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Applications of Capacitive Deionization (CDI): Desalination, Softening, Selective Removal, and Energy Efficiency

Desalination

Revised

May ##, 2018

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Abstract

Capacitive deionization (CDI) has drawn a great attention as a promising desalination technology and due to the great potential, studies on CDI are now significantly increased. However, there have been no guideline of the developing strategies of the CDI technology for specific applications. Therefore, this study presents a critical review of the recent advances in CDI to meet the technical requirements on various applicable areas, with an emphasis on a hybrid system. This review paper summarizes the major progresses on the novel electrode materials in CDI for the brackish water desalination application. Moreover, it reviews the CDI and reverse osmosis (RO) integrated systems for both ultrapure water (UPW) production and the wastewater treatment. The CDI applicability on various industrial processes has been further discussed through two distinctive areas: (1) water softening and (2) selective removal for valuable heavy metals and nutrients (nitrate/phosphate). Additionally, it summarizes the outstanding improvements on energy efficiency of CDI, specifically focusing on the energy recovery and hybridization with energy producing technology (i.e., reverse electrodialysis (RED) and microbial fuel cell (MFC)). This review paper is expected to suggest the practical applications and guidelines for electrode material developments on specific applications.

Keywords: Capacitive deionization, Applications, Hybrid system, Energy efficiency, Selective removal, Desalination

1. Introduction

Utilizing the non-conventional water resources for drinking water production is inevitable in these days [1]. As an alternative to the sustainable water resources, brackish water desalination is considered to be one of the key strategic solutions to the increasing fresh water demand. Today, numerous processes have been applied for brackish water desalination: membrane based pressure driven (i.e., reverse osmosis (RO) and nanofiltration (NF)) and electrochemically driven (i.e., electrodialysis (ED) and (electrodeionization (EDI)) processes are by far the dominant technologies producing fresh water from various water resources [2, 3]. Although significant improvement of desalination technologies has been achieved, limitations such as high energy consumption and/or membrane fouling have been reported to be the critical problem [4, 5]. Therefore capacitive deionization (CDI), an electrochemical process removing the salt or charged species from the feed solution by the porous electrodes has emerged as the next-generation desalination technique [6].

The principle of CDI process relies on the electrical potential difference over a pair of electrodes to remove the charged species from the water. The removal mechanism of CDI is represented by the electrical double layer models in which ion removal is induced by electrosorption on the electrodes [7, 8]. As the absence of hydraulic pressure not only implies the potential to reduce operating cost but can also be beneficial for fouling control, compared to the pressure-driven membrane processes. Furthermore, relatively low voltage is required for CDI (less than 1.8 V) over conventional electrochemical-driven processes, as a result, CDI displays significant advantages in terms of low energy requirements with substantial water recovery [9]. It should be noted that when the applied voltage exceeds certain point (theoretically 1.23 V, causing water hydrolysis), Faradaic reaction was occurred, which could

be followed by the decreased desalination performance, energy efficiency, and electrode lifespan [10].

In recent years, CDI technology has been developed for improving both removal and energy efficiency with advanced CDI system called membrane-CDI (MCDI) and flow electrode-CDI (FCDI). MCDI, modified CDI system with integrating ion exchange membranes on the surface of the electrodes exhibited superior desalination performance compared to conventional CDI due to preventing the impact of co-ions [11-13]. More recently FCDI exhibits great potential for being the next-generation CDI system [14, 15]. As the FCDI process utilizes the flowing suspended carbon electrode materials, salts could be continuously desalinated, that is, no discharging stage is required.

Numerous CDI research areas have been conducted and a total of 536 papers were published since 2012, as depicted in Fig. 1. Most of the CDI studies focused on the electrode development (54.5%), followed by CDI applications (19.6%), whereas the rest are dedicated to better understand the energy efficiency, fundamental mechanisms, and novel systems. Specifically, a total of 15 review papers related to CDI have been published in the last five years, which strongly indicates the rapid evolution of the CDI technology. As per the most of the earlier research papers, published review papers also focus on the recent advantages on the electrode material development. Other review papers summarized the overall aspects of CDI technology. However, there have been few studies on suggesting guidelines for practical applications of CDI technology.

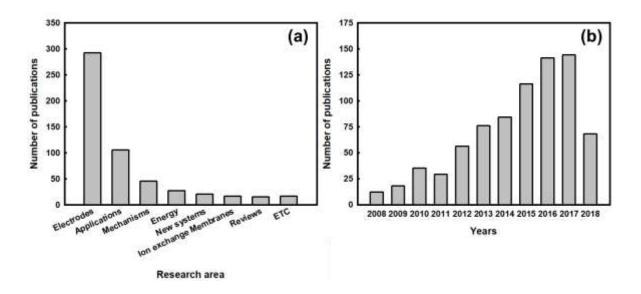


Figure 1 The number of publications depending on (a) the research areas of the CDI technology (2012-2017) and (b) the published year. Research papers were retrieved from the Scopus with capacitive deionization as the keyword in the title and the papers published in only past five years were categorized for research area analysis (Access on March 2018).

Therefore, this paper reviewed the practical applicability by treating various feed solutions with CDI strengths in terms of (i) brackish water desalination, (ii) water softening, (iii) selective removal (i.e., heavy metals or phosphate/nitrate), and (iv) energy efficiency as depicted in Fig. 2. This review paper is considered to suggest practical application areas and guideline on developing the CDI process for specific applications. For improving the desalination performance and energy efficiency, the hybrid system of CDI with other technologies (i.e., RO, reverse electrodialysis (RED), or microbial fuel cell (MFC)) were critically discussed.

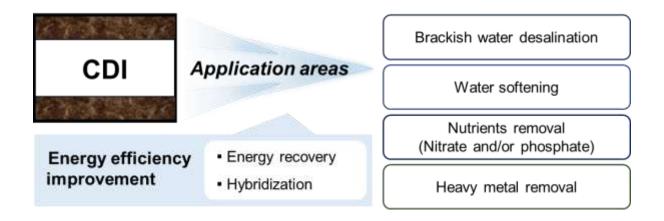


Figure 2 Possible application areas of CDI technology and practical strategies to improve the energy efficiency.

