

Commercialization Efforts of Capacitive Deionization Technology in Water Treatment Processes

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Although capacitive deionization (CDI) technology has been studied intensively for more than 20 years, its commercialization remains in the initial stage, which is partly caused by the insufficient knowledge exchange between academia and industry. This concept reviews multiple scaling-up efforts in the CDI technology for treating real water streams, following a case-by-case fashion. While the cell architecture in pilot scales is limited to the membrane CDI, highlighting the necessary role of ion-exchange components during scaling-ups, different ways of

electrode stacking, i.e., monopolar and bipolar, are available. The performance indicators of these CDI systems when treating real water streams are summarized to gain insights into industrial practices. Importantly, key discrepancies in cell components and performances between pilot-scale and lab-scale studies are emphasized. The main challenges in large-scale CDI systems for industrial purposes are discussed, providing hints for a better integration of research and applications.

1. Introduction

Successful utilization of saltwater reservoirs could largely resolve freshwater scarcity issues, but this requires reliable and economical desalination technologies. The global freshwater produced by desalinating saltwater in 2020 reached ca. 95 million tons per day.^[1] At the moment, membrane-based reverse osmosis (RO) seems the most promising technique in treating seawater or wastewater with high salinity, occupying ca. 70% of the total desalinated water production.^[2] In addition to membrane processes, an electro-sorption technology, capacitive deionization (CDI) was realized in the late 1990s, offering excellent prospects for treating brackish water streams.

CDI follows the working principle of an electrical double-layer capacitor (EDLC).^[3] Through polarizing the two porous electrodes, e.g., activated carbon (AC), carbon aerogel, etc., the charged species in the electrolyte (i.e., salt ions) can be adsorbed onto the electrodes, thereby desalinating feedwater and storing energy simultaneously during charging, and vice

versa for the discharging. Importantly, the processes are repeatable, enabling the periodic water treatment and energy harvesting/reuse, making CDI a distinct technology. The energy consumption of CDI is proportional to the salt concentration of feedwater, making the technology more efficient in handling brackish water ($\leq 3,000$ mg/L total dissolved solids, TDS).^[4] Compared with RO, CDI exhibits the advantages of higher water recoveries, less membrane waste, lower influent standards, and adjustable effluent salinities.

Despite various electrode materials and cell architectures being developed by researchers, commercially available CDI systems mostly rely on AC electrodes due to their mass-produced nature and low cost. However, the performance of AC needs further improvements, necessitating the ion exchange membranes (IEMs) or ion-exchange coatings, i.e., membrane CDI (MCDI). The two ways of electric connections distinguish the MCDI into monopolar and bipolar. The former (Figure 1a) was successfully commercialized by the Dutch company Voltea B.V., fulfilling the desalination requirements in fields like small-scale agricultural irrigation and the reuse of industrial cooling water.^[5] The Chinese company EST Water & Technologies, differently, developed the bipolar MCDI (Figure 1b), which seems working quite well as evidenced by the fact that the company has achieved its 8th product iteration recently.^[6] In short, the monopolar MCDI has evenly distributed electric potential but requires numerous welding points, whereas the bipolar MCDI system displays a faster desalination speed due to an intrinsic lower time constant with a simpler circuit connection.^[7]

While most research papers report some of the best performance indicators of CDI systems, the conclusions drawn from the laboratories cannot fully bridge the gap between academia and industry. Given the situation, this paper primarily focuses on the scaling-up efforts by reviewing the technical details and the performance indicators of pilot-scale cell designs when treating real water streams. Furthermore, case studies of these pilot CDI systems are evaluated to highlight the key

