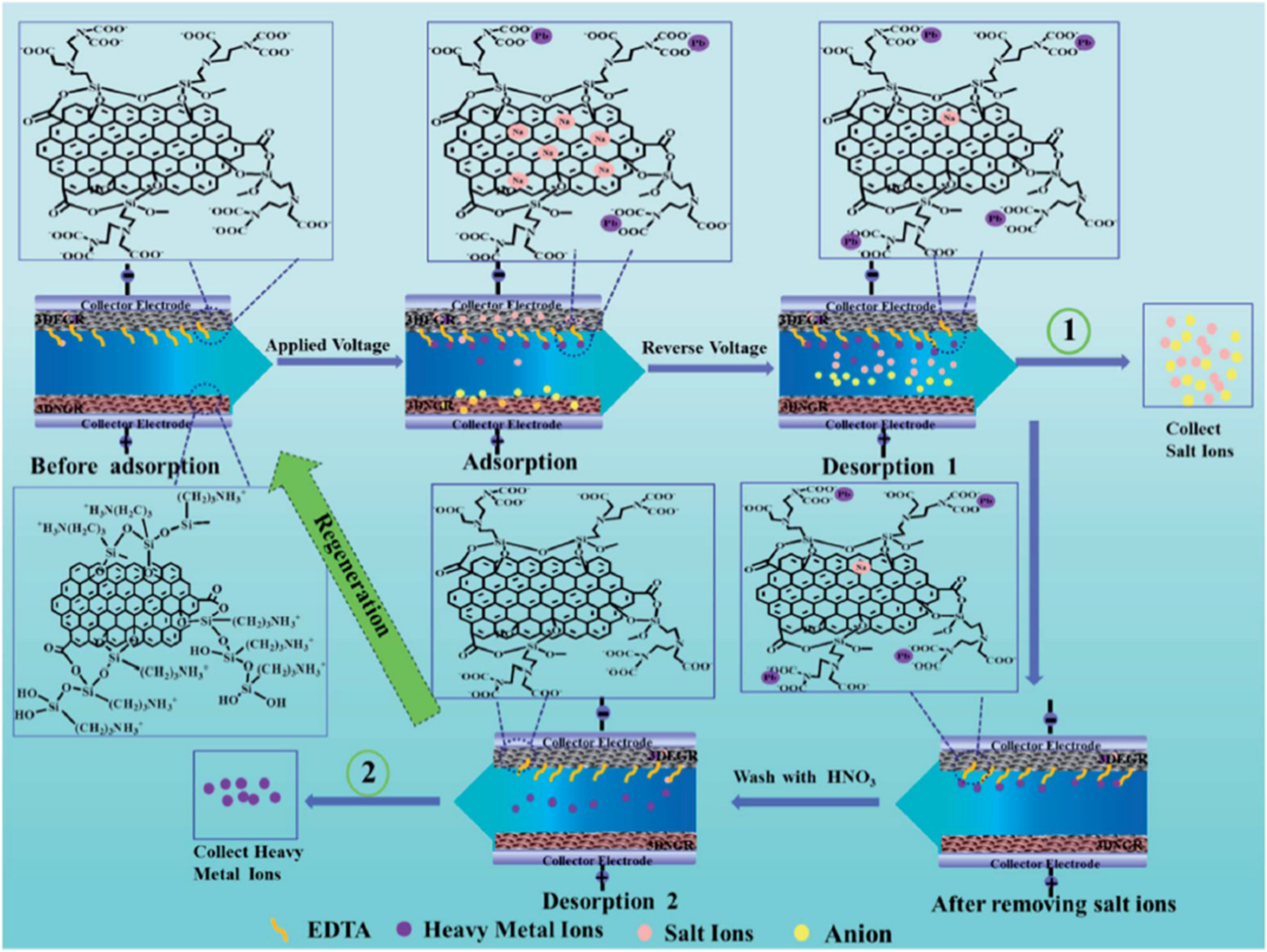

Fig. 13. Schematic illustration of the separation and recovery of heavy metal ions and salt ions from wastewaters via CDI. The insets are the 3DEGR and 3DNGR structures.
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were formed near the surface of the positive activated carbon cloth electrodes and then physically adsorbed inside the porous structure of the electrodes. When discharging the electrodes, Br_2 molecules were electrochemically reduced back to Br^- and returned to the solution. With the same initial molar concentration for Br^- and Cl^-, Br^- removal and recovery quantities were almost two orders of magnitude greater than Cl^-, consequently realizing the specific removal and recovery of Br^-.

