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Review

Application of Capacitive Deionization in Water Treatment and Energy Recovery: A Review

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Abstract: Water resources are the basis for human survival and development. However, human beings face severe challenges of water pollution and freshwater shortage. With the critical advantages of low energy consumption, high efficiency, low cost, green and pollution-free, and renewable electrodes, capacitive deionization (CDI) has become an up-and-coming water treatment technology. After decades of development, the application of CDI has expanded from seawater desalination to many fields. However, the existing literature still needs a comprehensive overview of the multi-functional application of CDI technology in water treatment. Therefore, our work critically reviewed the latest research progress of CDI in water treatment to meet the technical requirements of various application fields. This paper first summarizes the various applications of CDI in water treatment, focusing on CDI's representative research results in heavy metal removal, organic contaminants removal, water softening, phosphate and nitrate removal, and water disinfection. In addition, we also discussed the latest research progress of energy recovery and energy consumption assessment for the CDI process. Finally, this paper discusses the challenges and future opportunities facing CDI technology.

Keywords: capacitive deionization; water treatment; water softening; heavy metal; phosphate and nitrate; organic contaminant; water disinfection; energy recovery; energy consumption assessment

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1. Introduction

The rapid development of industry and agriculture in the world has caused severe water pollution. At the same time, the increase in population numbers has led to a shortage of water resources. According to statistics, 80% of the wastewater produced in the world is discharged into the environment without purification treatment. These water sources usually contain high concentrations of TDS, heavy metals, and organic and inorganic contaminants [1,2]. Moreover, the earth's water resources consist of 97% sea and 3% fresh water [3,4]. It is worth noting that seawater can only be used after being treated. The growing human demand for freshwater resources has exacerbated the global shortage. Water pollution and freshwater shortage are seriously threatening human survival. Therefore, the purification of water resources has become an urgent problem for human beings. At present, water treatment technology has been paid more and more attention to. Researchers have developed various water treatment technologies for seawater desalination and sewage purification, such as reverse osmosis, electrodialysis, distillation, adsorption, chemical precipitation, ion exchange, and other technologies [5–9]. However, these methods have some limitations in production and application, such as high energy consumption, high equipment consumption rate, easy-to-produce secondary pollution, and other problems.

CDI is a new electro-sorption technology. CDI has the advantages of environmental friendliness, low cost, low energy consumption, high efficiency, easy electrode regeneration, and no secondary pollution. More importantly, based on the reversible electrochemical process, the energy consumed by capacitor deionization can be partially recovered, and the energy consumption and economic cost of the system can be reduced.

Figure 1 shows the number of research papers on the application of CDI in water treatment from 2010 to 2022, as searched using the Web of Science. Since 2010, the experimental papers on CDI technology in water purification research have been overgrown, which strongly shows the rapid development of CDI technology. Therefore, in the face of the energy crisis and global warming, CDI technology will likely provide humans with the critical method of using fresh water.

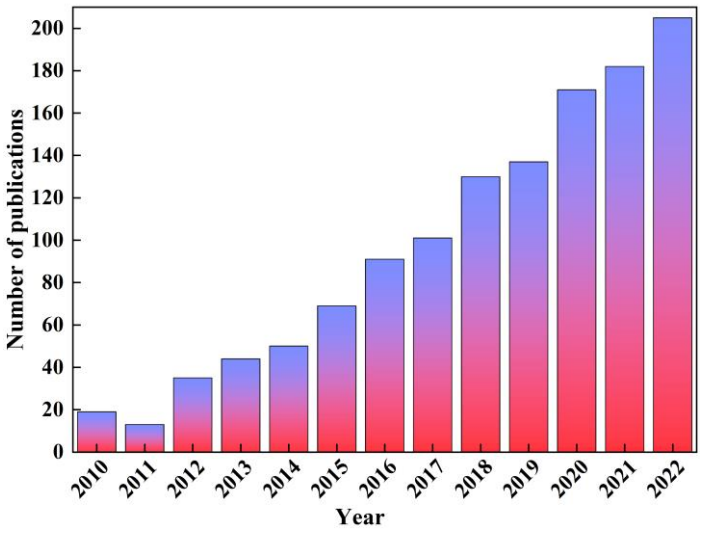

Figure 1. The number of research papers published on the application of CDI technology in water treatment from 2010 to 2022.

