

Review

# Selective Ion Removal by Capacitive Deionization (CDI)-Based Technologies

Shuqian Chai <sup>1</sup>, Jiarui Xi <sup>1</sup>, Ling Chen <sup>1</sup>, Wei He <sup>2</sup>, Junjie Shen <sup>3,\*</sup> and Hui Gong <sup>1,\*</sup> 

<sup>1</sup> State Key Joint Laboratory of Environment Simulation and Pollution Control, School of Environment, Tongji University, Shanghai 200070, China; 2032821@tongji.edu.cn (S.C.); 18220502355@163.com (J.X.); chenling@tongji.edu.cn (L.C.)

<sup>2</sup> School of Engineering, University of Warwick, Coventry CV4 7AL, UK; wei.he.2@warwick.ac.uk

<sup>3</sup> Department of Chemical Engineering, University of Bath, Bath BA2 7AY, UK

\* Correspondence: j.shen@bath.ac.uk (J.S.); gonghui001@tongji.edu.cn (H.G.)

**Abstract:** Severe freshwater shortages and global pollution make selective removal of target ions from solutions of great significance for water purification and resource recovery. Capacitive deionization (CDI) removes charged ions and molecules from water by applying a low applied electric field across the electrodes and has received much attention due to its lower energy consumption and sustainability. Its application field has been expanding in the past few years. In this paper, we report an overview of the current status of selective ion removal in CDI. This paper also discusses the prospects of selective CDI, including desalination, water softening, heavy metal removal and recovery, nutrient removal, and other common ion removal techniques. The insights from this review will inform the implementation of CDI technology.

**Keywords:** capacitive deionization; selective ion removal; electrode materials; desalination; water softening; heavy metal; nutrients



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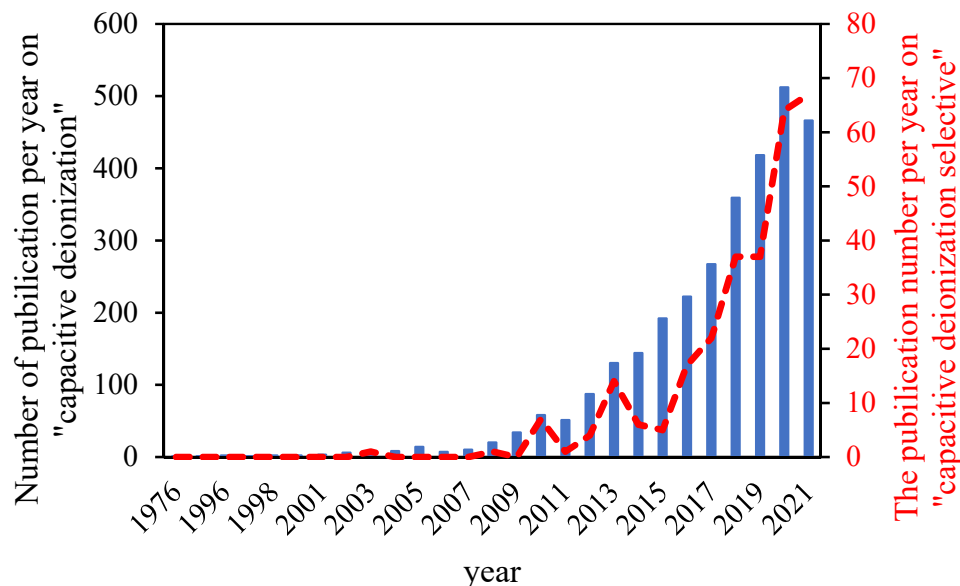
## 1. Introduction

Capacitive deionization (CDI) is an electrochemical-based technology that has drawn much attention recently due to its advantages of being energy-efficient and environmentally friendly [1,2]. The early research of CDI dates to the late 1960s; Blair et al. [3] and Arnold et al. [4] began to study brine desalination, which was then called electrochemical demineralization of water. In 1966, Caudle et al. [5] made electrodes with activated carbon powder for desalination in flow devices and introduced the concept of capacitive deionization (CDI). Until the last two decades, the academic interest in CDI exhibited an exponential increase, leading to tremendous advances [6].

As shown in Figure 1, CDI technology received more and more attention after 2000. From 2000 to 2010, the number of papers published each year generally showed an upward trend, although with fluctuations. After 2010, the high efficiency and energy-saving advantages of CDI were recognized, thus attracting more and more academic and application attention, with rapidly increasing publications.

The CDI process includes two stages of adsorption and desorption, achieving ion removal. After a small electrical potential (normally <1.5 V to avoid water electrolysis) is applied between two electrodes, the charged species in the water are adsorbed on the porous electrode material, generating desalinated water. The desorption process is that after the adsorption capacity of electrode materials is saturated, the two electrodes are short-circuited or reversed, and the adsorbed ions are released, producing a brine stream. The ion adsorption and desorption could be considered as charging and discharging of a capacitor, and the mechanism of ion adsorption on electrode could be explained by the electrical double layer (EDL) model [7]. In theory, CDI can remove any charged entity from water [8]. To overcome the shortcoming of frequent charging and discharging during the

CDI process, flow-electrode capacitive deionization (FCDI) was proposed, which makes use of a fluent electrode to achieve continuous ion removal. As technical requirement increases in ion removal in various regions, more CDI configuration is developed, including MCDI, FCDI, HCDI, and so on [9].