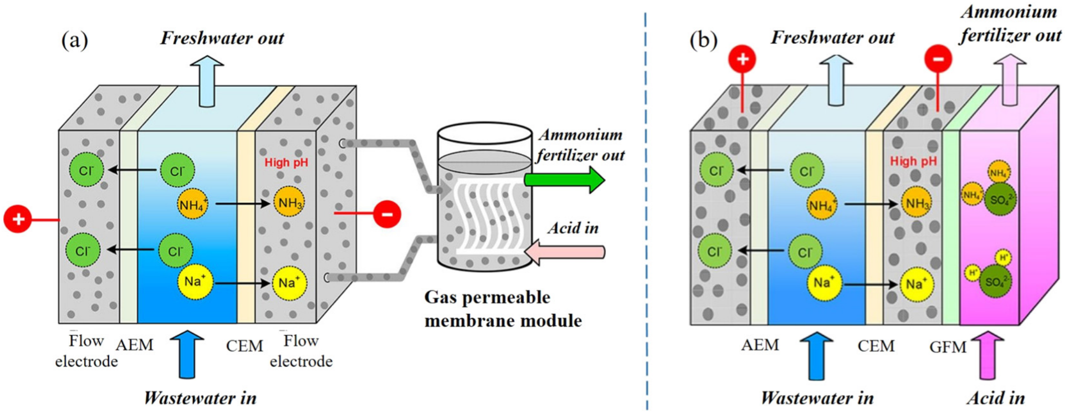

Fig. 14. (a) A FCDI unit combined with a hydrophobic gas-permeable hollow fiber membrane contactor for recovery of ammonia from wastewaters; (b) a FCDI unit integrated with a flat-sheet hydrophobic gas-permeable membrane for recovery of ammonia from wastewaters.
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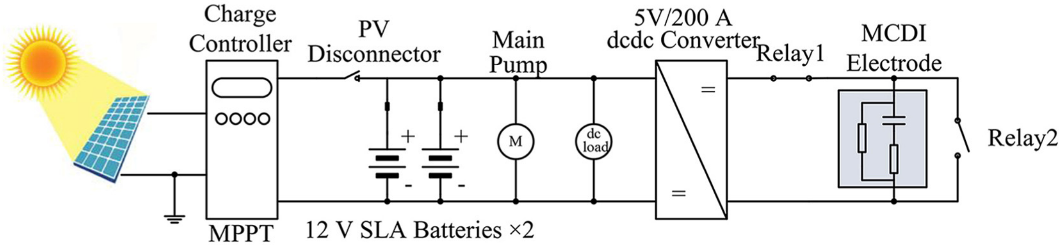

Fig. 15. Electrical diagram of a pilot-scale MCDI system powered with photovoltaics and battery storage.
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6. Combination with other technologies

Combining CDI with other technologies has been an active area of research in CDI community in recent years. CDI systems can be combined with other technologies synergistically to achieve various ultimate goals.

(1) Coupling of CDI with solar energy. Considering that CDI cells usually require a low operating voltage (about 1 V), renewable energy sources such as solar energy could be well utilized to fulfill such requirement, which makes CDI suitable for application even in areas remote from the power grid. Tan et al., for the first time, coupled with CDI technology (specifically MCDI cells) with a suitably-scaled, stand-alone, renewable power system comprising photovoltaic panels and battery storage, and designed and constructed a 1 kW pilot plant for bulk water desalination, which allowed for electrode charging currents > 100 A and product volumes on the order of 5m^3 day^{-1} (Fig. 15) [200]. Guidelines for designing and sizing such power systems were proposed including determining electrode charging current, photovoltaic panels and battery capacity. The experimental results of this prototype system were compared with those of photovoltaic powered systems incorporating other water treatment technologies such as reverse osmosis and electrodialysis, and it suggested that the use of photovoltaic-powered MCDI system, particularly when combined with energy recovery, is competitive against more mature water-treatment technologies for particular applications and at particular locations.