The principle of CDI technology is mainly based on the theory of electric double-layer adsorption, including adsorption and desorption processes [10]. In the adsorption process, an external voltage (<1.8 V) is applied to the two parallel plate electrodes, and an electrostatic field is formed between the two electrodes. When the ions in the solution flow through the electrode, they migrate to the electrode under the action of electric field force and concentration gradient and adsorb on the electrode material surface to form a double layer to achieve the purpose of ion removal [11]. When the adsorbed ion is nearly saturated, the voltage between the electrodes or the reverse voltage is removed. The ion is released into the solution to achieve electrode regeneration [12–14]. The CDI architecture diagram and removal mechanism are shown in Figure 2a. In recent years, people have begun to explore efficient electrode materials and innovate the battery structure to improve the removal rate and energy efficiency. With the rapid development of CDI technology, especially the continuous emergence of new CDI devices, representative advanced CDI systems such as membrane capacitor deionization (MCDI) technology, flow electrode capacitance deionization (FCDI) technology, and hybrid capacitor deionization technology (HCDI) have emerged [15]. The specific configurations are shown in Figure 2.

The conventional CDI system has a "common ion effect" [16]. When a voltage is applied to the electrode, ions with an opposite charge will be adsorbed on the electrode. At the same time, ions with the same charge as the electrode will enter the solution to be measured through the electrode's pores, lowering the ion removal efficiency [13,17]. In order to improve the charge efficiency and ion removal ability of carbon materials by reducing the influence of the common ion effect, Lee et al. [18] developed a new desalination technique for MCDI by placing ion exchange membranes on one or more electrodes. Compared with the conventional CDI, MCDI avoids the common ion effect, improves charge efficiency and energy efficiency,

and exhibits excellent desalination performance [19,20]. MCDI has become one of the most interesting new configurations in CDI technology.

However, MCDI technology still did not meet business needs, so FCDI technology came into being. For a long time, the optimization research of CDI focused on the fixed electrode and improved the electrode performance through the innovation of electrode materials. However, the adsorption capacity of fixed electrode materials in CDI technology is limited, and the system cannot operate continuously for a long time because of the desorption process during operation [21]. Therefore, the innovation of electrode materials has yet to effectively solve the limitation of electrode materials, which seriously restricts the application of CDI technology in treating high-concentration brine. Based on MCDI, Jeon et al. [22] developed Flow electrode capacitive deionization (FCDI) in 2013 by using porous material suspension as a mobile electrode. The appearance of FCDI technology breaks the limit of the fixed electrode, and the mobile electrode replaces the fixed electrode to adsorb ions in water. During the operation of the FCDI, the electrode suspension is continuously circulated and regenerated by desorption outside the electrode chamber. FCDI improves desalination performance and provides a system for long-term continuous operation [23–25].