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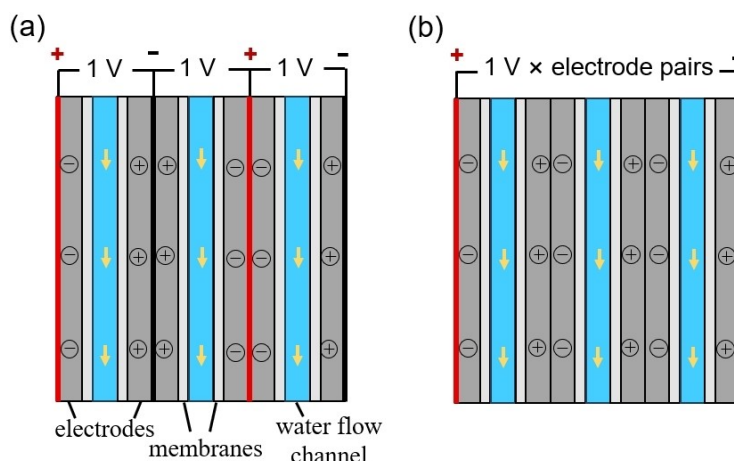


Figure 1. Schematic illustrations of the CDI stack: (a) monopolar MCDI and (b) bipolar MCDI.

obstacles to commercialization. Lastly, the economic and stability issues are discussed, providing guidelines for future industrialization and deployment.

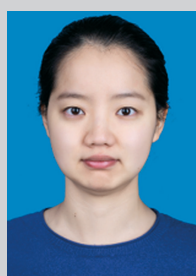
2. Scaling-Up Efforts of CDI Technology

2.1. Industrial Water Reuse

CDI technology can be applied for desalinating industrial water streams from cooling towers, desulfurization, steel manufacturing, and mining.^[5–6,8] There are three water streams in a cooling tower, namely feedwater, blowdown water, and recirculation water, of which CDI technology could be an ideal option for treating the first two.

2.1.1. Monopolar Pilot MCDI modules – Voltea B.V. and Wageningen University, the Netherlands – Cooling Tower Feedwater, 2014^[9]

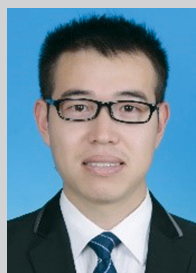
One of the demonstrations was in a Unilever site at Pratau, Germany, which lasted for one year. As demonstrated in Figure 2, the system consists of a prefiltration train and a butter tank, in addition to the two monopolar MCDI modules that were employed under constant current mode. Each CDI module contains 750 g AC electrodes, yielding a water productivity of 7 L/min (0.42 t/h). By treating the feedwater with a conductivity of 0.65 mS/cm, the conductivity removal of ~70% and water recovery of ~84% were achieved with an energy consumption of ~0.234 kWh/m³. With the energy consumption being advantageous to that of RO, water savings, wastewater savings, and chemical savings, over the entire evaluation also achieved competitive levels up to 28%, 48%, and 85%, respectively. Furthermore, MCDI is less susceptible to silica scaling compared



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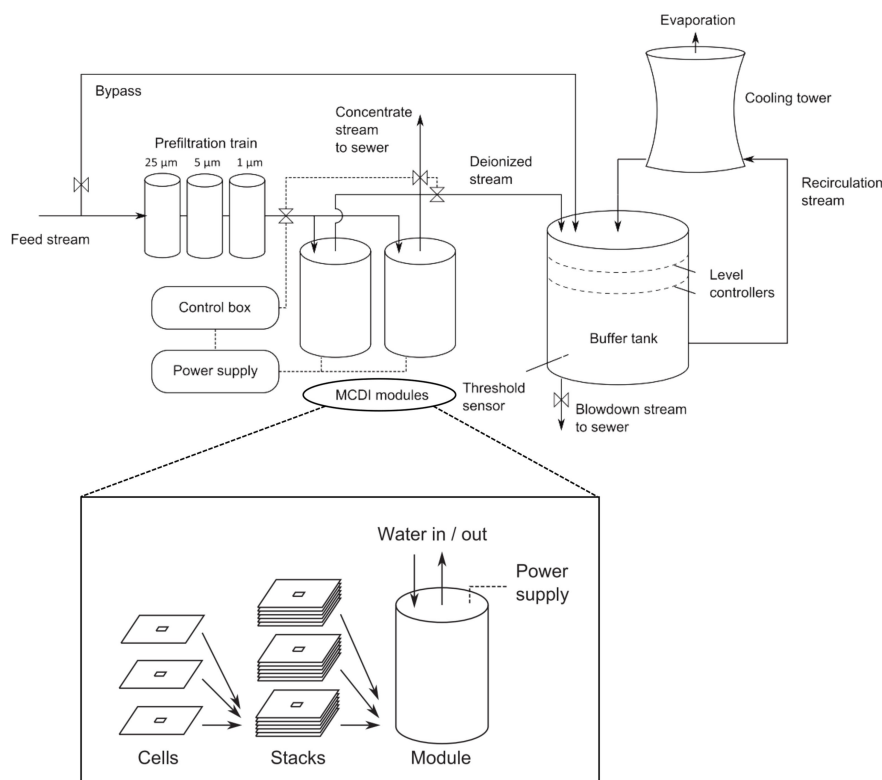