(2) Combination of CDI with membrane separation technologies such as reverse osmosis (RO), reverse electrodialysis (RED) nanofiltration (NF), and microfiltration (MF) for water treatment. For example, Minhas et al. integrated RO with flow-by CDI to desalinate seawater to simultaneously produce high-quality ultrapure water and freshwater at a reasonable energy consumption, which is of great interest in the locations where both fresh and ultrapure water are required [201]. In addition, simulation studies indicated that the hybrid RO-CDI desalination system could realize a higher overall water recovery and lower specific energy consumption than the existing two-stage RO system [202]. Considering that, in a typical two-pass RO system, the brackish water RO (BWRO) treatment for the purpose of meeting the produced water quality greatly increases the energy consumption of RO for seawater desalination, Choi et al. proposed a novel RO-MCDI-RED hybrid seawater desalination system in which MCDI and RED were respectively employed for substituting BWRO as the second pass desalination process and producing the energy by utilizing the brine from the first-pass RO and MCDI desalination processes (Fig. 16) [203]. Results indicated that MCDI was an excellent substitute for BWRO, and RED recovered 33% of the energy from the brine discharges. Due to the synergistic effect between MCDI and RED, the energy consumption of this RO-MCDI-RED system was reduced by 39% compared to that of a typical two-pass RO system under optimum conditions. A hybrid FCDI-NF system was reported with the potential for a high water recovery of 70% and with the potential to desalinate high-salinity brackish water (~10,000 mg L^{-1} NaCl) with lower energy consumption (~20% energy saving) compared to the best reported energy consumption of a BWRO unit [204], while the selective separation of monovalent and divalent anions (Cl^- and SO_4^{2-}) from aqueous solutions were demonstrated using the FCDI device assembled with a NF membrane and a conventional IEM with a concentration-normalized permselectivity between Cl^- and SO_4^{2-} between 1.28 and 7.03 achieved depending on the applied cell potential [205]. Most recently, integration of an FCDI cell and a MF system was proposed for brackish water desalination [206]. The integrated system enables the continuous separation of feed water into dilute and concentrated streams with extremely high water recovery (97%) at reasonable energy consumption (0.1–2.0 kWh m^{-3} depending on operational parameters).

(3) Integration of CDI with a bioelectrochemical system such as microbial fuel cell (MFC) and microbial desalination cell (MDC). MFC is an emerging technology which uses exoelectrogenic microorganisms to oxidize organic matter in the wastewater to produce electricity [207]. Taking advantage of the low voltage requirement of CDI, the electricity generated from continuous flow MFCs could be utilized to power the CDI for downstream deionization process, thereby enabling simultaneous removal of organic matter and salinity in wastewater as well as electricity production. As schematically shown in Fig. 17a. Feng et al. demonstrated that, in addition to the effective removal of carbonaceous pollutants and a variety of ionic species, the integrated MFC-CDI technology is also good at removal of ammonium nitrogen by biological assimilation and aerobic nitrification in the MFC bioreactors followed by the subsequent electrosorption of nitrate in the CDI cells [208]. Regarding the optimal arrangement of the MFC-CDI circuit, Feng et al. reported that, among the three connection types of MFCs (i.e., single-, series-, and parallel-configuration), the parallel connection of MFCs contributed to the highest voltage applied to CDI [207], while Liang et al. revealed that the performance depended on the characteristics of individual MFC and CDI reactors as well as the operating conditions [209]. As one of MFC's derivatives, MDC is a recently developed technology that can desalinate water in the middle chamber in-between the anode and cathode chambers by using the electrical potential generated by microorganisms during organic degradation to drive ion transport through a pair of CEM and AEM [210]. However, MDC suffers from pH fluctuation, and the resulting anolyte and catholyte solutions become concentrated with salt from the desalination chamber, making them inappropriate for certain reuse applications [211]. A microbial capacitive desalination cell (MCDC), which integrates CDI into the MDC design, employs porous carbon electrodes to adsorb ions thereby preventing salt transport to anode and cathode chambers, and addresses the problem of pH fluctuation by replacing the AEM with another CEM to allow free transfer of protons from the anode chamber throughout the reactor (see Fig. 17b) [212]. The MFC-CDI