**Figure 1.** Numbers of papers with the theme of “capacitive deionization” and the number of publications per year on “capacitive deionization selective” from 1976 to 2021.

From the perspective of application, CDI technology was first used in the field of desalination and now is regarded as one of the most promising approaches, because it can overcome the shortcomings of traditional desalination technology, such as high energy demand and/or membrane fouling [10,11]. Then, CDI application was extended to various water treatment fields to remove ionized pollutants such as heavy metals and hardness ions. Recent studies have reported a broad application of CDI, such as water softening, heavy metal and nutrient removal, and other common anion removal techniques. For example, CDI could treat wastewater from several industries, such as mining, metallurgy, and printing circuits containing various heavy metal ions; CDI could remove  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  in the water softening field; CDI could treat groundwater which contains excessive concentrations of nitrate and fluoride.

Currently, the state-of-the-art CDI research is rapidly developing and evolving beyond conventional fields into new potential applications of recovering resources via CDI technology. Considering the fact that the types of ions contained in different wastewaters vary greatly, selective removal has become one of the most crucial functions of future CDI applications. If CDI can achieve selective removal of ions, its application fields will be further expanded, and better environmental benefits can be obtained.

For now, research on selective ion removal by capacitive deionization (CDI)-based technologies has been performed with remarkable progress, with various selectivity mechanisms based on more and more new published literature, which suggests that an updated review is useful for the academic community. A deep understanding of selective removal of target ions by CDI is still under development. This paper provides a comprehensive review of selective removal of target ions using CDI-based technologies. The selective mechanism and the current status of selective various ion adsorption are discussed to provide possible directions for the subsequent research on CDI technology.

## 2. The Main Factors Affecting Ion Selectivity

### 2.1. Ionic Properties of Ions to Be Selectively Removed

The ion selectivity of CDI in multi-ionic solutions has not yet been clearly elucidated. Ionic properties such as hydrated radius and ionic charge will affect the sequence of removal of different ions by CDI [12].

It was generally reported that ions with smaller hydrated radii and higher valences can be more effectively removed due to the EDL overlapping [13,14]. Table 1 [15] listed the valence and hydrated radius of various ions. Gabelich et al. [16] studied the electrosorption behaviors of carbon aerogel electrodes in different solutions and reported that ion selectivity was based on ionic hydrated radius and that monovalent ions with smaller hydrated radii were preferentially removed compared to multivalent ions with larger hydrated radii. However, studies on CDI research using multi-ion systems sometimes presents contradictory results. Gao et al. [17] reported that multivalent ions were preferentially adsorbed from the aqueous solution (the electrosorption capacities in the following order:  $\text{Fe}^{3+} > \text{Cu}^{2+} > \text{Zn}^{2+} > \text{Na}^+$ ), because cations with larger charge will be more easily adsorbed onto the electrode surface under the effect of electric field. For cations with same charge, the one with the smaller hydrated radius would be more effectively adsorbed (the electrosorption capacities in the following order:  $\text{Cu}^{2+} > \text{Zn}^{2+}$ ), because smaller cations can pass through the pores more easily. Yanting Li et al. [18] observed that the electrosorptive rates revealed an inversely proportional relationship with ionic hydrated radii. The order of electrosorptive rates followed  $\text{K}^+ > \text{Na}^+ > \text{Ca}^{2+} > \text{Mg}^{2+}$ . Mohamed Mossad et al. [19] came to the same conclusion as Gao, who found that the selectivity of cations followed the order  $\text{Fe}^{3+} > \text{Ca}^{2+} > \text{Mg}^{2+} > \text{Na}^+$ .

**Table 1.** Valence and hydrated radius of each ion.

Ions	Valence	Hydrated Radius (Å)
$\text{Li}^+$	+1	3.82
$\text{Na}^+$	+1	3.58
$\text{K}^+$	+1	3.31
$\text{Mg}^{2+}$	+2	4.28
$\text{Ca}^{2+}$	+2	4.12
$\text{NH}_4^+$	+1	3.31
$\text{Fe}^{2+}$	+2	4.28
$\text{Cu}^{2+}$	+2	4.19
$\text{Cd}^{2+}$	+2	4.26
$\text{Pb}^{2+}$	+2	4.01
$\text{Cr}^{3+}$	+3	4.61
$\text{Fe}^{3+}$	+3	4.57
$\text{F}^-$	-1	3.52
$\text{Cl}^-$	-1	3.32
$\text{Br}^-$	-1	3.30
$\text{I}^-$	-1	3.31
$\text{NO}_3^-$	-1	3.35
$\text{SO}_4^{2-}$	-2	3.79

Xu et al. [20] examined the ion selectivity of the carbon aerogel and reported that the amount of adsorbed ions by carbon aerogel (in mol/g aerogel) in treating produced water increased in the order of  $\text{Cl} (7.7 \times 10^{-5}) > \text{Br} (3.6 \times 10^{-6}) > \text{I} (2.6 \times 10^{-6})$  for anions. In their study, feed concentration seemed to play a more important role in ion uptake than the ionic hydrated radius. Ying et al. [21] reported that the sorption capacity of anions followed the order of  $\text{Br} > \text{Cl}$  at similar initial solution concentrations.