Figure 2. Schematic of the cooling tower configuration equipped with MCDI. Solid lines (—) indicate water flows, dotted lines (...) indicate control lines, and double triangles indicate valves. Adapted with permission from Ref.^[9]. Copyright (2019) Elsevier.

to membrane separations, as silica is not charged under normal operation conditions.

2.1.2. Bipolar Pilot MCDI Modules – EST Water & Technologies, China – Cooling Tower Blowdown Water, 2015^[10]

The pilot was deployed at a power plant in Inner Mongolia, China, with a water productivity of ca. 6.7 L/min. Pretreatment was first used to decrease the alkalinity (~38.5% removal rate) and the hardness (~75.5% removal rate) of the feedwater. By passing through the MCDI modules, the average conductivity of feedwater decreased from 3,277 to 882 $\mu\text{S}/\text{cm}$ (~74.9%). The scaling/fouling issue of the module was prevented from both the pretreatment and the post-cleaning. The total cost was estimated to be 1.5 RMB (0.21 USD) for treating one ton of the water stream, among which 0.83 RMB (0.12 USD) was covering the energy consumption (1.78 kWh/m³). Treating cooling tower wastewater is the primary application for CDI technology in China. Financially, the reuse of blowdown wastewater is favored due to the high sewage costs in the region.

2.2. Drinking Water Supplement

CDI also shows promise in producing drinking water. Thanks to the low energy consumption, it can be integrated with photovoltaic panels, providing fresh water for remote locations where

grid power is hardly accessible.^[11] The commercialization product (Voltea B.V.) can already be seen in a 1.5-year field trial in central Australia, producing drinking water with competitive energy requirements.^[12] While RO removes all minerals after the desalination process, essential ions may remain after CDI treatment. Generally, water with a TDS level lower than 500 mg/L is suggested to be acceptable for the human consumption.^[13]

2.2.1. Portable Monopolar MCDI Prototype – LT Green Energy, Australia – Inland Brackish Groundwater, 2013^[14]

The prototype was demonstrated in Wilora, Northern Territory, Australia. As shown in Figure 3, the system consists of a prefiltration unit and a CDI cell matrix. Each CDI unit consists of 100 pairs of coconut-derived AC electrodes (a specific area of 800 m²/g and a total weight of 1,354 g). An incoming water stream of 7 L/min was suggested to be the optimum, giving an energy consumption of 1.89 kWh/m³ (1.5 V). The TDS level of ca. 1,500 mg/L was reduced to ca. 500 mg/L after one working cycle that lasted for 120 seconds, with a water recovery rate of between 75% and 80%. As for the long-term operation, the removal efficiency of the CDI unit decreased by 7% after 15 days, which was then recovered to its original state after cleaning with 20 L of 0.01 M citric acid (for metal scaling) and 0.01 M sodium hydroxide (for organic fouling).