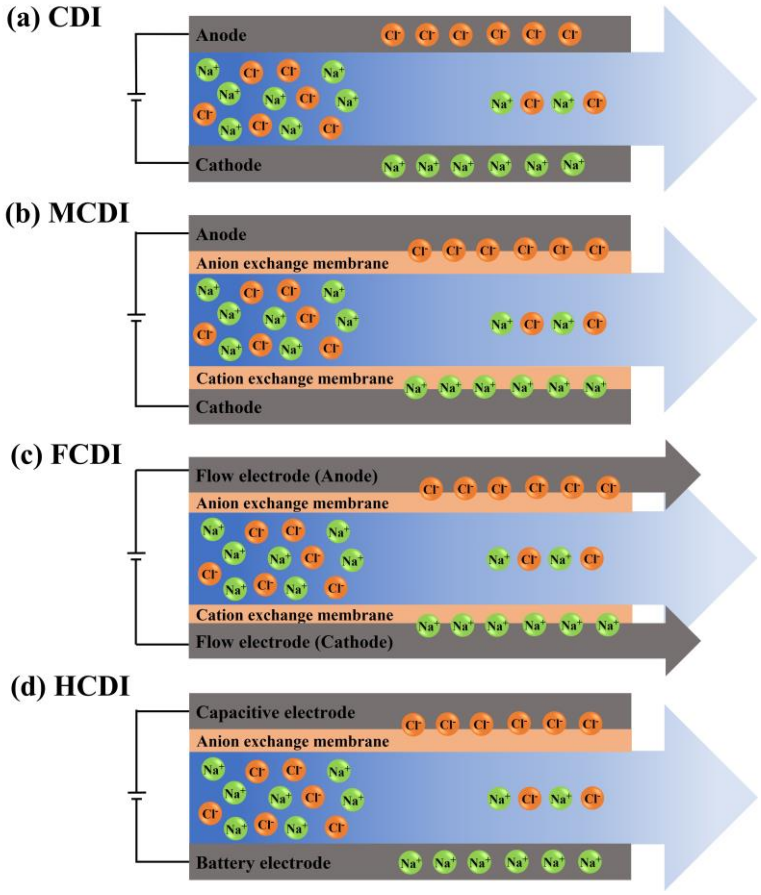

Figure 2. Architecture diagrams and removal mechanisms of (**a**) CDI, (**b**) MCDI, (**c**) FCDI, and (**d**) HCDI.

Another new system has emerged with the development and application of MCDI, FCDI, and other structures. Lee's team reported another new seawater desalination technology called Hybrid capacitive deionization (HCDI) [20,26]. HCDI system combines CDI and asymmetric capacitance system of battery material and is made of two different electrode materials, a carbon electrode and the battery material [27,28]. During desalination, sodium ions are retained in the redox electrode through a chemical reaction. Chlorine ions

are trapped in the double-layer electric layer formed by the surface of the porous carbon electrode. This design increases the efficiency of desalination.

CDI technology has unique advantages in water treatment. With the development and improvement of electrode materials and the continuous optimization of different types of CDI structures, the functional application areas of CDI technology have been widely expanded to include brackish water desalination, seawater desalination, heavy metal removal, water disinfection, organic contaminants removal, energy recovery, and other areas. Other review papers summarized CDI electrode materials and CDI battery structure. However, the existing literature still needs a comprehensive overview of the multi-functional application of CDI technology in water treatment. Therefore, this paper comprehensively summarizes the latest research advances in CDI technology in water treatment. It focuses on representative applications in various fields, providing a specific theoretical basis and the latest research results for relevant researchers. At the same time, it further promotes the application and development of CDI technology in various fields. It summarizes and examines the future development prospects and challenges of CDI technology in scientific research and practical applications.

2. Application Progress of CDI Technology in Water Treatment

2.1. Water Desalination

In 1960, Blair and Murphy first proposed the concept of desalination, called "electrochemical desalination of water" [29]. They conducted pioneering research on the application of CDI technology in seawater desalination. Porous-activated carbon is the earliest electrode material used in CDI technology, but it has some defects, such as poor conductivity, wettability, and low capacity. Therefore, researchers have improved the adsorption performance of the electrode through modification, activation, doping, and other methods. Ryoo et al. [30,31] doped the activated carbon electrode with titanium dioxide. The existence of titanium dioxide significantly increases the electro-adsorption of electrode materials on NaCl, which proves that the desalination performance of activated carbon materials can be improved by doping some substances. After a period of research on conventional activated carbon, researchers were not satisfied with its adsorption capacity. Therefore, other carbon materials are constantly being developed to improve the desalination performance of electrode materials. Manganese dioxide (MnO_2) is a TMO material with high theoretical specific capacitance (>1300 F/g), but the lower electrical conductivity limits the practical application of MnO_2 [32–36]. MnO_2 is commonly compounded with carbon to enhance the electro-adsorption performance and is used as an HCDI electrode. Biomass carbonization usually produces biochar under high temperatures and in an oxygen-free atmosphere, so it is considered a new substitute for traditional activated carbon. Adorna et al. [37] prepared coconut shell-derived activated biochar (AB) with a high specific surface area using waste coconut shells and combined AB with MnO_2 to prepare AB-MnO_2 nanocomposites. The material exhibited excellent CDI performance at 1.2 V, with a specific electro-sorption capacity of 68.4 mg/g for Na^+ removal. Other researchers have synthesized MnO_2 nanowires with different pore crystalline structures using hydrothermal methods and investigated the effect of crystal structure on desalination performance [38]. As shown in Figure 3a, the crystal structure has a significant influence on the desalination performance of HCDI. It can be adjusted according to the ion hydration radius to optimize desalination performance further.

Graphene has excellent conductivity and a hierarchical porous structure compared with other carbon materials. Its specific surface area reaches 2630 m^2/g, showing that graphene can have excellent desalination performance when used as a CDI electrode [39–43]. Zhang et al. [44] fabricated graphene electrodes with excellent desalination performance by assembling core-shell composites with incomplete graphene oxide shells into bulk membranes using the compression molding method and heat treating them under an NH_3 atmosphere (Figure 3b).