This means that ions with greater charge were more easily adsorbed. For ions having the same charge, the selectivity was completely dependent on hydrated radius.

## 2.2. Operation Parameters

### 2.2.1. Applied Voltage

According to the principle of CDI, voltage is the driving force that drives ions in solution to migrate from solution to the electrode surface, making them affect desalination efficiency and adsorption capacity. Tang et al. [22] described a CDI system to remove low concentrations of  $F^-$  in the presence of NaCl content and found that the higher the applied voltage (1.2 V), the lower the fluoride ion concentration in the effluent. Shu-Yun Huang et al. [23] found that the performance of the electric double-layer capacitor of carbon electrodes could be improved at a higher voltage (1.2 V). The removal capacity of  $Cu^{2+}$  at 1.2 V was 2.50 mg/g, which was considerably higher than that at 0.0 V (1.50 mg/g). Kim et al. [24] also demonstrated that the charge efficiency of a CDI can be improved by increasing the discharge voltage.

Theoretically, the higher is the voltage, the stronger is the electrostatic force, but the applied voltage of CDI is controlled below 1.5 V, as faradaic side reactions such as water electrolysis would occur when the voltage exceeds 1.23 V. In addition, the strong electrostatic forces may drive the migration of ions with high mobility, thus decreasing the selectivity to target ions. Li Wang et al. [25] reported that increasing the current density could reduce the ion selectivity. It should be noted that there is a trade-off between selectivity and removal rate, according to previous literature.

### 2.2.2. Solution Flow Rate

In general, a larger solution flow rate promoted more ions to reach the electrode surface, and, accordingly, the adsorption capacity of the electrode would reach the saturation state faster. If the flow rate is too large, the contact between the electrode surface and the solution ions will be insufficient, resulting in a decrease in the adsorption capacity and desalination efficiency. In addition, if the solution flow rate is low, it will lead to more time and energy consumption. Olga Pastushok et al. [26] reported that the decline of the electrosorption by the increase of the solution flow rate was attributed to the low contact time of the solution with electrodes inside the CDI. Huyskens et al. [27] found a similar phenomenon, which reported that a high flow rate increased the charge efficiency while at the same time resulting in a lower overall ion removal.

Therefore, the trade-off between deionization efficiency and energy consumption should be considered when using solution flow rate to achieve selective removal. Normally, the negative effect of a high flow rate could be minimized by the increase of a CDI unit size via a physical scaling-up. Generally, flow rate has a minor effect on selectivity of both anions and cations [28].

### 2.2.3. pH

The balance of ionization is strongly influenced by solution pH. The molecular forms of ions that belong to weak electrolytes depend on solution pH; for example, when the pH of the solution is above the acid dissociation constant ( $pK_a$ ) of hydrofluoric acid (HF,  $pK_a = 3.2$ ),  $F^-$  is the dominant species [29].

Tianting Pang et al. [30] studied the effect of different DIC species ( $H_2CO_3$ ,  $NaHCO_3$ , and  $Na_2CO_3$ ) on the removal of  $F^-$  in the MCDI system. DIC species in aqueous solution have three forms ( $H_2CO_3$ ,  $HCO_3^-$ , and  $CO_3^{2-}$ ), which vary with the pH. The results showed that different DIC forms have different effects on  $F^-$  removal. Enhao Zhu et al. [31] reported that a high pH is not conducive to selective removal of phosphate, as the  $OH^-$  strongly competes with  $PO_4^{3-}$  to be adsorbed when  $pH > 7$ . Choi et al. [28] found that the removal selectivity of  $Na^+$  and  $Ca^{2+}$  is the highest at neutral pH. The pH affects the electrical (zeta) potentials by adsorption of  $H^+$  or  $OH^-$  ions on the surface of electrodes. For example, in acidic conditions, the presence of more  $H^+$  causes the zeta potential to be shielded, thereby reducing the repulsive force between the cation exchange membrane and the cations.

The selective ion removal could be achieved by pH adjustment with various ion forms, including neutral state and charged stage.

### 2.3. Construction of CDI Reactor

#### 2.3.1. Electrode Materials

Generally, electrode materials can be divided into carbon electrodes and faradic electrode materials. Conventionally, carbon electrodes are usually the material of choice, in which ions are stored within the electric double layers (EDLs). Numerous carbon materials have been studied as potential electrode materials for CDI, including activated carbon (AC) [32], carbon aerogel [33,34], carbon nanotubes (CNTs) [35], and ordered mesoporous carbon [36].