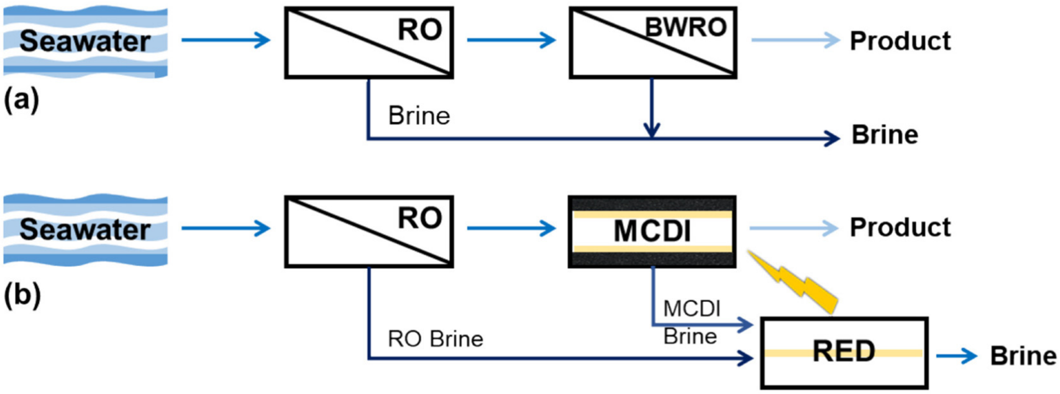

Fig. 16. Conceptual illustration of (a) typical two-pass RO and (b) novel RO-MCDI-RED hybrid systems.
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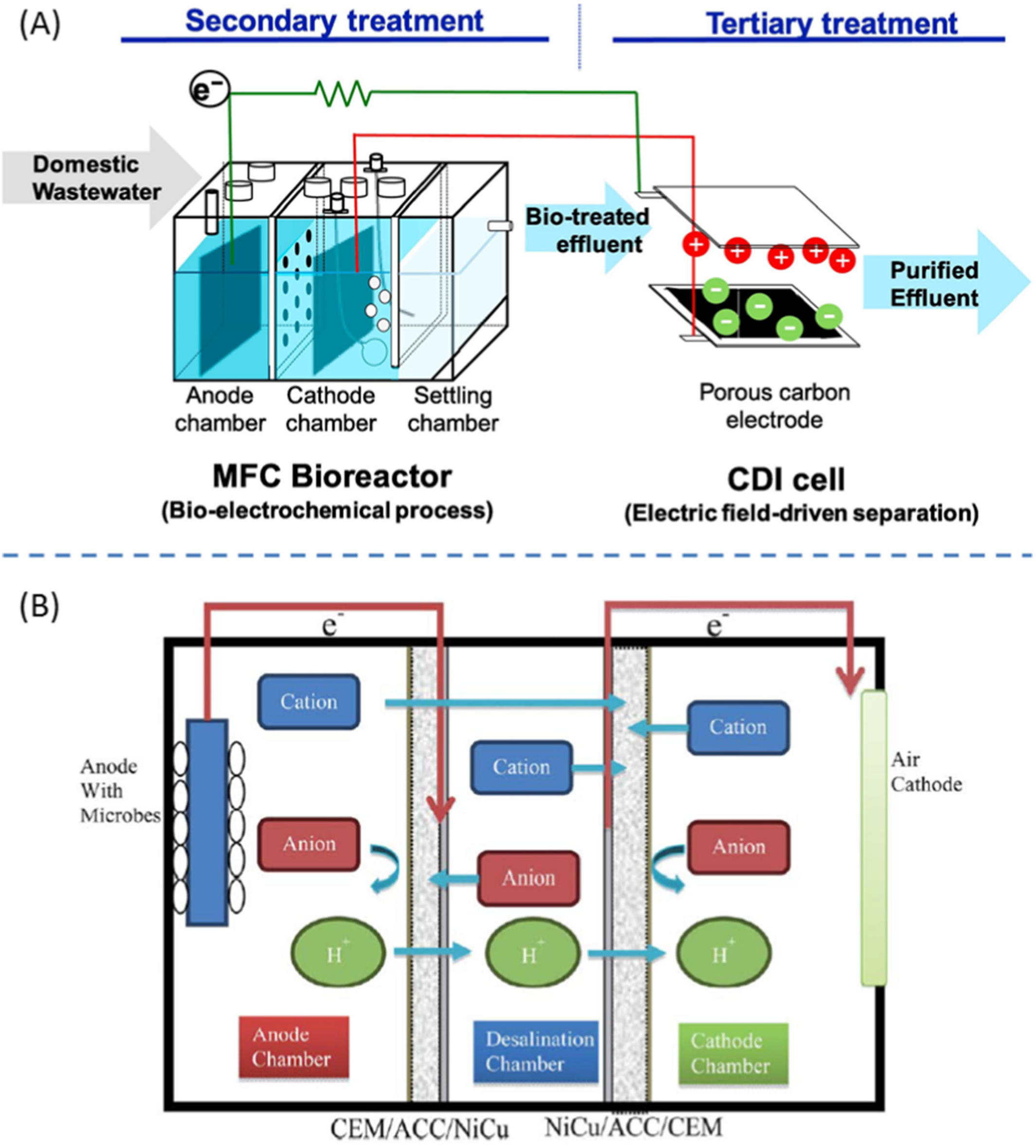

Fig. 17. (A) Schematic design of the coupled MFC-CDI system for the simultaneous removal of organic contaminants, desalination and electricity generation. Reproduced with permission from Ref. [208]. Copyright 2017 Elsevier. (B) Schematic of a microbial capacitive desalination cell (MCDC). Reproduced with permission from Ref. [212]. Copyright 2012 The Royal Society of Chemistry.