When graphene sheets are rolled into nanoscale cylindrical structures, single walls (SWCNTs) or multi-walled carbon nanotubes (CNTs) with high mechanical strength, good chemical

stability, high conductivity, and good capacitance retention will be formed [45–47]. Thus, CNT has considerable potential for electro-sorption, indicating that it is very suitable as a CDI electrode material. Meanwhile, CNT is considered an ideal candidate for CDI electrode materials. In the research by Hu et al., they prepared N-doped carbon nanotube materials containing cobalt and cobalt oxides (Co-CO_3O_4/N-CNT) by the pyrolysis method [48]. They uniformly encapsulated Co-CO_3O_4 nanoparticles in situ in the internal channels of conductive CNTs (Figure 3c). The electrode material has an excellent desalination ability of 66.91 mg/g NaCl. The interaction between cobalt and cobalt oxide is key to improving desalination capacity.

However, the electro-adsorption performance of a single carbon can no longer meet the application requirements of the rapidly developing CDI technology. In order to obtain electrode materials with better electro-sorption performance, researchers combined MOFs derived carbon, Prussian blue (PB), and blue analogues (PBAs) with other carbon materials, such as MOFs-derived carbon-graphene composites, PBA-CNTs, MOFs-derived carbon-carbon nanofiber composites, and MOFs-derived carbon–carbon nanotube composites [49–52]. Shi et al. [53] synthesized Fe-MOF and graphene at 800 °C and prepared a new type of MCDI electrode. The electrode material exhibits an excellent electro-adsorption capacity of 37.6 mg/g and high desalination stability in 1000 mg/L NaCl solution and 1.2 V voltage. The excellent performance of this material is important for two reasons. On the one hand, graphene provides a highly conductive pathway, and on the other hand, Fe-MOF provides a highly porous carbon structure.

Prussian blue (PB) and blue analogues (PBAs) belong to intercalation materials. Compared with other carbon materials, this material has the advantages of high theoretical specific capacity, safety, low toxicity, and easy synthesis [54,55]. However, this material has some disadvantages, such as poor electronic conductivity, particle aggregation, and poor cycling stability [56,57]. Therefore, the defects of PBA limit its practical application in CDI. Combining carbon materials with PBA has proven to be an effective way to improve the desalination performance of PBA [58,59]. Gong et al. [60] successfully prepared a stepwise hollow Prussian blue/CNTs composite (SHPB/CNTs) by combining PBA with CNTs (Figure 3d). When the material is used as a CDI electrode, it shows an ultra-high desalination capacity of 103.4 mg/g and excellent cycle stability.