2. Brackish water desalination

2.1 Novel electrode developments in CDI

Electrode material development is one of the most fundamental solutions for higher desalination capacity as well as the removal efficiency of CDI process and therefore, the majority of recent studies focused on the development and synthesis of superior electrode material. Significant efforts have been made to develop high performance electrodes by synthesizing different carbon based materials such as activated carbon, carbide derived carbon (CDC), carbon aerogel, carbon nanotubes (CNTs), carbon nanofibers (CNFs), composites, graphene, etc. (Table 1).

Activated carbon was considered to be the most common electrode material due to its high surface area and optimized pore size distribution for the CDI process with relatively cheap and easy manufacturing characteristics [16]. However for better removal efficiency, other aforementioned materials have been widely investigated as the next generation electrodes due to (1) improved surface area (carbide-derived carbon (CDC) for about 1100 - 1300 m²/g, graphene for 2630 m²/g, and CNT for 77,415 m²/g) [17-19], (2) high salt adsorption capacity

(CDC for 28-44% increment) [17], (3) low electrical resistivity (carbon aerogel for <40 m Ω /cm) [20], or (4) optimized pore size distribution (carbon aerogel for < 50 nm) [21]. Furthermore, anion and cation exchange polymers integrated with CNTs electrodes were fabricated. It showed much higher removal efficiency (93%) compared to that of CDI (25%) or MCDI (74%) [22].

As summarized in Table 1, great efforts have been conducted on improving the performance of CDI by employing the novel electrodes. The newly synthesized or developed electrodes exhibited innovative removal efficiency, however, essence lies not only on the improved electrode performance but on the commercialization (i.e., cheap and easy to scale up). That is the reason why most of the companies Voltea (Nederland), EST Water & Technologies CO., Ltd (China), and Siontech (South Korea) supplying CDI units manufactured the CDI modules with the conventional activated carbon. Therefore consideration for feasibility of novel electrode on practical implication should be accompanied with improving the removal performance.

Table 1 Salt adsorption rate and removal efficiency of different types of CDI electrode materials and system types. As the salt adsorption rate and removal efficiency results were obtained from the lab-scale experiments, capacity of the CDI system was not stated.

Feed water TDS conc. (mg/l)	Flow rate (mL/min)	Salt adsorption rate (mg/g)	Salt Removal efficiency (%)	Applied voltage (V)	Electrode materials	CDI types	Remarks	Ref.				
1506	7000	-	75-82	- 1.5	And the London		Effect of hardness on long-term CDI operation	[23]				
1676 1500	7000	<u>-</u>	55-86 75-80	1.5	Activated carbon		Performance of commercial portable CDI unit					
2500	7000	10.5	27	1.6		-	renormance of commercial portable CDI unit	[24]				
1500	- 5	8.9	32	1.6	Activated carbon cloth		Desalination and disinfection properties of CDI					
2000		7.7	84	1.0		-						
2500	25	10.4	82	1.8	Activated carbon with		Fabrication and performance evaluation of the electrodes	[9]				
3000		11.2	71	- 1.0	TiO_2		prepared with titanium dioxide and activated carbon	[>]				
4000	6	8.9		1.2	Activated carbon fibre	_	Evaluation of performance of different types of activated carbon fibre electrodes in desalination	[26]				
2000	-	7	-	1.3			Desalination performance of brackish water and iodine recovery using CDI					
1168	-	13	-	1.2	Carbon aerogel CI	CDI	Desalination application using carbon aerogel electrode in CDI	[28]				
600	_		66			-						
700	_		60	– Mesonoroi	- Mesonorous carbon	Mesonorous carbon	Mesoporous carbon	Mesoporous carbon	Mesonorous carbon		Electrode fabrication and assessment of desalination	
1000	500	8.4	80	1.2	aerogel		performance	[29]				
1200	_		86	_								
1500			84			-						
1000	80	90	90	60	1.4	Carbon composite		Fabrication of composite electrode and evaluation of desalination performance	[30]			
1000	80	-	00	1.4	Carbon composite		Preparation of 3-Dimensional hierarchical porous carbon composite electrode	[31]				
		20.76	-	1.2	Graphene oxide/resorcinol- formaldehyde							
800	20	33.52	-	1.8			Synthesis of graphene-composite electrode for CDI	[32]				
500	25	16.1	-	1.2	Flexible cotton derived carbon sponge		Fabrication of cotton-derived carbon sponge electrode	[33]				
3970	_	14.6	35	_	1 5	-						
4460	30	14.5	31	1.2	Mesoporous carbon flat sheet with graphite		Synthesis of mesoporous carbon electrode for desalination	[34]				
3000		5.3	17.6		sheet with graphite							

1000	-	78.73	-	1.4	Polypyrrole/graphite		Investigation of dopants on the adsorbing performance of polypyrrole/graphite electrodes for CDI	[35]
500	1000	-	90.6	-	Activated carbon		Study demonstrating energy recovery potential of MCDI during desorption phase	[36]
503	-	-	70	-	Activated carbon	_	Practical application of MCDI for desalination of cooling tower feed water	[37]
1000	40	-	92	1.2	Activated carbon cloth		performance comparison between CDI and MCDI	[38]
-	40		93.5	1.6	Carbon nanotube- nanofibre	MCDI	Fabrication of carbon nanotube-carbon fibre composite electrode for MCDI desalination	[39]
-	50	5.6	93	1.2	Carbon nanotubes	_	Incorporation of ion exchange polymer directly on the electrode as modified MCDI	[22]
1000	9		90	1.2	Carbon fibre	_	Performance of different types of ion exchange membranes on MCDI	[40]

2.2 RO-CDI hybrid system

RO technology takes a significant place in desalination due to its excellent salt rejection, despite of some drawbacks, in terms of membrane fouling/scaling and high energy consumption. On the contrary, CDI can supplement the limitations of RO process and therefore integrating these two processes, RO-CDI hybrid system can improve the performance and energy efficiency which can possibly replace the conventional RO system. Two scenarios of RO-CDI hybrid systems: (1) RO-CDI pass system (CDI as RO permeate treatment) for ultrapure water (UPW) production and (2) RO-CDI stage system (CDI as RO brine treatment) for maximizing water recovery rate in wastewater treatment were critically discussed as below.