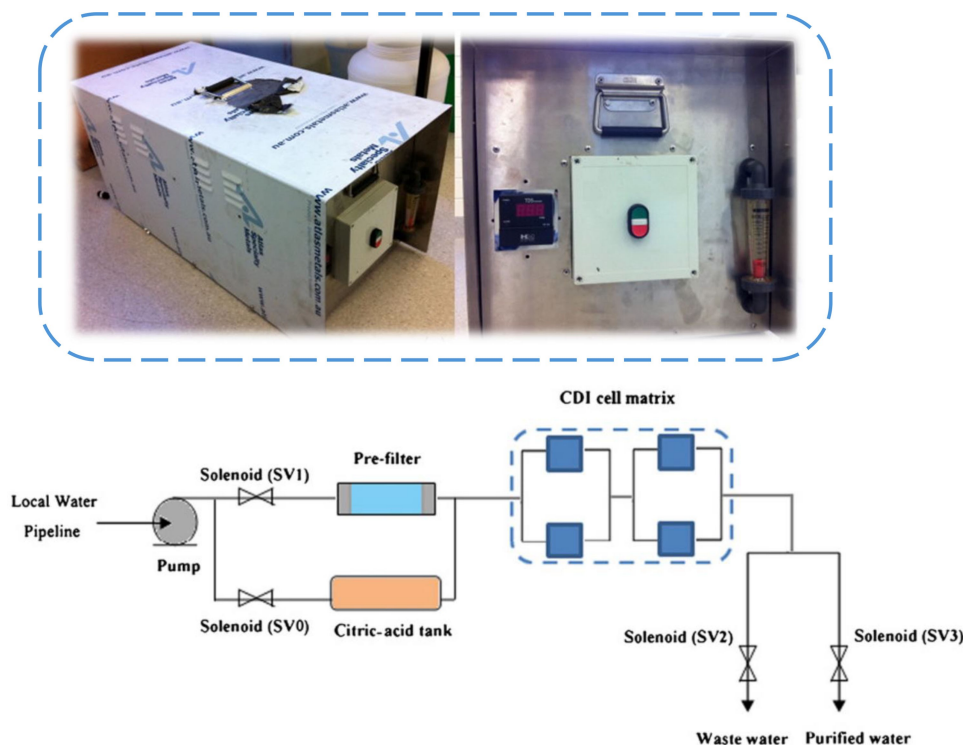


Figure 3. Photographs and schematic diagrams of the CDI unit. Adapted with permission from Ref.^[14a]. Copyright (2013) Elsevier.

2.2.2. Charge-Barrier MCDI Pilot Unit – ENPAR Tech. Inc., Canada – Synthesized Water Samples Containing NaCl or Nitrates/Ammonium, 2009^[15]

The model DesEL 9 K pilot unit shown in Figure 4 followed an MCDI design with charge barriers placed on both electrodes of the cell. Each unit had a cylindrical geometry ($\varnothing = 298$ mm, $h = 366$ mm) contributed by an AC electrode (0.25 mm thick) stack, leading to the overall size of the pilot system that weighs ca. 10 kg and occupies ca. 1.5 L in volume, i.e., 1.2 (l) \times 1.0 (w) \times 1.8 (h) m. The adsorption (deionization) stage continued until the conductivity was above a certain threshold and moved to the desorption (regeneration) stage. A purge process

was involved between each adsorption-desorption cycle to remove the concentrate. The TDS of raw water ranging from 150 to 3,000 mg/L was removed with high efficiencies, i.e., 80.9%–94.3% upon energy consumptions of 0.45 to 5.35 kWh/m³. In addition, the removal rates for nitrates and ammonium ions achieved 88%–98% and 71.9%–88.1%, respectively.

2.3. Domestic Wastewater Treatment

Municipal wastewater treatment usually consists of several major processes: 1) Physical separation – removal of solids; 2) sludge activation – degradation of organics; 3) adsorption/