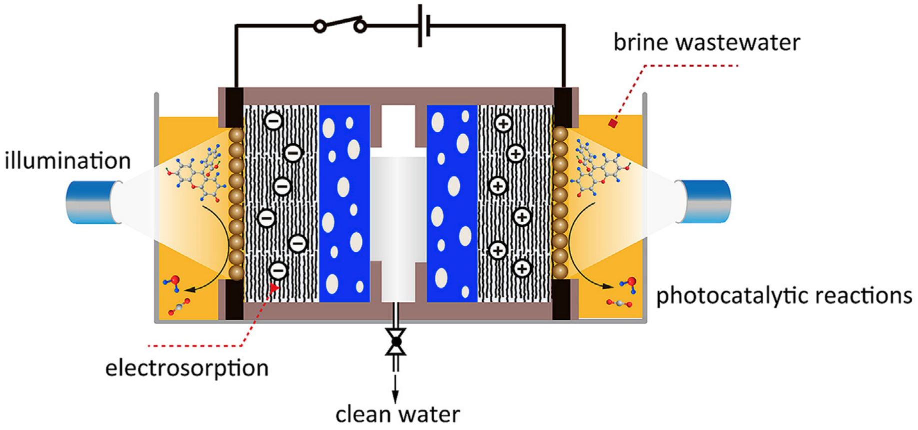

Fig. 18. Schematic demonstrating the integration of CDI with photocatalysis in the flow-through setup.
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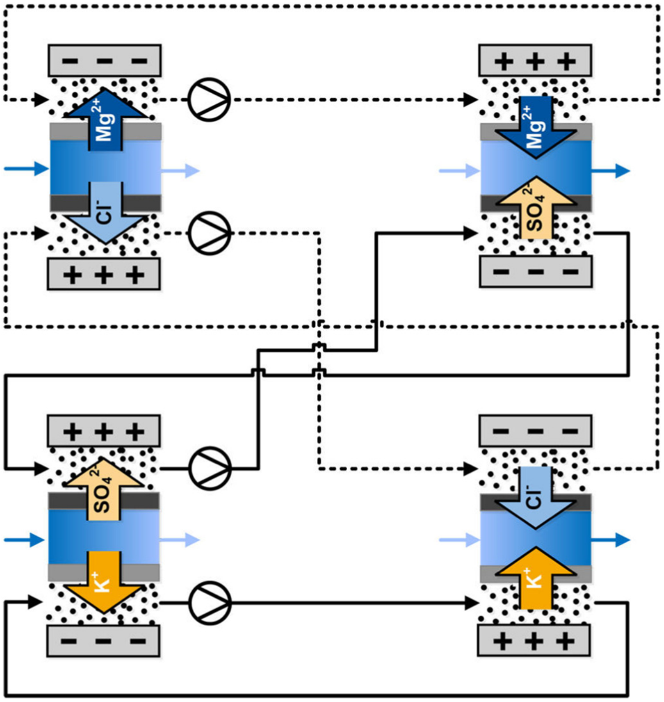

Fig. 19. Proposed process for the salt metathesis using a four-module FCDI system. $MgCl_2$ and K_2SO_4 are decomposed in the left two FCDI cells. The electrode flows are swapped into the upper and lower flow cells on the right for the recombination into $MgSO_4$ and KCl.
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and MCDC systems provide new integrated processes to complement current technologies for organic matter degradation, salt removal and energy recovery from real wastewaters.

(4) Combination of CDI with photocatalysis. The first example is a system combining CDI and photocatalysis for synergistic conversion and removal of total chromium (Cr) from aqueous solutions [152], in which the metal-organic framework MIL-53(Fe) with wide visible-light absorption was applied as positive photoelectrode material to convert Cr(VI) to Cr(III), and the activated carbon was used as negative electrode material to absorb Cr(III). Through optimization, a maximum Cr(VI) conversion ratio of 81.6% could be achieved, and the removal of the total Cr could reach 72.2%, which is difficult to be realized using current other technologies. More recently, Ye et al. designed a flow-through device integrating CDI and photocatalysis for concurrent removal of organic contaminants and ionic species by utilizing ternary membranes composed of the outmost graphitic carbon nitride (g-C_3N_4) layer as metal-free photocatalyst for degrading organic pollutants, the self-assembled carbon nanotubes membrane interlayer as electrode and active material for CDI, and the innermost poly(vinyl alcohol)-formaldehyde foam guaranteeing the mechanical stability of the membrane system, as shown in Fig. 18 [213]. Results of this study demonstrated that many types of wastewaters (e.g., solution of dyes and antibiotics) passing through the setup could rapidly turn to clean water with significantly decreased salinity. Moreover, the salt adsorption capacity of this device could be easily recovered through a simple discharge process by short-circuiting the two membrane electrodes. These encouraging results revealed the excellent practical application promise of the hybrid technology integrating CDI and photocatalysis as an efficient and environment-friendly purification method for brine wastewaters.