2.2.1. RO-CDI pass system (UPW production)

These days, for stringent drinking water regulation and increased demand for highly purified water for industrial areas, auxiliary processes for further RO permeate treatment is essential [41]. UPW, where the contaminants are removed to extremely low levels, is used in many industries such as power plants, pharmaceutical industries, etc. As such, the demand and market potential for UPW is on the rise [41] and the two-pass RO system is now widely adopted to meet the required water quality. Despite its high removal efficiency of the two-pass RO system, composed of first-pass RO and second-pass brackish water RO (BWRO) (Fig. 3(a)), high energy consumption has explored a new system. Therefore, RO-CDI pass system has been suggested to replace the conventional two-pass RO system (Fig. 3(b)).

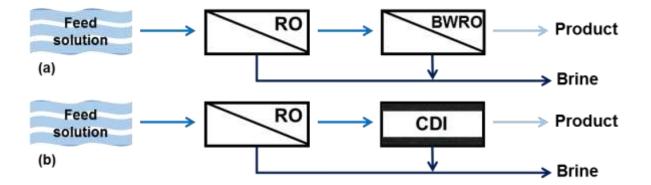


Figure 3 Schematic diagram of (a) conventional two pass RO and (b) RO-CDI pass system for producing the UPW. In RO-CDI pass system, RO permeate is further treated by the CDI process to achieve extremely high permeate water quality such as UPW.

Several studies demonstrated the CDI application potential for UPW production. An introduction of CDI as RO permeate polishing step can be an alternative solution due to the great energy efficiency [15, 41-43]. Furthermore, there should be little concerns on organic or inorganic fouling [44], which are the two main pollutant groups in the water as the feed water to CDI. Therefore, performance of CDI could be maximized without any fouling or scaling problems. In this respect, RO-CDI pass system was investigated to produce not only drinking water but also highly purified water such as UPW [42].

Previous studies simulated the water quality of UPW and fresh water from the RO-CDI pass system. With the increase of the feed solution concentrations, TDS concentrations of UPW and fresh water were gradually increased. Furthermore, by optimizing the system configuration and improving the CDI performance, TDS concentration of the UPW was drastically dropped to 0.035 mg/L (resistivity as 18.8 M Ω /cm) which could be used for semiconductor industry requiring the highest level of purity [41]. Furthermore, performance of the CDI was investigated under 10 mg/L of the feed solution TDS concentration and the UPW with resistivity from 2 to 9 M Ω /cm was produced under 1.5 V of the applied voltage [45]. Another

research simulated that a permeate quality of 2.1 mg/L can be produced even from the seawater (32,702 mg/L) with a removal efficiency of 99.9% [46].

Advanced researches exhibited a great potential of the RO-CDI pass system for substituting the conventional RO-EDI system with satisfactory product water quality and energy efficiency. EDI, combined with ED and ion exchange resin, is currently the most common process employed for UPW production system. Although EDI could produce extremely highly purified produced water, specific energy consumption (SEC) of EDI (0.39-2.11 kWh/m³) was relatively higher than that of CDI process (0.02-0.22 kWh/m³) depending on the applied voltage, flow rate, and feed solution TDS concentration [43, 47]. Another possible application of RO-CDI pass system could be bromide removal in seawater desalination. Recent study reported that adopting CDI on behalf of the conventional BWRO as the second pass process could decrease the energy consumption up to 40% when treating the 1st pass RO permeate [48]. There was no direct comparison of the cost related to capital investment between CDI and BWRO, however, feasibility of RO-CDI pass system for seawater desalination was definitely proved.

Previous studies convinced the successful application of the CDI process as the RO-CDI pass system for the UPW production as well as other various industrial applications due to the competent removal and energy efficiency. However, as most of the results were obtained from the simulation, further researches through the lab- and real-scale experiment should be investigated.

2.2.2 RO-CDI stage system (Wastewater treatment)

Besides the seawater desalination, RO recently broadens its application to industrial and municipal wastewater treatment. However, the production of RO brine is unavoidable during the RO process. It may cause adverse impacts on the environment or require high costs for treatment [49]. Therefore the treatment of RO brine is a global challenge for enhancing the overall water recovery rate [50] and thus, RO-CDI hybrid systems treating the RO brine by CDI process was proposed [51].

The simplified flow diagram of conventional two stage RO and RO-CDI stage systems is presented in Fig. 4. In the RO-CDI stage system (Fig. 4(b)), the RO brine is fed to the CDI instead of another RO process. When treating the municipal wastewater with high organic matter concentration, pretreatment was adopted before CDI process (Fig. 4(c)).

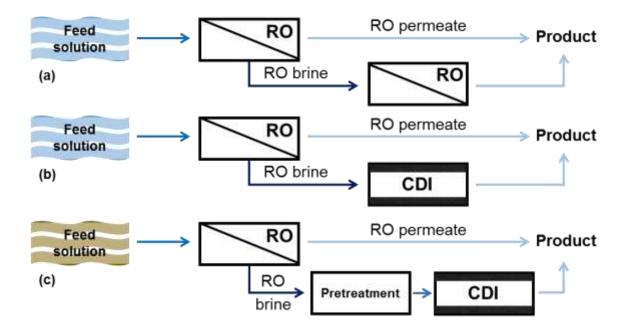


Figure 4 Schematic diagrams of RO-CDI stage systems (a) conventional two-stage RO system, (b) RO-CDI stage system for brackish water treatment, and (c) RO-CDI stage system for wastewater reclamation system. Biological activated carbon (BAC), microfiltration (MF), ultrafiltration (UF), pH adjustment, ozone, and sodium bisulfite were applied for pretreatment to remove organic matters before CDI process [52].

RO-CDI stage system was proposed for treating the industrial wastewater with 1,146 mg/L TDS concentration for water recovery increment [53]. The simulated results based on the advanced researches concluded that the CDI produced the permeate water quality as 497 mg/L when the RO brine was 1,686 mg/L, satisfying the drinking water regulation from World Health Organization (WHO). Here, RO brine was the feed solution to the CDI system. Furthermore, the energy consumption of RO-CDI stage system was about 19% decreased compared to the two-stage RO system depending on the energy recovery device efficiency [53]. Energy consumption of conventional two stage RO and RO-CDI stage system were briefly summarized in Table 2. Although the reported performances of RO-CDI stage system were simulated, verification with the experimental data was clearly conducted, which convinced the successful application of RO-CDI stage system for industrial wastewater treatment.