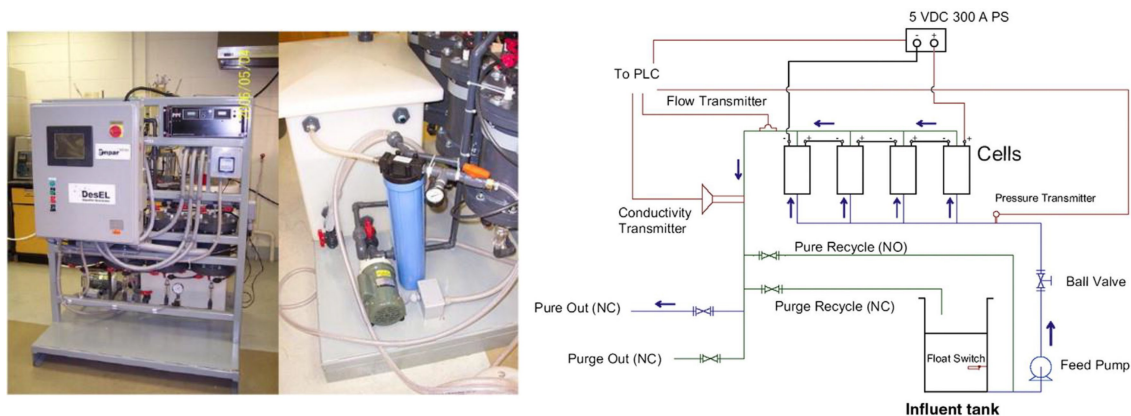


Figure 4. Photographs and instrumentation schematic of the DesEL 9 K pilot plant (ENPAR Tech. Inc.). Adapted with permission from Ref.^[15]. Copyright (2009) Elsevier.

filtration – removal of phosphates and nitrates; 4) chlorine/ozone/UV – disinfection.^[16] CDI is anticipated to be performed in the third process since both phosphates and nitrates may be presented as polarized/polarizable ions in domestic wastewater.

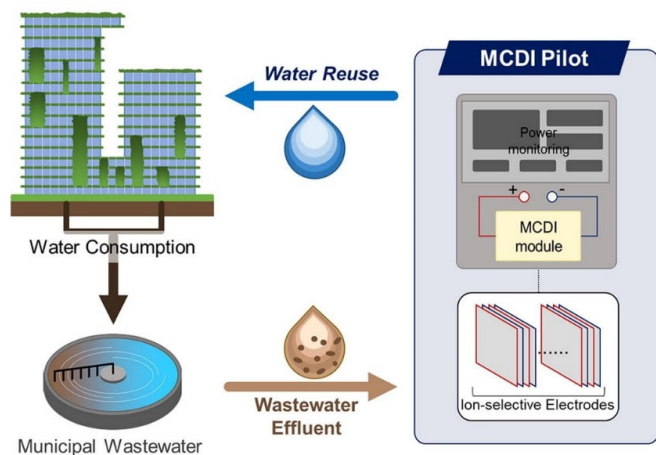


Figure 5. CDI concept for reuse of municipal wastewater: Schematic of an MCDI module designed for treating secondary wastewater effluent. Adapted with permission from Ref.^[17]. Copyright (2019) Elsevier.

2.3.1. Monopolar Pilot MCDI Systems – University of Technology Sydney and Siontech. Co., Republic of Korea – Effluent From a Decentralized Wastewater Treatment Plant, 2019^[17]

In 2019, Kim et al. attempted to reuse municipal wastewater by installing a pilot-scale MCDI system with 50 pairs of AC electrodes (P-60, Kuraray, Japan) at a decentralized wastewater treatment plant located in Sydney, Australia (Figure 5). The electrodes were coated with either cation- or anion-exchange polymers, referring to the ion-selective electrodes with low membrane/contact resistance. By treating a real wastewater stream of $800\ \mu\text{S}/\text{cm}$, the MCDI module exhibited a total salt removal rate of ca. 51% with a NO_3^- removal efficiency of 75.7%. The ion selectivity followed the sequence of $\text{Cl}^- > \text{NO}_3^- > \text{SO}_4^{2-}$, which was suggested to originate from the anion perm-selectivity of the anion exchange membrane. Moreover, the pilot-scale MCDI unit operated for 15 days and did not show evidence of performance degradation, even though the organic concentration of the wastewater was non-negligible (i.e., $12.4\ \text{mg}/\text{L}$ dissolved organic carbon).