Activated carbon is one of the most widely used and cost-effective electrode materials. Its price is low, and its specific surface area can reach as high as 1000~3500 m<sup>2</sup>/g. The conductivity of ordered mesoporous carbon is improved compared with activated carbon, thereby improving the CDI desalination performance. Carbon aerogel is another commonly used electrode material. It demonstrates a moderate specific surface area (about 400~1100 m<sup>2</sup>/g) and excellent electrical conductivity (conductivity can reach 25~100 S/cm) [37]. Carbon nanotubes were discovered in 1991 [38]; in these, pores are mainly located on the outer surface, and almost all of the specific surface area can be directly used, resulting in excellent CDI performance. Ordered mesoporous carbon possesses tunable and large pore sizes, high surface areas, periodically arranged monodispersed mesopore space, and alternative pore shapes [39].

As carbon materials store charge through the surface EDL, they are limited by the amount of charge storage. In addition, they display an insufficient selectivity toward various ions. Therefore, the new research interest on carbon electrodes is to use various methods to modify traditional materials in order to improve their adsorption capacity. Hawks et al. [40] reported carbon electrodes with subnanometer scale pores that could be used as an electro-sorbent for selectively removing nitrate. Kaustub Singh et al. [41] introduced an intercalation material called vanadium hexacyanoferricyanate (VHCF) as electrode material and observed a preference for VHCF toward divalent ions. Meanwhile, the addition of selective ion exchange coatings on the surface of a carbon electrode was also an applicable approach to achieve selective removal of target ions. Jun Kim [42] applied a Ca<sup>2+</sup>-selective nanocomposite (CSN) coating on the activated carbon electrode to achieve calcium-selective removal.

Although carbon-based materials have made great progress, unavoidable side reactions (such as oxidation of carbon anode [43] and co-ion expulsion effect [44]) hinder carbon-based materials' advancement and application in desalination of water with high salt concentration [45].

To overcome the limitations of the CDI in carbon-based electrode materials, faradic electrodes have been proposed. Recent studies have found that faradic electrode materials have salt removal capacities (>40 mg/g) much higher than carbon materials (~15 mg/g) [46] and indicate the advantages of low energy consumption and ion selectivity. Faradic electrode materials capture ions by relying on faradic reactions. Ions are inserted or stored in the lattice crystal structure sites of the intercalation host compounds because of redox reactions [45]. Intercalation materials such as MXenes [47], Prussian blue [48], and manganese oxides [49] have been studied as CDI electrode materials and showed good performance.

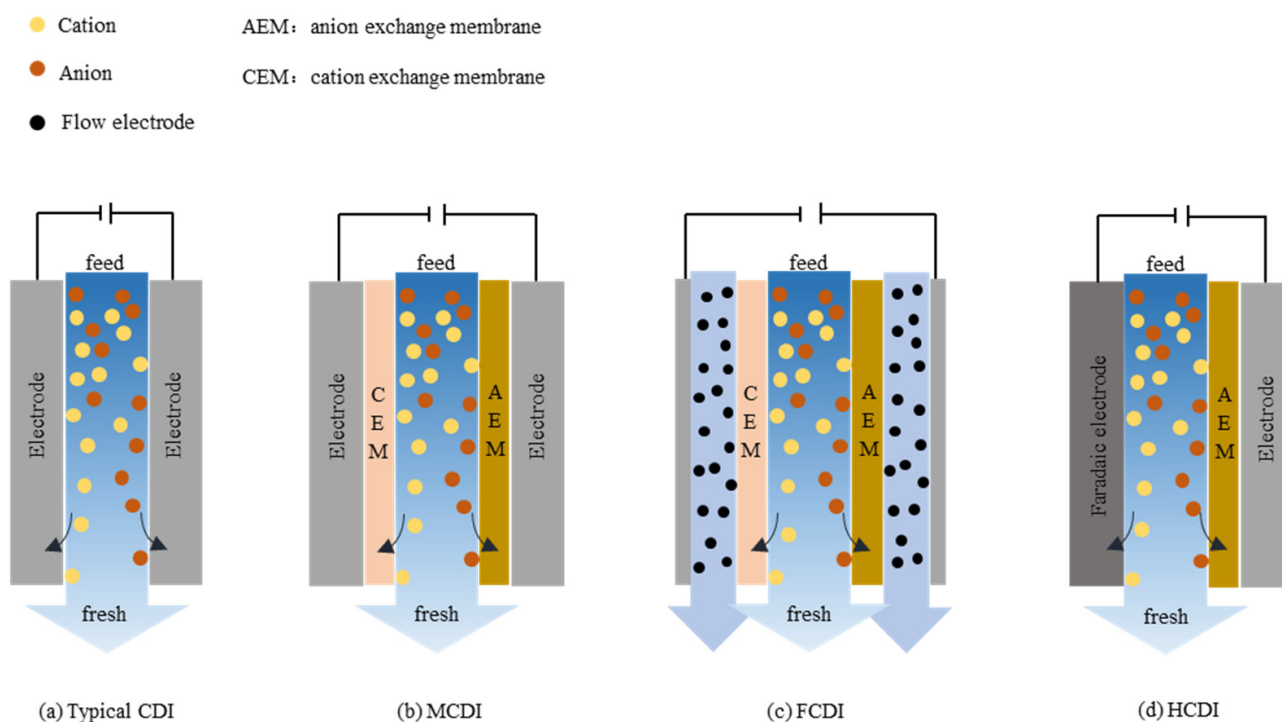
The current research on faradic materials mainly focuses on the removal of sodium ions and chloride ions [50]. Limited studies reported that faradic electrode materials have good selectivity for cations (such as Li<sup>+</sup>, Na<sup>+</sup>, K<sup>+</sup>) and have limited ability to remove anions (such as Cl<sup>-</sup>) [45]. Thus, ion exchange membranes have been used to overcome this limitation. Leandro L et al. [51] reported that Li<sup>+</sup> can be inserted in  $\lambda$ -MnO<sub>2</sub> cubic phase and extracted from aqueous solutions by a topotactic Li<sup>+</sup> insertion reaction. Seonghwan Kim et al. [52] studied the selectivity of sodium manganese oxide (Na<sub>0.44</sub>MnO<sub>2</sub>) for Na<sup>+</sup>, K<sup>+</sup>, Mg<sup>2+</sup>, and Ca<sup>2+</sup>. The results showed the high selectivity of Na<sub>0.44-x</sub>MnO<sub>2</sub> for Na<sup>+</sup> over