Table 2 Specific energy consumption of two stage RO and RO-CDI stage system for treating the industrial wastewater. The feed water to the second stage process was the brine from RO process and the TDS concentration was 1,686 mg/L.

System configuration	Pump efficiency (%)	SEC (kWh/m³)
	80	0.184
Two stage RO system	100	0.414
	80	0.149
RO-CDI system	100	0.335

In addition to the industrial wastewater treatment, the application of RO-CDI stage system extends its application to the domestic wastewater reclamation even in some major cities such as Barcelona, Tokyo, and Singapore [54]. Specifically in Singapore, NEWater project was developed to increase the feasibility and viability of using reclaimed water as a water source

for freshwater supply. As the feed water to NEWater project is the municipal wastewater, RO brine from water reclamation facility contains extremely high organic concentration (TOC: 15.0 - 31.1 mg/L). As the organic matters in the feed solution cause a severe fouling in the CDI cell, several pretreatments (i.e., biological activated carbon (BAC), microfiltration (MF), ultrafiltration (UF), pH adjustment, ozone, or sodium bisulfite) for removing substantial amount of organic matter were applied before CDI process. The inorganic matter removal efficiency of CDI process was from 86 to 92% for the water recovery from 78 to 89%. Furthermore, the overall water recovery of RO-CDI stage system was over 90% with about 15% lower energy consumption compared to the conventional two-stage RO system even considering the energy consumption for BAC [52]. Here, SEC of the pilot-scale CDI process with BAC pretreatment was 0.85 kWh/m³.

Even though the RO-CDI stage system exhibited the improved removal efficiency and water recovery rate, it was found that the tested pretreatment processes were not enough to remove the organics in the RO brine and thus, organic fouling was still the major problem in the CDI system. Furthermore, the energy consumption of CDI process was only evaluated, not for the entire system including pretreatment processes and RO process. Therefore, for sustainable CDI hybrid system operation, effective organic fouling control as well as fouling cleaning methods for CDI process should be further developed in addition to the system improvement.

3. Water softening

Water hardness due to presence of minerals such as calcium and magnesium ions causes serious problems for boilers and heat exchangers due to scale formation. Several processes have been adopted to reduce water hardness such as chemical precipitation, ion exchange processes, NF, RO, and ED [55]. However, most of these unit operations either consume high energy or require excessive chemicals usage, or generate a significant scaling on the surface of the membranes. Therefore, CDI has been examined as a new softening process because the CDI is operated by the electrical potential difference. Stronger attraction between the electrodes and multivalent ions convinces the feasibility of the CDI technology to be applied for the water softening. As such, the advanced researches investigated the feasibility of the CDI technology on the water softening application. (Table 3) [55-58].

Table 3 Removal efficiency of hardness depending on the CDI types and electrodes under various operating conditions (i.e., applied voltage and flow rate).

Hardness ion/ concentration	Applied voltage (V)	Flow rate (mL/min)	Removal efficiency	CDI type/electrode	Remarks	Ref.
350 mg/L as CaCO ₃	1.5	4	73%	CDI with carbon cloth electrode	Application of CDI in hardness removal using different types of activated carbon electrodes	[55]
10 mM, CaCl ₂	1.2	2	14.2 mg Ca ²⁺ /g	Ca-alginate coated on carbon electrode	Application of Ca-alginate coated MCDI for hardness control	[50]
10 mM, CaCl ₂	1.2	2	9.8 mg Ca ²⁺ /g	CDI with carbon electrode	— Application of Ca-arginate coated MCDI for narthess control	[59]
35 mg/L as CaCO ₃	2	10	3.5 mg CaCO ₃ /g	CDI with purified reduced graphene oxide electrode	Ultra-pure graphene oxide and reduced graphene oxide electrode for hardness control	[60]
266 mg/L, CaCl ₂	2	20	90%	CDI with activated carbon clotl electrode	Fabrication of carbon cloth electrode for hardness removal	[61]
46 mg/L, Ca ²⁺	1.2	28,000	74%			[37]
$5.1 \text{ mg/L}, \text{Mg}^{2+}$	$\overline{Mg^{2+}}$ 1.2 $\overline{28}$,		71%	electrode	desalination	[31]
1.45 mM, Ca ²⁺	2	10	58%	CDI with activated carbon electrode in a multi-ionic	In a colorativity study units CDI in a multi-legic consistence of	[62]
2.41 mM, Mg ²⁺	·	10	47%	environment	Ion selectivity study using CDI in a multi-ionic environment	
2.5 mM, CaCl ₂	1.5	-	~98% ~89% ~16% ~82%	CDI with SiO ₂ coating Carbon nanofoar carbon electrodes Carbon sheet	Removal efficiency of CDI depending on carbon materials and the effect of coating on the surface of the electrodes	[63]
	0.8	_	1.35 mg, CaSO ₄ /g			[64]
3 mM, CaSO ₄	1.0		2.97 mg, CaSO ₄ /g	CDI with SiO ₂ coating carbon	Effect of applied voltage and flow rate on the ion removal/regeneration, charge efficiency and energy	
3 mivi, Ca5O4	1.2		4.38 mg, CaSO ₄ /g	electrodes	consumption charge efficiency and energy	
		45	3.38 mg, CaSO ₄ /g			

Specifically, novel CDI electrodes for water softening have been aggressively developed. The removal of calcium ions was 44% higher with Ca-alginate coated electrode compared to conventional CDI due to increased charge efficiency (55% for CDI against 85% for Ca-alginate-CDI (CA-CDI)) [59]. Another coating material, nanoporous γ-Al₂O₃ or SiO₂ was coated on Porvair carbon for increasing the Ca²⁺ removal efficiency [65, 66]. With the coating on the surface of the carbon electrodes, specific surface area as well as the electrochemical properties of the electrodes was improved and the quantity of the removed Ca²⁺ was between four and five times higher than the uncoated electrodes. Coating material significantly influenced the Ca²⁺ removal efficiency, however, carbon material also takes great importance on the removal efficiency. When SiO₂ was coated on the carbon foam and/or nanofoam exhibited great removal efficiency (about 89-98%), on the other hands, the removal efficiency of the carbon cloth with SiO₂ coating was only 16% [63]. It should be also noted that applied voltage and flow rate directly determine the removal efficiency. Increasing voltage and flow rate resulted in increased removal efficiency [64].