7. Emerging applications

CDI-based technologies are expanding into a diverse range of applications that make full use of their various configurations and unique features. Three notable aspects of the emerging application areas that hold great promise, i.e., salt metathesis, energy harvesting and CO_2 capture, are presented in this section.

As another benefit of the FCDI technique, FCDI for a salt metathesis process, also known as double displacement reactions, was demonstrated by Linnartz et al. where four identical FCDI cells were used to generate a concentrated valuable magnesium sulfate ($MgSO_4$) solution from dilute $MgCl_2$ and Na_2SO_4 (or K_2SO_4) solutions [115]. The metathesis process involves interchange of cations and anions between two salts: $K_2SO_4 + MgCl_2 \rightarrow MgSO_4 + 2KCl$, and the point of carrying out such a process is justified by the availability of the substrates and the demand for the products. The principle of FCDI for the salt metathesis is displayed in Fig. 19. In the system, feed solutions (K_2SO_4 and $MgCl_2$) were recycled through the two decomposition FCDI modules, respectively. Product solutions ($MgSO_4$ and KCl), initially made with an appropriate volume of ultrapure water, were also recycled through the two recombination FCDI modules, respectively. The pairs of flowing electrodes were recirculated through the corresponding decomposition and recombination FCDI modules and the storage vessels. Performing the metathesis process under appropriate conditions allowed the concentration of the resulting product solutions by a factor as high as 81.5 with $MgSO_4$ purity of about 80% and current efficiencies of 96%, and it is believed that $MgSO_4$ purity could be enhanced via further improvements in the ion selectivity of IEMs.

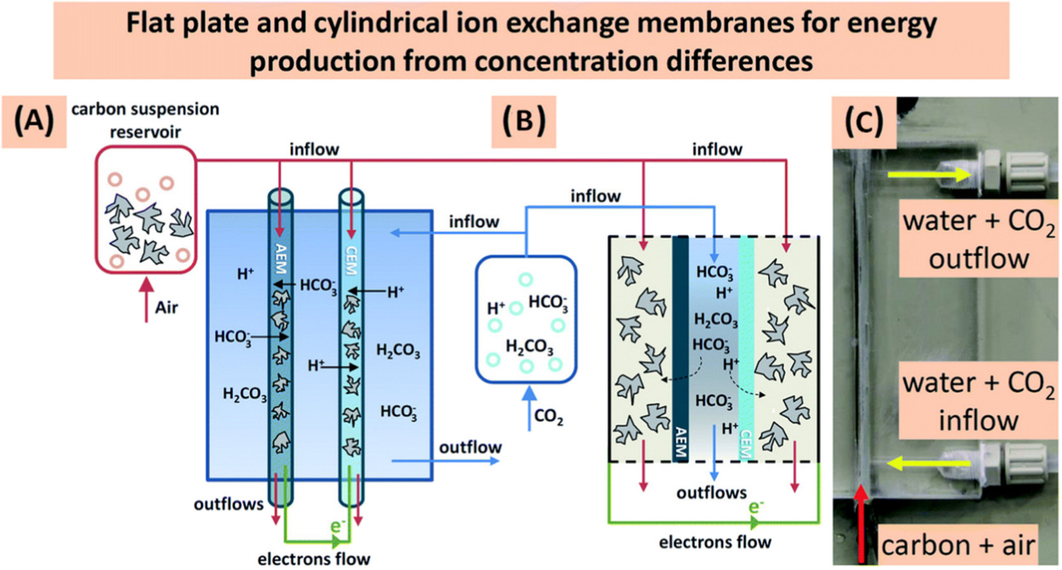

Fig. 20. Schematic view of (A) cylindrical and (B) flat plate ion-exchange membranes used for continuous energy production from capacitive mixing associated with CO_2 emissions and from differences in water salinity. (C) Picture of continuous system utilizing cylindrical ion-exchange membranes for energy production. Adapted with permission from Ref. [215]. Published by The Royal Society of Chemistry.