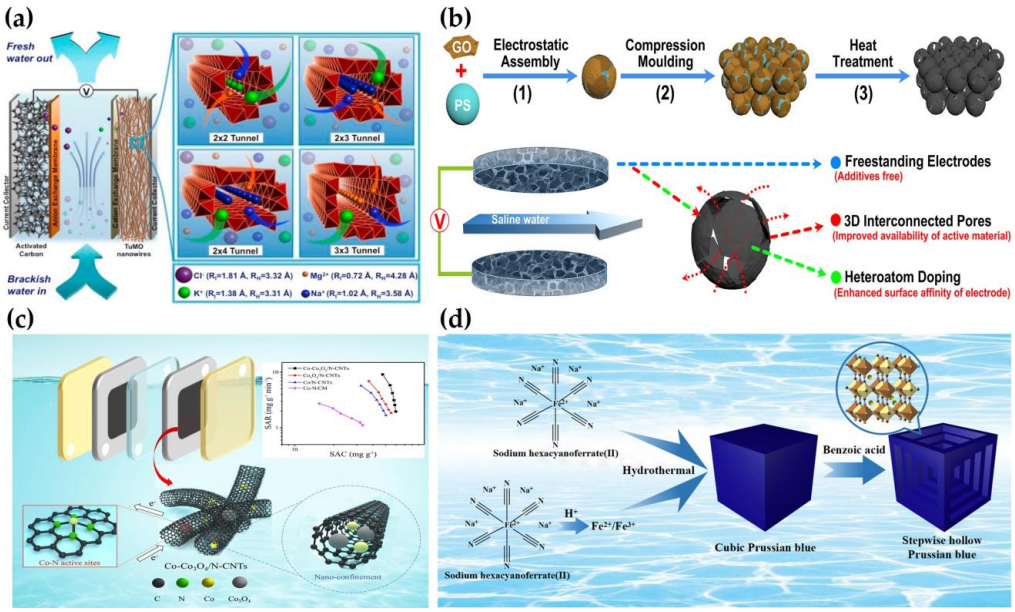

Figure 3. (**a**) Manganese oxide nanowires with different tunnel crystal structures as HCDI electrodes for ion removal [38]. Copyright 2018, Elsevier. (**b**) Schematic diagram of the construction process of graphene-based freestanding membrane electrodes and seawater desalination [44]. Copyright 2022, Elsevier. (**c**) Co-Co_3O_4 encapsulated in nitrogen-doped carbon nanotubes for capacitive desalination [48]. Copyright 2022, Elsevier. (**d**) Schematic illustration of the synthesis process for SHPB [60]. Copyright 2022, Elsevier.

In addition, in order to find high-performance materials that can remove salt ions through other mechanisms, Younes et al. [61] prepared cobalt ferric oxide (CFO) metal oxide nanoparticles (NPs) through the hydrothermal method and then prepared high-performance nanocomposites by vacuum filtration or the freeze-drying method. The electrode material exhibits an excellent electro-adsorption capacity of 64.5 mg/g and high desalination stability in 50 mg/L NaCl solution and 1.2 V voltage. The excellent performance of this material benefits from the porous and interconnected 3D structure of the 3DrGO, and it provides a larger surface area to form EDL capacitance. In addition, the added porous 3DrGO entangled with the spinel crystals ($CoFe_2O_4$) in the composite allowed for a quick ion diffusion across the interconnected open macroporous structures.

2.2. Water Softening

Hard water refers to water containing more soluble calcium-magnesium compounds. Although hard water will not directly endanger human health, it can bring many troubles to life and industry, such as reduced washing efficiency of soap and detergent and scaling of water appliances [11,62,63]. More seriously, the existence of hard water will increase the production cost of enterprises and reduce their profits. Water hardness (expressed in units of mg/L) is the total amount of divalent cations such as Mg^{2+} or Ca^{2+} in the solution [64]. The more calcium and magnesium salts dissolved in the water, the greater the water's hardness. At the same time, the greater the hardness of the water, the easier it is for minerals such as calcium and magnesium to precipitate and eventually cause blockages in equipment such as water pipes and boilers. As the cost of equipment maintenance increases gradually, the hardness of water has become one of the criteria for judging production costs. Therefore, the softening of hard water has important practical significance. Various methods have been developed to address the problem of water hardenings, such as chemical precipitation, ion exchange, adsorption, electrodialysis, electrolysis, electrochemistry, and nanofiltration [65–72]. CDI is a chemical-free and cost-effective technology that has been increasingly applied to water softening. Table 1 summarizes some research results of hard water treatment with CDI in recent years.

Table 1. Removal efficiency of hardness ions by CDI.

Hardness Ion/Concentration	Applied Voltage (V)	Flow Rate (mL/min)	Removal Efficiency	CDI Type/Electrode	Ref.
100 mg/L $CaCl_2$	1.4	25	42.8 mg/g	HCDI with copper hexacyanoferrate as cathode electrode	[73]
10 mM $CaCl_2$	1.2	10	248 µmol/g	CDI with porous mordenite modified activated carbon electrode	[74]
10 mM $CaCl_2$	1.2	2	14.2 mg/g	Ca-alginate coated on carbon electrode	[75]
0.95 g/L $CaCl_2$	1.4	25	21.0 mg/g	CDI of KOH carbonized and activated carbon electrode	[76]
150 mg/L $CaCl_2$ 75 mg/L $MgSO_4$	1.2	20	534.6 µmol/g 980.4 µmol/g	PHCI with manganese spinel ferrite as cathode electrode	[77]
350 mg/L $CaCO_3$	1.5	16	80%	CDI with activated carbon cloth electrode	[78]
35 mg/L $CaCO_3$	2	10	3.5 mg/g	CDI with purified reduced graphene oxide electrodes	[79]

Preparing advanced carbon materials such as carbon aerogels, graphene, and CNTs requires expensive precursors and complex procedures. Low costs and renewability have led to biomass materials attracting the attention of researchers. Deng et al. [76] synthesized laminated activated carbon with an ultra-high specific surface area (1943.2 m^2/g) and large oxygen-containing functional groups by KOH charring and activating chestnut inner shells at high temperatures (Figure 4a). A mixed solution containing NaCl and $CaCl_2$ is injected into the CDI device. The carbonized and activated chestnut shell was used as a CDI electrode. Under the condition of the 30 mL/min volume flow rate, 1.4 V voltage was

applied to the CDI device for 20 min. It was found that the adsorption capacities of $CaCl_2$ and NaCl were 21.0 mg/g and 17.7 mg/g, respectively.