In addition, a higher removal for calcium ions was reported for carbon cloth over composite electrode due to better wettability of the carbon cloth [55]. The authors also noted the importance of optimizing the pore size of the electrodes to improve the ion selectivity for divalent ions for possible water softening applications [67]. The interesting approach for hardness removal using ion exchange process under CDI was also reported by adopting zeolite modified carbon electrode for hardness removal through CDI-assisted ion exchange mechanism [68]. The application of external voltage led to remarkable increase in calcium removal. In a slightly different study, a novel concept was introduced, where a monovalent cation selective membrane was used in MCDI to produce divalent cation-rich solution as a means to stabilize permeate from NF or low pressure RO (LPRO) [69]. This is another example where MCDI can be innovatively configured to serve specific needs for different applications.

CDI has distinct advantages in water softening mainly for its low energy consumption as well as chemical-free process and because of that, Dutch company Voltea already supplies modular based CDI units for water softening. Although a previous research reported that there was little concern on the scaling problem in CDI technology [70], however, scaling on the spacer or the presence of ferric ion may deteriorate the product water quality. For that reasons, further investigation for sustainable operation of CDI on pilot or real scale plant could help meet overall system optimization which could be followed by the improved performance.

4. Selective removal

In recent years, the selective removal of specific ions from the feed solution has become critical issues in many industrial processes such as valuable resources recovery or toxic ion removal [1, 71, 72]. As a result, there are lots of innovations in technologies, however, generation of the secondary waste stream, higher energy consumption and higher capital cost were noted to be the inherent challenges [73, 74]. Therefore, the feasibility of CDI process for selective removal has been extensively explored in this section.

4.1 Heavy metal removal

Due to rapid industrial development, increasing amount of toxic heavy metals such as lead, cadmium, chromium etc. are released into the environment. A comprehensive review on the existing technologies for heavy metal removal was conducted [74]. The most common heavy metal removal technologies include chemical precipitation, ion-exchange, adsorption, membrane processes, coagulation and flocculation, and electrochemical processes. The review found certain disadvantages of the above processes. For example, chemical precipitation produces large volume of sludge and secondary waste, and they are good only for water

containing high concentration of ions. Similarly, ion-exchange also generates secondary waste during regeneration of resin, and they have limited large-scale applicability. Pressure driven membrane processes are known to be efficient in heavy metal removal but they have higher operating cost, and electrochemical processes have higher energy and capital cost. Since CDI is proven to be an energy efficient technology for low TDS water, and it does not use any chemical in the processes, it can be a viable technology for heavy metal removal.

There are several studies on CDI application in removing different species of heavy metals from various water sources (Table 4). Removal efficiency difference between lead and sodium was critically investigated depending on the pH, initial lead concentration, and time [75]. Lead was more sensitively affected by the pH changes and the removal efficiency was highest at the neutral pH condition (pH was 6). Another interesting results were the adsorption rate of lead and sodium. Due to the relatively high charge of lead compared to sodium, the adsorption equilibrium of lead was reached in a very short time, which indicates that highest removal selectivity between lead and sodium could be achieved with relatively lower operating time and neutral pH condition.

In addition to the study on lead removal, fundamental research on the removal efficiency of the two kinds of arsenic (As(\mathbf{V}) and As(\mathbf{II})) was also studied with different initial concentration of arsenic and applied voltage on the CDI system [76]. As expected, increased applied voltage and initial arsenic concentration resulted in the increased removal efficiency of both As(\mathbf{V}) and As(\mathbf{II}). However, the sorption capacity of As(\mathbf{V}) was higher than that of As(\mathbf{II}) under all of the operating conditions. Additionally, with the presence of NaCl or natural organic matter (NOM), removal efficiency of both arsenic ions were declined due to the competition effect. Specifically, NOM could potentially obstruct the electrode pores and therefore, specific surface area was decreased.

As the CDI process is operated with the potential difference between the pairs of the electrodes, charge and hydrated radius of the ions in the water determine the removal efficiency. Therefore, when considering the individual removal test, although cadmium and lead exhibits identical charge valence, due to the relatively large hydrated radius of the cadmium (4.26 and 4.01 Å of cadmium and lead, respectively), lead was more favorable removed. However, it should be noted that despite the larger hydrated radius of chromium than that of cadmium or lead, removal efficiency of the chromium was much higher [77]. Similar removal efficiency trend was observed when three metals were mixed in the solution with same concentration. Specifically, the removal efficiency of cadmium was significantly inhibited due to the combined effect of hydraulic radius and charge valance [73].

 Table 4 Removal efficiency of heavy metal ions by CDI.

Pollutants	Concentration	Electrodes	Removal efficiency	Remarks	Ref.	
	0.02-0.1 mg/L	Activated carbon	86-98%	Use of solar-powered CDI unit for arsenic removal from synthetic water	[78]	
Arsenic	0.1-200 mg/L	Activated carbon	0.01 - 0.025 mg, As(V)/g 0.005 - 0.014 mg, As(III)/g	Effect of applied voltage and initial concentration on the removal efficiency of As(V) and As(III)	[76]	
	0.2 mg/L	Activated	98.51%	Practical application of commercial CDI module for arsenic and lead	[79]	
	0.0267 mg/L	carbon	>60%	removal in Mexico	[//]	
Lead	5-100 mg/L	3D graphene	60-99%	Heavy metal separation from wastewater by CDI with 3D graphene- based asymmetric electrodes depending on pH, voltage, and	[75]	
	0.5 mM	A .: . 1	43%	A (CON 11 / 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1		
Cadmium	0.5 mM	Activated carbon	32%	- Assessment of CDI application in heavy metal (i.e., lead, cadmium, and chromium) removal	[77]	
	0.5 mM		52%	- and chromium) removar		
Chromium	2 mg/L	Carbon 99.6% aerogel		Application of CDI in removal of chromium from wastewater using carbon aerogel electrode	[73]	
	0.035 mg/L	Carbon	94%	Chromium ions removal using carbon aerogel electrodes for ground		
	0.025 mg/L	aerogel	88%	water remediation	[80]	
Copper	50 mg/L	Activated carbon	>90%	Copper ions removal from aqueous solution with CDI	[81]	

4.2 Phosphate and nitrate removal

Similarly to heavy metal species, high concentration of phosphates and nitrates contained in the wastewater from agriculture and industrial applications should be removed for preventing the environmental concerns such as eutrophication of water source [82, 83]. While phosphorus is an environmental pollutant, it is also an essential element for food production, which is in short supply, therefore, it is not only important to treat water for phosphorus removal but it also has to be recovered [84]. The common unit process for phosphorus removal and recovery from wastewater is the struvite precipitation and sludge incineration where P recovery between 90-98% and 75% respectively can be achieved but it has substantial investment due to high chemical and energy demand [84, 85].