other cations, having over 13 times higher selectivity for  $\text{Na}^+$  than for  $\text{K}^+$  and 6–8 times higher selectivity for  $\text{Na}^+$  than for  $\text{Mg}^{2+}$  and  $\text{Ca}^{2+}$ .

### 2.3.2. Cell Architectures

Under certain operating conditions and electrode materials, the selective removal performance can be enhanced by optimizing the cell architectures.

As shown in Figure 2, a typical CDI cell is composed of two current collectors used for electron transmission and a spacer channel for feed water transportation [6].



**Figure 2.** Schematic of (a) typical CDI; (b) MCDI; (c) FCDI; and (d) HCDI.

Typical CDI mostly uses carbon-based materials as electrodes, which realize selectivity mainly by different physical properties of ions or modifying carbon materials. In the CDI device, not all of the consumed electric charges are used to adsorb the counterions, and part of the charges will be used to drive the same ions away from the electrode surface, resulting in the current efficiency being much lower than the theoretical value. In order to reduce the influence of the common ion effect and improve ion removal ability, researchers have designed several CDI cell architectures to enhance the CDI performance.

Membrane capacitive deionization (MCDI) is a modification of CDI by placing ion exchange membranes (IEMs) in front of each electrode. IEMs allow the transport of counterions and repulse co-ions by fixed charge groups on the membrane skeleton. Recent studies demonstrate that membranes can participate in selective ion removal in CDI.

Dong et al. [53] studied the influence of IEMs on the selective removal of  $\text{Pb}^{2+}$  by CDI. The results showed that IEMs allow the selective removal of  $\text{Pb}^{2+}$  against  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$ . The mechanism of selective removal of  $\text{Pb}^{2+}$  is believed to be a replacement process. The relatively strong affinity between  $\text{Pb}^{2+}$  and the functional groups allows  $\text{Pb}^{2+}$  to replace the  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  arriving earlier on the electrode. Jongmoon Choi et al. [28] reported that a monovalent cation permselective exchange membrane is more effective for selective removal of monovalent cations and requires less energy. M. Bryjak et al. [54] reported that MCDI with lithium-selective membranes can achieve lithium recovery effectively.

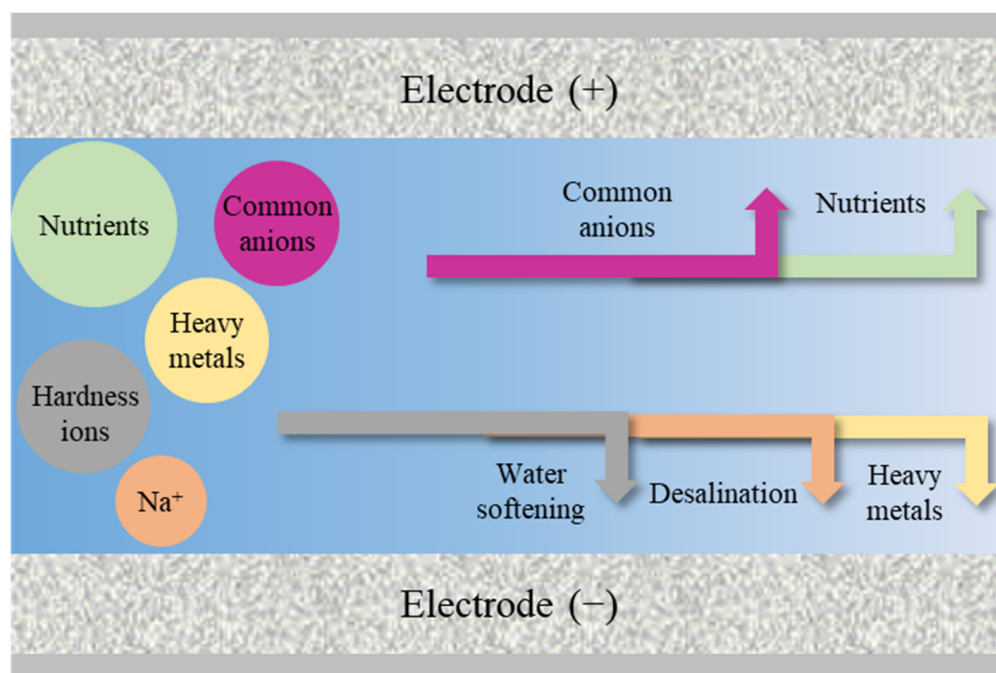
The major problem with CDI and MCDI is the requirement of an additional regeneration step to remove ions from saturated electrodes after the desalination step, which leads to discontinuous running. To overcome this limitation, the flow-electrode capacitive