2.3.2. Monopolar Pilot MCDI System – National Taiwan University – Secondary Effluent From a Domestic Wastewater Plant, 2021^[18]

As demonstrated in Figure 6, the system was installed in a domestic wastewater plant in Taoyuan, Taiwan, consisting of a membrane bioreactor (MBR) unit, a pretreatment unit, an MCDI

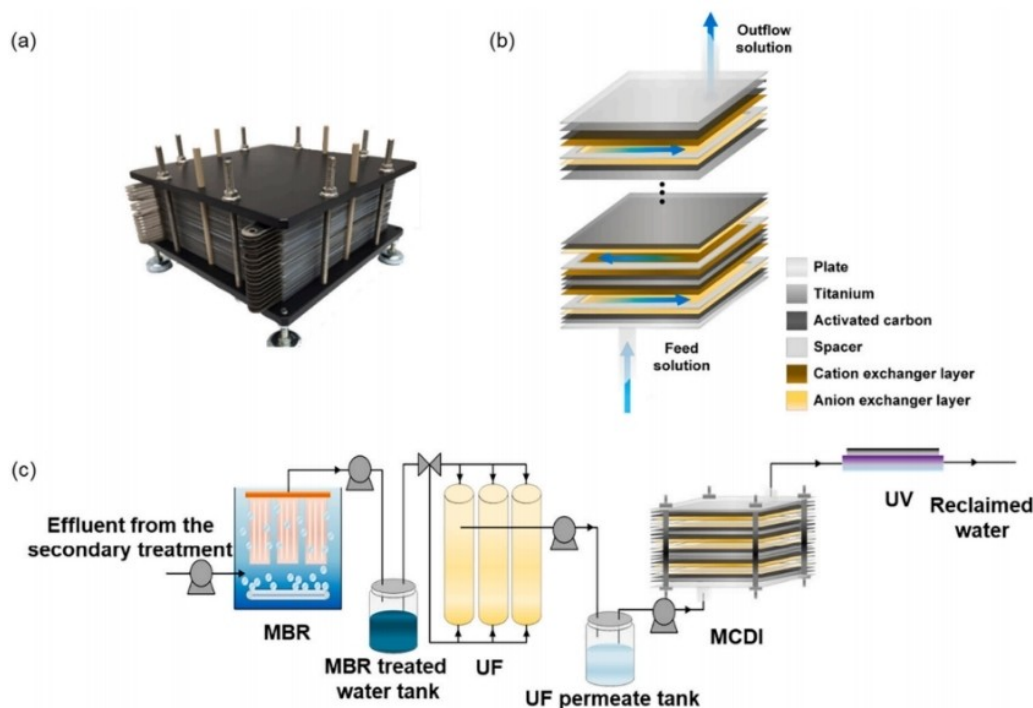


Figure 6. Schematic diagram of the MCDI stack: (a) macroscopic view, (b) cell configuration, and (c) the MCDI-based system installed after an MBR for wastewater reclamation. Reprinted with permission from Ref.^[18b]. Copyright (2013) Elsevier.

stack, and a disinfection unit, in which an MCDI stack contains 40 pairs of $20 \times 20 \text{ cm}^2$ commercial AC electrodes obtained from China Steel Chemical Corporation (ACS20). A charge efficiency of $\sim 95\%$ and an energy consumption of $\sim 0.172 \text{ kWh/m}^3$ were achieved following a single-pass mode and a potentiostatic (1.2 V) charge-discharge cycling. The conductivity of the secondary effluent from the plant was decreased from 550 to $103 \mu\text{S/cm}$, which met the Taiwanese standard for industrial reuse ($250 \mu\text{S/cm}$). The pilot scale MCDI system exhibited promising ion removal capabilities, giving 94.4% , 93.7% , and 83.9% for Ca^{2+} , Mg^{2+} , and NO_3^- , respectively. During the long-term operation for >5 months, the water quality of effluent remained stable, while the deionization capacity and charge efficiency decayed due to scaling and biofouling.