Energy harvesting based on conventional capacitive mixing is primarily limited by the non-continuous electricity generation resulting from intermittent charging and discharging cycles. The use of flow electrodes is beneficial to providing a means to harvest mixing energy continuously from naturally available concentration differences. Hatzell et al. demonstrated that a capacitive mixing system based on a four-reactor process with two separate flow-electrode slurries could generate constant and continuous energy from water salinity gradients (9.2 ± 0.6 mW m^{-2} for the overall system at 15% carbon mass content and 10 g L^{-1} flow-electrode electrolyte ion concentration), thereby providing a new platform for mixing energy extraction [214]. Porada et al. also demonstrated the continuous energy production via capacitive mixing using differences in water salinity, and moreover, concentration differences in bicarbonate ions (HCO_3^-) dissolved in water from the exhaust gases of a power plant with high levels of CO_2 versus the low CO_2 concentration in air, with the flow-electrode cell operated either with flat plate IEMs or with novel hollow cylindrical IEMs, as shown in Fig. 20 [215]. These results may contribute to establishing sustainable energy strategies when tapping in energy from otherwise unused sources.

The increasing rising level of atmospheric carbon dioxide (CO_2) resulting from anthropogenic emissions is one of the greatest environmental concerns of our age, and the concerns (global warming and climate change) have triggered efforts to reduce the concentration of atmospheric CO_2. Efficiently capturing CO_2 from existing emission sources is considered an important strategy for achieving this goal. Legrand et al. demonstrated that MCDI can be used to capture CO_2 in the form of bicarbonate and carbonate ions produced from the reaction of CO_2 with water at room temperature and atmospheric pressure without any addition of chemicals or heat (see Fig. 21) [216]. Under similar conditions, this system exhibited energy consumption comparable to or lower than that of similar technologies such as supercapacitive swing adsorption. In practical applications, $CO_2(g)$ should be absorbed from a diluted gas mixture when MCDI is charged and desorbed into a concentrated stream when MCDI is discharged. A membrane contactor could be used to separate the gas from the liquid phase after the charging and discharging steps.

8. Summary and outlook

In summary, a state-of-the-art and comprehensive review about the versatile applications of CDI has been presented here. The past two decades has seen tremendous advances for CDI, and CDI-based technologies have grown from the application of brackish water desalination to many new application areas such as seawater desalination, water purification, resource recovery, water disinfection, synergistic combination with other technologies, energy harvesting and CO_2 capture. Despite the remarkable progress achieved, the research on the applications of CDI necessitates ongoing efforts and there are several important issues requiring focus in the near future.

(1) A great many works in the literature observed the performance of CDI based on the first or only a few initial cycles. It is of importance to investigate the long-term performance and report the cyclic stability. In terms of electrode materials, although numerous capacitive, battery and hybrid materials have been synthesized to substantially improve desalination capacity [17,217], few studies were conducted to develop electrodes with improved longevity. Evidence and explanations for the decreasing electrode stability have been provided in many articles, but practical solutions to enhancing their long-term stability are still required. The research group of Pan et al. recently made contributions by providing several improvement strategies for the HCDI's cycling performance. One route is introducing redox-active polyimide as the cathode of HCDI instead of conventional inorganic intercalation compounds suffering from drawbacks such as decomposition and dissolution [218]. Another approach is using nickel hexacyanoferrate/reduced graphene oxide cathode with high theoretic capacity and low Na+ intercalation/extraction potential that could be operated a low voltage, which thereby effectively suppresses the oxidation reactions on the activated carbon anode [219].

(2) The cost of the commonly utilized commercial IEMs remains prohibitively high for large-scale applications. Moreover, the IEMs are usually thick and have high electrical resistance. The weak contact adhesion between the electrodes and the IEMs might also produce a noticeable contact resistance [17,220]. Research efforts are required to develop alternatives to the IEMs with lower cost and/or lower resistance. In addition, considering that IEMs could play an