deionization (FCDI) process has been proposed [55]. The FCDI systems are made up of two flow electrodes and ion exchange membranes. Because the electrode of FCDI is flowable during its operation, the electrode flowing out of the electrode chamber can be desorbed by collision outside the electrode chamber without a separate desorption process, which achieves continuous charge–discharge operations and greatly improves desalination efficiency. However, there are few studies on selective ion removal in the FCDI field [56]. Most research and applications are aimed at the removal of salt ions from influent water, and the flow electrode materials used in FCDI usually lack ion selectivity in the adsorption process. In order to remove and recover specific ions from wastewater, how to make flow electrodes with high adsorption selectivity is critical and challenging in future research and might become a new research hotspot.

Hybrid CDI (HCDI) [57,58] is a new CDI architecture that is composed of a faradaic electrode, anion exchange membrane, and a carbon electrode. In HCDI, cations are removed from the solution by faradaic electrode, while anions are held in the EDL formed at the surface of the carbon electrode as shown in Figure 2d.

### 3. Possible Application Areas of CDI

We systematically reviewed the possible application of CDI, as shown in Figure 3: (I) desalination, (II) water softening (e.g., hardness ions), (III) heavy metal removal and recovery, (IV) nutrients ( $\text{NO}_3^-$ ,  $\text{NO}_2^-$ , and  $\text{PO}_4^{3-}$ ), and (V) other common anions (e.g.,  $\text{SO}_4^{2-}$ ,  $\text{F}^-$ ,  $\text{Br}^-$ , and  $\text{Cl}^-$ ).



**Figure 3.** Possible application areas of CDI.

#### 3.1. Desalination

Commonly utilized desalination techniques include reverse osmosis (RO) and thermal separation methods. These techniques have downsides, including high energy consumption and complex operation [59]. Capacitive deionization (CDI) is an emerging and attractive process for salt removal, which has the potential to become an alternative to existing seawater and brackish water desalination technologies.

Recent studies have focused on electrode material development, because it is one of the most fundamental methods to achieve higher desalination capacity and removal efficiency in CDI [60]. The  $\text{Na}^+$  removal electrode has evolved from a carbon-based electrode to a faradaic electrode, and the desalination performance increased significantly. The development of  $\text{Cl}^-$  removal electrodes has been relatively slow; most research employed

activated carbon, Ag/AgCl, Bi/BiOCl to remove  $\text{Cl}^-$  [61]. Xu et al. [62] reported that using mesoporous carbon nano-polyhedra as electrode materials to treat brackish water could achieve a high desalination capacity of  $14.58 \text{ mg g}^{-1}$ . Xingtao Xu et al. [63] fabricated a novel nitrogen-doped graphene sponge (NGS) as CDI electrodes. The results showed that NGS exhibits an ultrahigh electrosorption capacity of  $21.0 \text{ mg/g}$  in  $\sim 500 \text{ mg/L}$  NaCl solution. Mauro Pasta et al. [64] reported that Ag// $\text{Na}_2\text{Mn}_5\text{O}_{10}$  used as the electrode could achieve a better selective removal of NaCl. When the salt removal rate was 25%, the Coulombic efficiencies of  $\text{Na}^+$  and  $\text{Cl}^-$  removal were 47% and 87%, respectively, significantly higher than the other coexisting ions. Chen et al. [65] were desalted using the BiOCl// $\text{Na}_{0.44}\text{MnO}_2$  electrode;  $\text{Cl}^-$  is captured at the BiOCl electrode, and  $\text{Na}^+$  is chemically inserted into the  $\text{Na}_{0.44}\text{MnO}_2$  electrode. Min et al. [66] reported that the Bi/rGO electrode had a high selectivity for  $\text{Cl}^-$  ( $\text{Cl}^- > \text{F}^- \gg \text{SO}_4^{2-}$ ), and the adsorption capacity for  $\text{Cl}^-$  in the mixed solution could reach as high as  $62.59 \text{ mg}\cdot\text{g}^{-1}$ .

Researchers also showed interest in the selective adsorption of other common anions (Table 2), such as  $\text{SO}_4^{2-}$ ,  $\text{F}^-$ ,  $\text{Br}^-$ , and  $\text{Cl}^-$ .