Furthermore, full-scale technologies are already applied for nitrate removal [86], where maximum nitrate removal efficiency reported was 100% for biological process followed by membrane process such as RO (97%), ion-exchange (90%) and chemical process (70%). While the removal efficiencies have been reported to be quite good, there still remains the issues of operational cost, requirement of pre-treatment and post-treatment, and generation of secondary pollution as some of the challenges of these technologies. Therefore, several studies evaluated the removal efficiency of the phosphate and nitrate by the CDI process.

A pilot CDI unit (AQUA, EWP) was tested to evaluate phosphate removal and the removal efficiencies were 86% and 77% for feed water containing 50 and 300 mg/L phosphate, respectively [87]. They also observed lower removal efficiencies for higher initial phosphate concentration and higher flow rates with optimum pH between 5 and 6, achieving 98% removal with a three staged system. Another study also utilized same CDI unit for investigating the removal efficiency of nitrate at different initial nitrate (100, 300, and 500 mg/L of nitrate with 2.0 g/L, NaCl) and NaCl (300 mg/L of nitrate with 1.5, 2.0, and 3.0 g/L, NaCl) concentration, respectively [88]. Although the experimental data was obtained from the batch mode CDI

system operation, developed transport model was in good agreement with the tested results. Based on the model, removal trends of more diverse ion species could be further investigated. Likewise, a commercial CDI unit (DesEL Technology, ENPAR Tech. Inc.) was also used to assess nitrate removal from wastewater (88-98%, removal rate) [89].

In the separate study, the removal efficiency and selectivity of nitrate was significantly improved with the coated anion-exchange resin (BHP 55) with high nitrate selectivity [90, 91]. The resin employed in both studies preferably adsorbs nitrate ions than the other anions. Similar to BHP 55 coating, asymmetric carbon electrodes coated with SiO₂ and Al₂O₃⁻ were employed and higher nitrate removal capacity was observed compared to the uncoated symmetric carbon electrodes [92]. It could be explained by the faster ion transport due to the reduced hydrophobicity, increased specific surface area, and increased adsorption sites. Aforementioned studies improved the nitrate removal efficiency with coating on the surface of the carbon electrodes, on the other hands, a novel CDI electrode (Pd/NiAl-LMO film electrode) was newly developed [93]. Pd/NiAl-LMO electrode effectively capture nitrate and convert nitrate to nitrogen (N₂) through the regeneration period.

Removal selectivity of CDI is governed by the characteristics of feed contaminant (i.e., ionic charge, hydrated radius, and initial concentration of ions), operating conditions (i.e., applied voltage), and electrode properties (i.e., pore size, pore size distribution, and structure) [62, 73, 77, 94]. Therefore, comprehensive understanding for the factors influencing the removal selectivity is required and the electrode with optimized pore size and distribution should be further developed for the better removal selectivity.

5. Energy efficiency improvement

Due to the intensive energy consumption of conventional desalination techniques, CDI has emerged as the new technology for energy and cost effective desalination process. However, there still exists tremendous potential for reducing or recovering the energy. With further improvement of energy efficiency, CDI can be more cost competitive and broaden its applications. There are two primary strategies to enhance the energy efficiency of CDI: (1) energy recovery of CDI process during the desorption period and (2) hybridization of CDI with energy producing process.

5.1 Energy recovery of MCDI process

There have been great efforts on improving the energy efficiency of the MCDI technology and the energy recovery during the desorption stage [95-99]. The adsorbed ions in the electrical double layers move from the electrode into the feed stream and the electrons were transported over the external load simultaneously, which resulted in the energy recovery [36]. As summarized in Table 5, with the introduction of nanoporous activated carbon electrode, 30 to 45% of the consumed energy could be recovered [100], while under the optimized operating condition, up to 83.2% of energy recovery can be achieved. Indeed, the energy recovery rate can be more than 70% with inexpensive hydrophilic activated-charcoal based electrodes [101]. It was also argued that the energy recovery reached 40% for the conventional CDI cell [98]. Moreover, up to 83% of the used energy can be recovered in the regeneration phase under constant current condition using a membrane CDI stack [36]. Energy recovery was tested under two major operating conditions, constant voltage (CV) and constant current mode (CC). In order to identify optimum operating mode for higher energy recovery, energy recovery was tested under different voltages and currents for CV and CC mode, respectively [102]. From the

results, CC mode operation was found to be more advantageous than CV mode due to the higher energy storage.

The energy recovery ratio of various operating conditions (operating time, feed solution salinity, and charging current) and system configuration of CDI (cell size and electrode surface) were also investigated [98, 103]. The energy recovery ratio of the CDI system increased with the increment of feed solution salinity, applied voltage, but decrement of the cycle duration. Specifically, optimum electrode surface and current for minimizing the energy losses was critically simulated and validated [103, 104]. On the other hand, the size of the CDI system has little impact on the energy recovery ratio although increased system size may enhance the thermodynamic efficiency.

Table 5 Recent improvements on energy recovery of the CDI process. CC and CV respectively indicates constant current and constant voltage.