The low selectivity and electro-adsorption properties of carbon-based electrode materials severely limit the application of CDI technology in water softening [13,80]. Pseudocapacitive electrode materials selectively interact with specific ions through Faradaic redox reactions or ion (de)intercalation, providing high selectivity for the electro-sorption of Ca^{2+} in water [81,82]. Xu et al. [73] used the co-precipitation method to prepare copper hexacyanoferrate (CuHCF) as a pseudo-capacitive electrode. CuHCF is used as a cathode and activated carbon as an anode. The HCDI cells without ion-exchange membranes were used to study the Ca^{2+} adsorption performance of CuHCF, as shown in Figure 4b. A high electrical adsorption capacity of 42.8 mg/g was obtained, which exceeded the reported carbon-based electrode. At the same time, the adsorption selectivity of CuHCF to Ca^{2+} is 3.05, higher than Na^+ and Mg^{2+}.

Yoon et al. [75] successfully prepared a Ca-alginate coated electrode and used it for hard water softening. The results showed that the removal rate of Ca^{2+} by Ca-alginate coated-CDI was 44% higher than that by conventional CDI. Further studies revealed that the calcium alginate coating showed comparable performance to conventional MCDI when used as an MCDI electrode.

Pseudocapacitor-type HCDI, which is carried out through the intrinsic pseudocapacitive effect, is now gradually being used in deionization. Xu et al. [77] prepared a manganese spinel ferrite (MFO) selective adsorption electrode by one-step solvothermal synthesis (Figure 4c). MFO electrode has a highly selective ability to adsorb hardness ions 534.6 μmol/g ($CaCl_2$) and 980.4 μmol/g ($MgCl_2$), mainly because of its unique crystal structure and pseudocapacitive property. It was also found that the electro-affinity of Mg^{2+} for Na^+ was much higher than that of Ca^{2+} in the mixed solution, achieving a super high hardness selectivity factor of 34.76. The electrochemical response of unary and multiple electrolytes revealed a unique pseudocapacitive affinity based on the cation (de)intercalation-redox mechanism, which is the main reason for the selective removal of hardness ions by electrode materials. This work provides an in-depth study of the intrinsic mechanism of redox reactions and efficient ion selectivity, which provides a new reference for developing new Faraday materials with high selectivity for water softening.

The coexistence of Tetracycline (TC) and hardness ions in natural water are common [83]. More importantly, TC and hardness ions quickly form TC-metal compound pollutants in the environment [84]. Therefore, the simultaneous removal of TC and hardness ions has important practical significance for environmental protection. Considering this, Sun et al. [85] assembled a symmetrical CDI device using kelp-derived hierarchical porous carbon (KHPC) to remove TC and hardness ions from water simultaneously (Figure 4d). The excellent deionization performance of KHPC electrodes is mainly caused by the improved potential-induced electrical adsorption performance of KHPC, high specific surface area, and suitable hierarchical pore structure. Although CDI water treatment technology has been applied to water softening, more research still needs to be done in this area. Furthermore, most of the research is still in the laboratory stage. The scaling formed during the operation of the CDI device may deteriorate the water quality, which still needs further study. Therefore, it is necessary to develop electrode materials with low cost, no pollution, and high selectivity and to conduct in-depth research on the sustainable operation of CDI in practical applications.