2.4. Agricultural Irrigation

Thanks to the ion selectivity of CDI, an excess amount of Na^+ can be removed while retaining a certain number of mineral ions (e.g. Ca^{2+} , Mg^{2+} , K^+ , SO_4^{2-}) to achieve a lower sodium adsorption ratio (SAR) of fed streams for better plant growth. Commercial CDI products are already seen in irrigation practices of greenhouse agriculture.^[5,19]

2.4.1. Lab-Scale CDI With Modified AC – Technion – Israel Institute of Technology – Synthesized Water With Multi-Ions, 2021^[20]

In 2021, Guyes et al. proved the feasibility of CDI in irrigation on a lab scale, of which the operational concept is illustrated in Figure 7. By using sulfonated cathodes, their CDI cell achieved a $\text{Na}^+/\text{Ca}^{2+}$ separation factor of up to 1.6, simultaneously reducing ionic conductivity from 1.75 to 0.69 mS/cm and SAR from 19.8 to $13.3 \text{ mM}^{1/2}$. Furthermore, the salt adsorption

capacity (SAC) and the monovalent selectivity barely degraded after 1000 charge-discharge cycles while maintaining a low energy consumption of ca. 0.38 kWh/m^3 .

2.5. Lithium Extraction

CDI technology can also be extended to resource recovery, e.g., selectively extracting Li from brine water sources. Either using Li-selective membranes or Li-selective electrodes in CDI would achieve Li recovery from aqueous streams^[21]. Ion selectivity is enabled by mono-valent IEMs (e.g., CIMS, ASTOM, Japan) for the former,^[22] whereas the latter exhibits the ion selectivity by manipulating the interlayer spacing of the battery electrodes.^[23]

2.5.1. a. Lab-Scale Hybrid CDI – Anhui University, China – Natural Water From a Salt Lake.^[24]

Although large-scale demonstrations seem limited, Zhou et al. developed a carbon-coated lithium vanadium phosphate electrode that was highly selective in lithium extraction. Their hybrid CDI setup was tested with the water sample collected from a natural salt lake in Tibet, exhibiting very promising Li selectivity coefficients of 187.50 ($\text{Li}^+/\text{Mg}^{2+}$), 136.11 (Li^+/K^+), 116.8 (Li^+/Na^+), and 4.92 ($\text{Li}^+/\text{Ca}^{2+}$).

3. Application Concerns and Perspectives

Generally, pilot-scale CDI practices remain limited, although the technology has been developed for decades and is mostly viewed as a promising one. After going through these CDI cases, crucial factors are summarized in Table 1 for easy comparisons. Some discrepancies between the pilot-scale and the lab-scale system should be highlighted:

- 1) To cut down the capital cost by minimizing the weight percentage of current collectors and structural components, thick electrodes ($>200 \mu\text{m}$) with a high mass loading of $0.01\text{--}0.03 \text{ g/cm}^2$ are always favorable in pilot-scale CDI systems. However, the increase in electrode thickness/mass decreases the charge efficiency and the SAC in mg/g-electrode . In contrast, the electrode thickness/mass in experimental studies is usually significantly smaller ($<100 \mu\text{m}$, $<0.01 \text{ g/cm}^2$).^[24–25] The situation perfectly explains why the promising numbers achieved in laboratories are not seen in large-scale systems. In this regard, ideal CDI electrode materials for commercial use should balance the conductivity, water permeability, and robustness, rather than solely maximizing SAC.
- 2) The advantages of CDI, including low-cost, membrane-less, and pollution-free, seem no longer convincing in pilot scales. IEMs remain the crucial components in almost every state-of-the-art CDI system that is commercially available. Certainly, the performance indicators of CDI can be effectively improved by the presence of IEMs, which however, contribute to around half of the capital cost of the

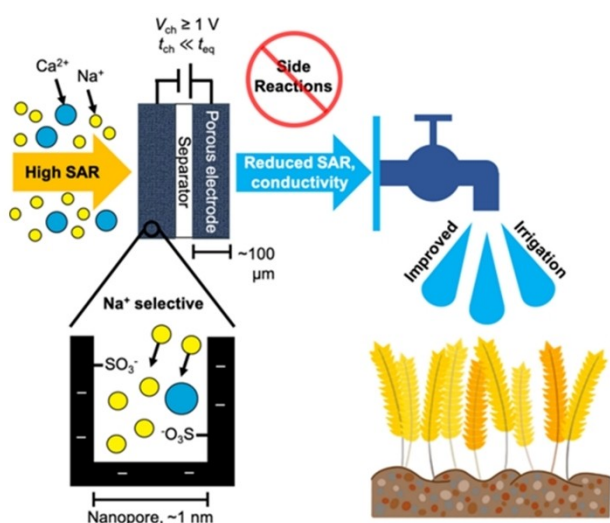


Figure 7. CDI concept for agricultural irrigation water treatment: Schematic of a CDI cell fed with water containing excessive Na^+ which must be treated for direct use in irrigation. Reprinted from Ref.^[20] CC BY.