Maximum	CDI	Experimental conditions					
energy recovery (%)	type	Electrodes	Recovery mode	Feed solution concentration	Applied current for desorption	Remarks	Ref.
83	MCDI	Activated carbon	CC mode	8.6 - 86 mM	2 - 15 A	Energy recovery of MCDI depending on the feed solution quality and applied current	[36]
40	MCDI	Activated carbon	CC mode	20 mM	8.64 mA	Energy recovery in MCDI and CDI under CC and CV mode	[96]
20	FCDI	Activated carbon	CC mode	35 g/L	0.1 A	Performance and energy recovery in FCDI	[97]
29			CV	0.5 g/L		Effect of operating time, feed solution	
39	CDI Carboi	Carbon aerogel	mode	1.0 g/L	1 mA	concentrations, and CDI system sizes on energy	[98]
63	_	-	mode	$\frac{1.5 \text{ g/L}}{1.5 \text{ g/L}}$		recovery in CDI	
83	MCDI	Activated carbon	CC mode	0.5 - 1 g/L	1 A	Effect of water recovery and flow rate on energy recovery in MCDI	[105]

5.2 Hybridization of CDI with energy producing process

The increasing attention to the water-energy nexus encourages the development of technologies that efficiently remove the contaminants during water treatment as well as energy production from the water resources [106]. In this respect, the energy efficiency of CDI process is expected to be greatly improved by integrating with energy producing processes such as (1) RED and (2) MFC as illustrated in Fig. 5.

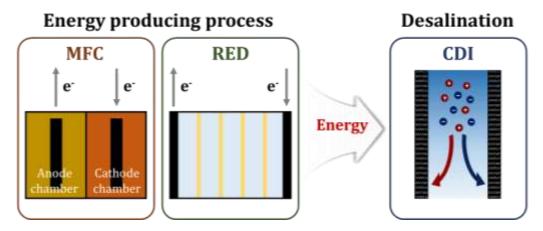


Figure 5 Schematic diagram of the CDI based integration system with the energy producing process (e.g., MFC and RED process). Harnessed energy by the MFC and RED process was utilized as the driving for of the CDI process.

5.2.1 CDI-RED hybrid system

RED, one of the emerging membrane-based technologies for producing energy from salinity gradient, has received attention these days. In a RED system, the chemical potential difference causes the transport of ions through the membranes from the concentrated solution to the diluted solution and therefore, electrical power can be sustainably generated [107].

The CDI-RED hybrid system has been investigated for decreasing the energy consumption of CDI operation by utilizing the inevitable waste brine as the electric energy resource [108, 109]. The feed solution is initially fed to the CDI cell and the outlet of the CDI system is divided

into four parts: fresh water, high concentration solution, low concentration solution, and the brine. Among them, high and low concentration solution were injected to RED system for energy production. Therefore, desalinated water and electric power can be produced at the same time by this hybrid system. The modeled result exhibited that the UPW (0.0001 mg/L) and fresh water (5.67 mg/L) were effectively produced with 46.5% decreased energy consumption compared to the simple CDI system.

5.2.2 CDI-MFC hybrid system

MFC has been developed to be a promising technology that enables both electricity generation and wastewater treatment with a single device [110]. MFC converts biochemical potential of the organic compounds to electrical energy through exoelectrogenic bacteria and thus, electricity is directly produced. However, the low quantity of the power generated by MFC hindered the direct applications of MFC [111].

In contrast with other electrochemical processes such as ED, CDI is operated at relatively low voltages and thus, the CDI-MFC hybrid systems were proposed for wastewater treatment and saline water desalination simultaneously without any energy input [112]. Typically, the CDI-MFC hybrid system is composed of two parallel processes: (1) MFC for wastewater treatment as well as electric power generation and (2) CDI for brackish water desalination (Fig. 6 (a)). The anode and cathode electrodes of MFC are directly connected with the activated carbon electrodes of CDI and the potential gradient generated in MFC system drives the ion transfer in the electrical field of CDI for brackish water deionization.

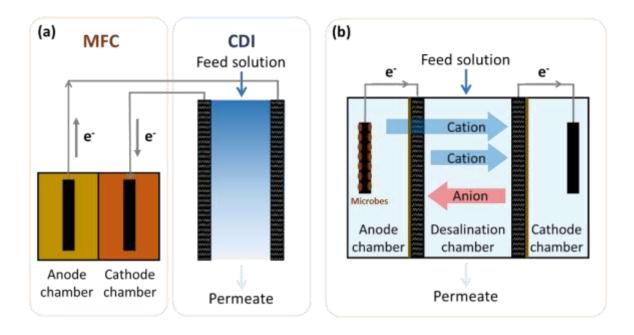


Figure 6 Schematic diagram of (a) CDI-MFC hybrid system and (b) MCDC system. The desalination chamber separated with anode and cathode chamber with two different assemblies. Cation exchange membrane is installed on the anode and cathode side assembly for preventing microbial growth.

The competent desalination efficiency of CDI process (73.0 - 79.2%) was observed when MFC supplied power to CDI [113]. Three different connections types of MFC to supply electrical potential to the CDI system: single MFC, MFCs in series, and MFCs in parallel were tested [112]. Three systems showed a similar trend of output voltage and removal efficiency of CDI system. Among them, parallel system exhibited highest performance, however, the lowest performance of the series system was observed due to the effectiveness of the electrons transfer leading to higher current in the system.

Furthermore, microbial desalination cells (MDCs), one of advanced MFC is emerged as a promising technology to simultaneously treat saline water and produce renewable energy [114]. On contrary to conventional MFC, the additional desalination function can be achieved in an MDC by adding a middle chamber containing saline water. Moreover, the anode–cathode

potential difference drives the migration of anions to the anode chamber and cations to the cathode chamber for charge neutrality [115]. In order to utilize the mutual benefits between the MDC and CDI, microbial capacitive desalination cell (MCDC), integrating MDC with the concept of CDI was investigated (Fig. 5 (b)) [116-118]. In MCDC system, saline water can be desalinated through electrical potential over the activated carbon driven by the microorganisms.

The average salt removal efficiency of the CDI (desalination chamber in MCDC system) during one cycle was 69.4% with 10 g/L NaCl feed solution [116]. In addition, to optimize the system configuration, several types of MCDC system were tested. When the feed water TDS was 1,000 mg/L, the removal rate of CDI was 61.2, 59.4, and 74.7% with single-, series, and parallel-configuration of MCDC system, respectively [116]. As shown in the CDI-MFC hybrid system, paralleled MDCs as the power supply to CDI exhibited the highest desalination performance due to larger amount of electrical potential generated from MDCs. Besides, the salt removal efficiency of the MCDI decreased with the increased operating time from 35.5 to 12.0%.