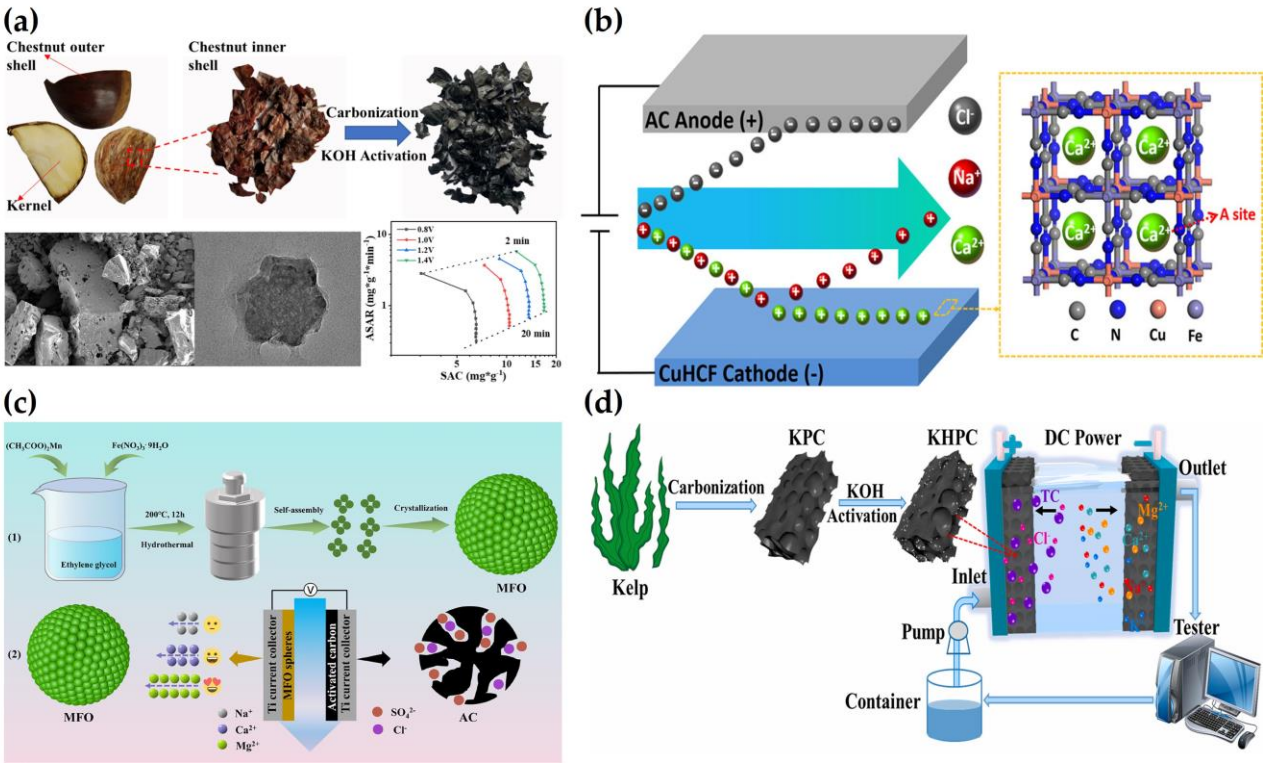

Figure 4. (**a**) Fabrication of CSx from chestnut inner shell and Structure diagram of CS500 [76]. Copyright 2022, Elsevier. (**b**) Application of CuHCF electrode material in hard water treatment [73]. Copyright 2020, American Chemical Society. (**c**) (1) Schematic illustration of the preparation of MFO nanospheres; (2) Schematic illustration of selective ion electro-sorption of the MFO electrode in the PHCDI system [77]. Copyright 2021, American Chemical Society. (**d**) Simultaneous removal of TC and hardness ions from water using the KHPC electrodes with a symmetrical CDI device [85]. Copyright 2021, Elsevier.

2.3. Removal of Heavy Metals

With rapid economic development, the industry has been expanding in recent years. Toxic and harmful heavy metals are released into the environment with industrial wastewater. Heavy metal pollution has seriously threatened the natural environment and human health. In the past decades, various techniques have been proposed to remove heavy metals from water, including chemical precipitation, ion exchange, electrochemistry, adsorption, membrane separation, coagulation, and other methods [86–89]. However, these techniques have disadvantages, such as high energy consumption and a tendency to cause secondary pollution [90–95]. CDI technology does not use chemical reagents and does not produce waste during water treatment. The advantages of CDI technology has attracted many researchers to explore its potential application in removing heavy metals in wastewater. Table 2 summarizes some research results of CDI application in heavy metal removal in recent years.