Table 1. Parameters and performance indicators of various CDI prototypes.

year	cell architecture	electrode materials	weight/pairs	mass (g/cm ²)	productivity (L/min)	operation mode	SAC (mg/g)	feedwater	salt removal rate	energy consumption (kWh/m ³)	water recovery	duration	module source
1 2014	monopolar MCDI	AC	750 g/Nil	0.0125	7	CC (60 A)	/	650 µS/cm	70.00%	0.234	85%	300 d	Voltea B.V.
2 2015	bipolar MCDI	AC	/	/	6.7	CV (51 V)	/	3277 µS/cm	74.90%	2.67	71.50%	15 d	EST Water & Technologies
3 2013	monopolar MCDI	Coated AC	1354 g/100	0.0246	7	CV (1.5 V)	1–5	1500 mg/L	70%	1.89	75–80%	15 d	LT Green Energy
4 2009	monopolar MCDI	Coated AC	/	/	8–10	CV (1.25 V)	3	150–3000 mg/L	80.9%–94.3%	0.45 to 5.35	63.9% to 95.8%	/	Current Water Technologies
5 2019	monopolar MCDI	Coated AC	80 g/50	0.008	2	CV (1.2 V)	/	800 µS/cm	51%	/	50%	15 d	Siontech
6 2021	monopolar MCDI	AC	Nil/40	/	0.1	CV (1.2 V)	9.2	1150 µS/cm	91%	0.172	75%	150 d	National Taiwan University

device. It is therefore necessary to search for cheaper alternatives. Compared with the free-standing IEMs ($\geq 100 \mu\text{m}$ thick), the ion-exchange coatings are significantly thinner ($2\text{--}50 \mu\text{m}$) and can be flatter, leading to lower membrane/contact resistances, higher energy efficiencies,^[26] and better fouling resistances.^[17] Nevertheless, research on ion-exchange coatings remains in the early stage and is worth further exploration.

- 3) Considering commercial standards, AC seems irreplaceable, although many advanced electrode materials/designs, e.g., intercalation materials and carbon nanotubes, are reported as promising candidates in scientific research. Even the lifetime of AC is far from the expected 1 to 2 years, equivalent to 35,000 to 50,000 cycles,^[27] which makes the implementation of IEMs or ion-exchange coatings necessary. In this case, optimizing the performance of AC (without IEMs/IEPs) is a financially feasible alternative to an industrial-scale CDI system. Surface chemistry is the key to enabling the stable cycling of AC electrodes.^[28] It should be noted that most commercialized AC used in CDI systems is primarily designed for EDLCs, which probably neglects the application standards of CDI. Those EDLC-AC are usually high-grade and more expensive (\$10–30/kg) than AC used in other applications (\$0.3–10/kg). In the case of the designated CDI-AC being developed in the future, CDI can potentially be more financially feasible.

To conclude, the high cost and high energy consumption of RO should not be overlooked, although it is considered a trusted desalination technology. Given that the deionized water produced by RO may be overqualified for many applications, CDI is suggested to take a supplementary role focusing on the treatment of brackish water streams, rather than directly competing with RO. Given the commercialization of CDI technology facing its bottleneck, the high capital cost (mainly due to IEMs) is the main challenge in AC-based MCDI. Omitting the usage of IEMs might be technically the easiest to maximize the cost-effectiveness but cycling stability remains a problem in membrane-free CDI. Modification/coating of AC electrodes holds great potential in resolving the existing issues while the development of new electrode materials should not be neglected on a long-term basis. Lastly, issues like brine water discharge and energy recovery process are also worth further investigations.

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Conflict of Interests

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords: Capacitive deionization • Pilot-scale • Membrane • Activated carbon • Real water stream

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