Several previous researches indicated that CDI could be competitive to RO in terms of energy efficiency, however, RO is generally applied for the seawater with 35,000 mg/L, TDS, not for the brackish water having the TDS concentration lower than 4,000 mg/L. This implies that there should be further improvement on the energy efficiency of CDI to overcome the conventional brackish water treatment process such as NF process. Fortunately, there have been great efforts for improving the energy efficiency of CDI process by utilizing the external elements including energy recovery during desorption period and integration with energy producing electrochemical processes. In addition to that, saving the energy consumption of the CDI process for desalination on adsorption phase and/or developing novel operating mode

[119] is considered to be the most critical. Therefore, guideline for optimization of the operating conditions, CDI module design, and novel electrode development may promise the commercialization of CDI process in various application areas.

6. Challenges in CDI technology: fouling, scaling, and scale-up

With expanding the application of CDI, harsher water resources (e.g., seawater or municipal wastewater) have been gradually investigated as the feed solution. Although the regeneration stage of CDI process mitigates formation of fouling layer or precipitation of components from the solution onto the electrode surface [120, 121], organic fouling or scaling potential still deteriorates the sustainability of CDI [7]. Specifically, fouling and/or scaling could decrease removal efficiency and permeate flow rate as well as increase energy consumption [70]. It is also noteworthy that sustainable operation of CDI is as critical as developing high removal performance electrodes or system, however, there have been only a few studies on organic fouling in CDI. For better understanding and improving the CDI sustainability, fundamental investigations of CDI on fouling/scaling mechanism should be a priority. Then, optimized pretreatment processes and cleaning methods with different feed solution properties for organic fouling control are considered to be essential. With these advances, successful and sustainable operation of CDI in the real application could be achieved.

Another limitation of recent studies on CDI technology is scale-up. There have been a few studies utilizing the pilot-scale CDI unit [122, 123], however, only the energy consumption of the CDI itself was reported. Even in a small pilot scale unit, the auxiliary energy consumption is considered to be essential to drive the cyclic operation which is unique for MCDI process: pre-charge step, charge step, discharge/cleaning step in a cycle, each step is delivered by electronically controlling the valves, and consumes extra energy. Therefore, in parallel with

the CDI technology development, specific equipment supporting the CDI system operation should be designed.

7. Perspectives and outlook for expanding the CDI applications

As reviewed in this paper, there are tremendous applicable areas of the CDI technology, however, most of the researches focused only on improving the removal efficiency of the CDI processes. Developing the optimized CDI system for specific application is considered to be more essential. Table 6 critically summarizes the practical guidelines for each application of the CDI process with respect to the optimized electrode, hybrid system and operating strategy.

For UPW production, electrodes with high removal efficiency should be employed with RO-CDI pass system or CDI-RED hybrid system. When employing the RO-CDI pass system, satisfactory product water quality is expected with great energy efficiency and CDI-RED system could guarantee extremely low energy consumption. On contrary, as wastewater caused severe fouling on the electrodes or ion exchange membranes, fouling control is considered to be the main concern. Furthermore, utilization of RO-CDI stage system or CDI-MFC system can significantly increase the water recovery and energy efficiency, respectively. For water softening and selective removal, optimized pore size and pore distribution of the electrodes for the target contaminant (e.g., divalent cations, heavy metals, phosphate, or nitrate) are recommended.

Another critical issue for practical application of the CDI process could be the pilot- and/or full-scale experiments. There have been some simulated performances and energy efficiency of the CDI processes from the previous studies, however, to implement the CDI process for the full-scale demonstrations, more actual applications through the pilot- and/or full-scale plants

should be further directed. Particularly, in addition to developing the CDI process itself, specific devices and system optimization for the successful operation of the CDI system need to be advanced. Based on our guidelines, the implementation of CDI technology is expected to be extended.

Table 6 Practical guidelines for expanding the application of CDI process with respect to electrode material, hybrid system and operating strategy.

Appli	cation	Electrode	Hybrid system	Operating strategy	
	UPW	High surface area High salt adsorption capacity	RO-CDI pass	High removal efficiency High energy efficiency High water recovery Fouling control	
	OFW	Low electrical resistivity Optimized pore size distribution	CDI-RED		
Desalination	Wastewater	Fouling resistance electrodes/ ion exchange membranes High surface area/salt adsorption	RO-CDI stage		
		Selective nutrients electroadsorption capacity	CDI-MFC		
Water softening		High wettability Optimized pore size to divalent cations	-	High removal efficiency Scaling control	
Selective removal	Heavy metal	- Optimized pore/ pore size distribution	-	Interaction between	
	Phosphate/nitrate	- Optimized pore/ pore size distribution	-	charged ions	

8. Concluding remarks

Although CDI is still an emerging technology, it is increasingly investigated as the next-generation water desalination process due to its competent removal, energy efficiency, and removal selectivity. The application potential for CDI seems promising and this review paper quantitatively estimates the feasibility of CDI and suggest the guidelines for various applicable areas. Major applicable areas and challenges of CDI are summarized as follows:

- Recent developments on novel electrodes of CDI technology encourage its application on brackish water treatment. Furthermore, RO-CDI hybrid system is expected to have significant potential for UPW production as well as wastewater treatment with great energy efficiency and sufficient removal efficiency.
- Removal selectivity of CDI has been reported to be the promising solution for water softening, heavy metal, and phosphate/nitrate removal. Most of the charged species were satisfactorily removed, however competition between the existing ions in the feed solution and scaling problem may deteriorate the removal selectivity. Thus, further fundamental studies of the interaction between the charged ions are recommended.
- Although CDI is considered as an energy efficient technology when treating brackish water, there still remains great potential in reducing the energy consumption. Energy recovery during desorption stage and hybrid system with energy producing technology (RED) significantly improved the energy efficiency and even no energy was required for CDI-MFC hybrid system. Consequently, practical guidelines for the CDI application were proposed.

Despite of the great applicability of CDI process, the sustainability of CDI process is questionable in terms of scaling and organic fouling. A large number of pre-treatments and cleaning methods were tested, however, CDI was still suffered from the fouling, which could

be followed by decreased charge efficiency and life span. Therefore, studies on the sustainable operation of CDI process should be carried out for further expanding the application of CDI technology.

Acknowledgements

This work is supported by the Korea Agency for Infrastructure Technology Advancement (KAIA) grant funded by the Ministry of Land, Infrastructure and Transport (Grant 18IFIP-B116952-03).

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