**Table 2.** Summary of the selective adsorption of other common anions in the literature.

Target Ions	Competing Ions	The Order of Electrosorption	Methods/Electrode Material	Ref.
$\text{SO}_4^{2-}$	$\text{Cl}^-$	$\text{SO}_4^{2-} > \text{Cl}^-$	Resin/QPVA Coated Electrode	[67]
$\text{SO}_4^{2-}$	$\text{Cl}^-$ , $\text{H}_2\text{PO}_4^-$	$\text{SO}_4^{2-} > \text{Cl}^- > \text{H}_2\text{PO}_4^-$	Amino-modified activated carbon	[68]
$\text{SO}_4^{2-}$	$\text{Cl}^-$	$\text{SO}_4^{2-} > \text{Cl}^-$	Optimization of operating parameters	[69]
$\text{F}^-$	$\text{Cl}^-$	-	Optimization of operating parameters	[22]
$\text{F}^-$	$\text{Cl}^-$	$\text{Cl}^- > \text{F}^-$	Optimization of operating parameters	[70]
$\text{F}^-$ , $\text{Cr}^{6+}$	-	-	Modeling and kinetic study	[71]
$\text{F}^-$	$\text{Cl}^-$ , $\text{NO}_3^-$	-	Using rGO/HA electrode	[72]
$\text{F}^-$	$\text{HCO}_3^-$ , $\text{CO}_3^{2-}$	$\text{CO}_3^{2-} > \text{HCO}_3^- > \text{F}^-$	Optimization of operation parameters	[30]
$\text{Br}^-$	$\text{Cl}^-$	$\text{Br}^- > \text{Cl}^-$	Activated carbon cloth electrode	[73]
$\text{Cl}^-$	-	-	Bi anode and Activated carbon cathode	[74]
$\text{Cl}^-$	$\text{SO}_4^{2-}$	$\text{Cl}^- > \text{SO}_4^{2-}$	Using Bi electrode	[75]

### 3.2. Water Softening

The presence of minerals of hardness in water, such as  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$ , causes scaling problems in boilers and heat exchangers. Various methods have been applied to remove hardness ions in water, including chemical precipitation, nanofiltration, electrodialysis, and so on [76]. However, these processes suffer from high energy consumption and overuse of chemicals. CDI has been considered as a new softening process. Table 3 summarized the recent studies of CDI in water softening.

Jun Kim et al. [42] developed a calcium-selective nanocomposite (CSN) electrode and showed that the  $\text{Na}^+$  cations are preferentially adsorbed in the EDLs and that later they are gradually replaced by the  $\text{Ca}^{2+}$  cations. Similar displacement phenomena were also observed in other studies [77], referred to as “time-dependent ion selectivity” [78].

It is well known that higher valence ions with smaller hydrated radii can be more effectively removed because of stronger electrostatic force [79]. The magnesium ion is reported to have a similar deionization trend to the calcium ion, because magnesium has the same charge valence and similar hydrated radius as calcium ion [80]. Therefore, the preferential ion electrosorption is estimated as follows:  $\text{Ca}^{2+} > \text{Mg}^{2+} > \text{Na}^+$  [81].



**Table 3.** Summary of the selective adsorption of hardness ions in the literature.

Target Ions	Competing Ions	The Order of Electrosorption	Methods/Electrode Material	Ref.
Ca <sup>2+</sup>	Na <sup>+</sup>	Ca <sup>2+</sup> > Na <sup>+</sup>	Applying a calcium selective nanocomposite coating	[42]
Ca <sup>2+</sup>	Na <sup>+</sup>	Ca <sup>2+</sup> > Na <sup>+</sup>	Ca-alginate as coating material on a negative electrode	[77]
Ca <sup>2+</sup> , Mg <sup>2+</sup>	Na <sup>+</sup>	Ca <sup>2+</sup> > Mg <sup>2+</sup> > Na <sup>+</sup>	CNTs/Ca-Selective zeolite composite electrodes	[81]
Ca <sup>2+</sup>	Na <sup>+</sup>	Ca <sup>2+</sup> > Na <sup>+</sup>	Parameter control	[78]
Ca <sup>2+</sup> , Mg <sup>2+</sup>	Na <sup>+</sup>	Ca <sup>2+</sup> > Mg <sup>2+</sup> > Na <sup>+</sup>	Activated carbon cloth and composites electrodes	[80]

### 3.3. Heavy Metal Removal and Recovery

Heavy metals are increasing in the environment due to urbanization, industrialization, and chemicalized agricultural activities [82]. Most metals are non-biodegradable and mobile in aqueous systems and tend to accumulate in living tissues and cause diseases [83]. Arsenic, lead, and mercury occupy the topmost position in the list of hazardous substances toxic to public health as per the US Environmental Protection Agency (EPA) and the Agency for Toxic Substances and Disease Registry of the US Department of Health and Human Services [84]. These heavy metals have cumulative effects in the human body, causing varying degrees of damage to human organs, even after exposure to minor amounts [85,86].