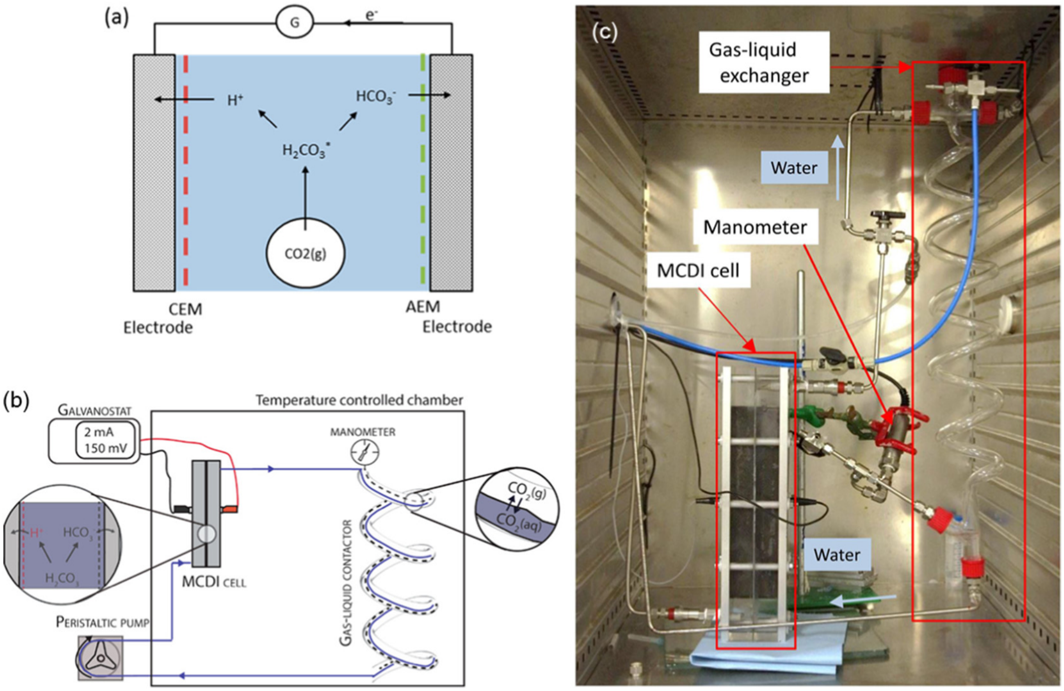

Fig. 21. (a) Schematic of CO_2 capture in the form of bicarbonate and carbonate ions produced from the reaction of CO_2 with water using a MCDI cell during charging. (b) Research setup scheme. The blue line represents the CO_2-flushed deionized water while the black dashed line represents the CO_2(g) volume. (c) Photo of the research setup during operation. The peristaltic pump is located outside the temperature controlled chamber. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)
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important role in the selective separation for CDI cells assembled with membranes, development of novel membranes with high selectivity/affinity towards specific ions allows for new application possibilities in the selective ion removal from multi-ionic solutions.

(3) As one great advantage of CDI-based technologies, in parallel with ion desorption, the energy consumed during the charging process can be partially recovered. Full recovery of the energy released during the discharging process to lower the associated energy consumption is the key to the large-scale commercialization of CDI-based technologies. Although many researchers have calculated and reported the energy consumption of CDI-based technologies for particular applications, the minimum net energy input in the case of energy recovery remains largely unknown. Consequently, the energy consumption with energy recovery is most likely to be a more appropriate performance metric than the energy consumption without energy recovery in future research.

(4) More future research for applications of CDI-based technologies should shift to real water conditions such as surface waters, underground waters and wastewaters. An overwhelming majority of CDI research explored the applications using feed waters with simple components that are synthesized in a laboratory using deionized water. Organic substances (e.g., natural organic matter) and certain ions (e.g., harness ions, bicarbonate ions) which are abundant in real waters may have negative influence on the operation performance due to the occurrence of scaling/fouling on the CDI electrode surface and the competition effects. Synergistic combination of CDI with additional pre-processing to minimize such effects needs to be explored.

(5) Applications of CDI-based technologies may be expanded to a diverse range of non-aqueous media. While CDI has been proven to be a versatile tool for separation processes in aqueous solutions, little attention has been paid to separations achievable by CDI in electrolyte systems using organic solvents. Liu et al. investigated the electrosorption of LiCl from different solvents (e.g., water, methanol, ethanol and *N*, *N*-dimethylformamide) by carbon nanotube film electrodes with results showing that the solvent plays an important role in the removal process owing to the characteristics of the solvent [221]. One study conducted by Porada et al. demonstrated the potential of CDI-based technologies for the application of electrosorption of organic electrolyte (tetraethylammonium tetrafluoroborate salt) dissolved in organic solvents (propylene carbonate) [222]. The operating voltage window of CDI was extended to 2.5 V and the system showed higher salt removal capacity and charge efficiency compared to CDI applied for treatment of aqueous electrolytes (NaCl). It is believed that the same concepts can be widened to other organic solvents, charged molecules, and applications such as extracting valuable resources or removing impurities from organic solvents in the chemical and pharmaceutical industries.

(6) Despite some industrial efforts to commercialize CDI-based technologies by companies such as Voltea B.V. (The Netherlands), EST Water & Technologies (China) and Siontech (South Korea) [37], there is a long way to go before CDI ultimately becomes a competitive means for water treatment or other purposes. Studies about the stability of large-scale CDI applications are rarely reported and the difference of stability between lab-scale and large-scale CDI applications is largely unknown. To further promote their commercial applications, tests based on pilot-scale or even large-scale CDI facilities manufactured using the innovative ideas from lab-scale studies are desirable. Such studies will provide valuable information about scalability, system cost, concentration limits, electrode/membrane's behavior after long-term operation and so on, thereby paving the way for a new generation of CDI systems.

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.

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