Previous studies showed that the CDI process can effectively remove individual metals (Cu<sup>2+</sup> [87], Cr<sup>3+</sup> [88,89], Ni<sup>2+</sup> [90], and Fe<sup>3+</sup> [13]) from aqueous solutions. Since it is common for multiple metal ions to coexist in wastewater, it is significant to understand the competitive removal behavior of these ions in CDI [91].

Zhe Huang et al. [83] investigated the viability and preference of CDI in removing cadmium (Cd<sup>2+</sup>), lead (Pb<sup>2+</sup>), and chromium (Cr<sup>3+</sup>) ions and reported that the removal of Cd<sup>2+</sup> was largely inhibited in the presence of Pb<sup>2+</sup> and Cr<sup>3+</sup>, because Cr<sup>3+</sup> had a higher charge and Pb<sup>2+</sup> had a smaller hydraulic radius. Minlin Mao et al. [92] found the MoO<sub>2</sub>/C spheres as cathodes showed a high selective capacitive removal efficiency of Pb<sup>2+</sup> from the multi-component system. Kim et al. [93] studied the recovery of palladium (Pd) from plating industry wastewater and achieved highly competitive Pd removal. Seoni Kim et al. [94] found that using the λ-MnO<sub>2</sub> electrode could effectively capture lithium ions. Fan et al. [95] investigated the feasibility of removing arsenic from groundwater in the presence of multiple ions. The results showed that the effluent arsenic concentration reduced to 0.03 mg/L.

### 3.4. Nutrient Removal and Recovery

The concentration of nutrients (such as NO<sub>3</sub><sup>-</sup>, NO<sub>2</sub><sup>-</sup>, and PO<sub>4</sub><sup>3-</sup>) is gradually increasing in municipal and agricultural wastewater due to contamination of soil and rivers caused by the excessive use of fertilizers and various organic pollutants [96,97]. Nitrate ions in drinking water are known to cause methemoglobinemia in infants. The EPA [98] has established the maximum contaminant level (MCL) for NO<sub>3</sub><sup>-</sup> of 10 mg/L in drinking water with reference dose (RfD) 1.6 mg/kg/day. NO<sub>2</sub><sup>-</sup> is abundant in the environment due to the extensive utilization of highly nitrogen-rich compounds. The EPA [98] has regulated the MCL for NO<sub>2</sub><sup>-</sup> of 1 mg/L in drinking water with RfD 0.16 mg/kg/day.

Aquatic plants such as algae can absorb and utilize various forms of nitrogen and obtain nitrogen from the atmosphere through biological nitrogen fixation to make up for the lack of nitrogen in the growing environment. However, phosphorus can only be obtained through the water environment, so the proliferation of algae in freshwater systems is generally controlled by phosphorus. Meanwhile, phosphorus is a non-renewable resource and its presence in nature is limited [99]. Removing and recovering nutrients in wastewater can mitigate the challenges of both environmental systems and fertilizer demands while improving water quality [100].

Today, several technologies have been already applied for nutrient removal, such as RO, chemical, and biological processes. However, these technologies face the problems of

high operational costs and secondary pollution. CDI is a promising alternative for nutrient removal and recovery, because the concentrations of nutrient species (such as  $\text{NO}_3^-$ ,  $\text{NO}_2^-$ , and  $\text{PO}_4^{3-}$ ) after secondary treatment are not very high. These species can be separated when exposed in a low electrical field (normally  $< 1.5$  V).

Recent studies showed the feasibility of using CDI for nutrient removal and recovery. Yu-Jin Kim et al. [101] fabricated a novel nitrate-selective composite carbon electrode (NSCCE) that exhibited high selectivity for nitrate ions. The results showed that the adsorption of nitrate ions was  $19 \text{ mmol/m}^2$ , a 2.3-fold increase over the adsorption achieved using the MCDI system. Olga Pastushok et al. [26] observed a slight decline in the removal efficiency of nitrate ions from 48 to 37% in the multi-ions system in comparison to the single-component nitrate system. Xu et al. [102] achieved the selective recovery of phosphorus by adjusting the charging and discharging progress of the FCDI. Yanhong Bian et al. [103] presented that FCDI could concurrently remove nitrogen and phosphate from wastewater and recovered them as concentrate efficiently.

#### 4. Conclusions and Future Perspectives

CDI is one of the novel ion removal technologies which has gradually attracted people's attention in recent years. The selective removal of ions will greatly expand the application scenarios of CDI. Seawater desalination is the original application field of CDI, and it is also the most researched and mature field. In addition, CDI also showed good ion removal performance in the fields of water softening, heavy metal removal, and nutrient element removal, but most of the research was still in the laboratory stage.

Promoting CDI from laboratory to practical application requires further exploration from the following aspects:

- (I) For different target ions, the ionic properties need to be paid attention first, and the hydration radius and valence of the ions often have an important impact on the selective adsorption of CDI.
- (II) Choosing appropriate operating parameters will also effectively improve the ion selectivity of CDI.
- (III) From the initial carbon-based materials to Faraday electrode materials, the development of electrode materials is one of the key factors to achieve CDI ion selectivity. In addition, under certain operating conditions and electrode materials, the selective removal performance can be enhanced by optimizing the cell architectures.

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