Electrostatic interaction is the primary mechanism of CDI technology to remove heavy metals, but non-electrostatic adsorption (physical or chemical adsorption) may also play a role [96]. Tang et al. [97] used the activated carbon cloth electrode as the CDI electrode to adsorb Zn^{2+} under the condition of applying 1.2 V voltage to the CDI device (Figure 5a). It was found that electrostatic interactions caused 87% of the total adsorbed Zn^{2+}, and the remaining 13% was caused by non-electrostatic adsorption. The limited double-layer (EDL) capacitance and desalination capacity of traditional carbon-based electrodes seriously affects their practical applications. Li et al. [98] prepared a self-supporting porous carbon nanofiber membrane (ZnO@N-PCNM) with high nitrogen doping and nano-Zn content by electrospin-

ning technology using ZIF-8 as a template (Figure 5b). The prepared ZnO@N-PCNM-10 was used as a CDI electrode. At an applied voltage of 1.8 V, the material successfully removed Pb^{2+} (99.42%), Cu^{2+} (68.46%), and Cd^{2+} (70.36%) ions. The results indicate that the material effectively removes heavy metal ions. High N doping and Zn loading make the material obtain higher specific capacitance and better electro-sorption performance.

Generally, solutions contain many kinds of ions, so the process of removing target ions will inevitably be interfered with by other ions. Therefore, the selective removal of target ions by CDI systems remains a significant challenge in practical applications where multi-ion hybrid solutions are treated. Zhang et al. [99] used the FCDI device to remove and separate Cu^{2+} from a salty solution with the assistance of (electro)deposition. During migration, Cu^{2+} is deposited on the carbon particles, whereas most Na^{+} is retained in the solution. It is noteworthy that Na^{+} desorption is strong; therefore, the separation of the two ions is more pronounced in the short-circuited closed cycle (SCC) mode. At an initial pH of 8.0, only Cu^{2+} was present on the cathode. In contrast, at an initial pH of 2.5, there was more Cu than Cu^{2+}, indicating that Cu^{2+} ions were effectively electrodeposited onto the cathode and formed Cu. The presence of Cu^{2+} in the carbon particles could be caused by weak electro- or Physico-chemical adsorption. It was found that the difference in ion distribution in the flow electrode led to the slower decay of Cu^{2+} removal performance than that of Na^{+}. This shows that FCDI technology has advantages in removing Cu^{2+}.

Many pseudocapacitive electrode materials based on Faraday redox reactions are used to efficiently and selectively remove various heavy metal ions. Xu et al. [100] prepared polypyrrole-coated dual-metal perovskite-type oxide as a pseudocapacitive cathode (Figure 5c). The maximum Cd removal capacity of 144.6 mg/g was achieved by asymmetry pseudocapacitive deionization. The CDI process maintained a retention rate of 93.4% after 150 h of operation. Cd is easily affected by pH value. With the decrease in pH value, the adsorption capacity of Cd decreased. The main reason for this phenomenon is that Cd competes with H^{+} for active sites. When the voltage value is changed, the removal capacity increases with the increase of voltage, but the energy consumption also increases. Compared with single metal oxide electrode materials, bimetallic oxides have better electrochemical performance and cycle stability and are easier to be popularized in industry. Therefore, bimetallic oxide is a potential CDI electrode material.

From the perspective of environmental chemistry, V^{5+} is the most toxic vanadium ion in various valence states, so V^{5+} is the main target for removing vanadium ions. Bao et al. [101] combined ion exchange resin with activated carbon to prepare a composite electrode, which was used as the electrode of the CDI battery to adsorb V^{5+}. Finally, a 106.89 mg/g high electric adsorption capacity was obtained. In another work, the team prepared a composite material that combines porous carbon derived from ZIF-8 with activated carbon [102]. The electrode is used as the electrode of CDI. A high adsorption value of 77.27 mg/g was obtained at a voltage of 1.2 V.

Generally, the ion radius, ion charge, and an ion's physicochemical affinity for the electrode will affect the selectivity of the electrode material to the ion [103,104]. It has been shown that ions with a small hydration radius, higher valence ions, and homovalent ions are more readily adsorbed by electrode materials [105–107]. For example, Huang et al. [105] carried out batch electro-sorption experiments using an activated carbon cloth for Cr^{3+}, Cd^{2+}, and Pb^{2+} (Figure 5d). It was found that the order of removal efficiency of these three cations was $Cr^{3+} > Pb^{2+} > Cd^{2+}$. The order of the hydration radius of these three cations was $Cr^{3+} > Pb^{2+} > Cd^{2+}$. Cr^{3+} has the highest hydration radius and valence state compared to the other two ions. Therefore, the removal efficiency of Cr^{3+} is better than the other two ions. Generally, CDI technology is very effective in removing ions when the ion concentration is low.

Coincidentally, CDI technology matches most heavy metal-polluted water sources and can be combined with other water treatment technologies to enhance the removal effect. Therefore, CDI technology has broad prospects in heavy metal removal.

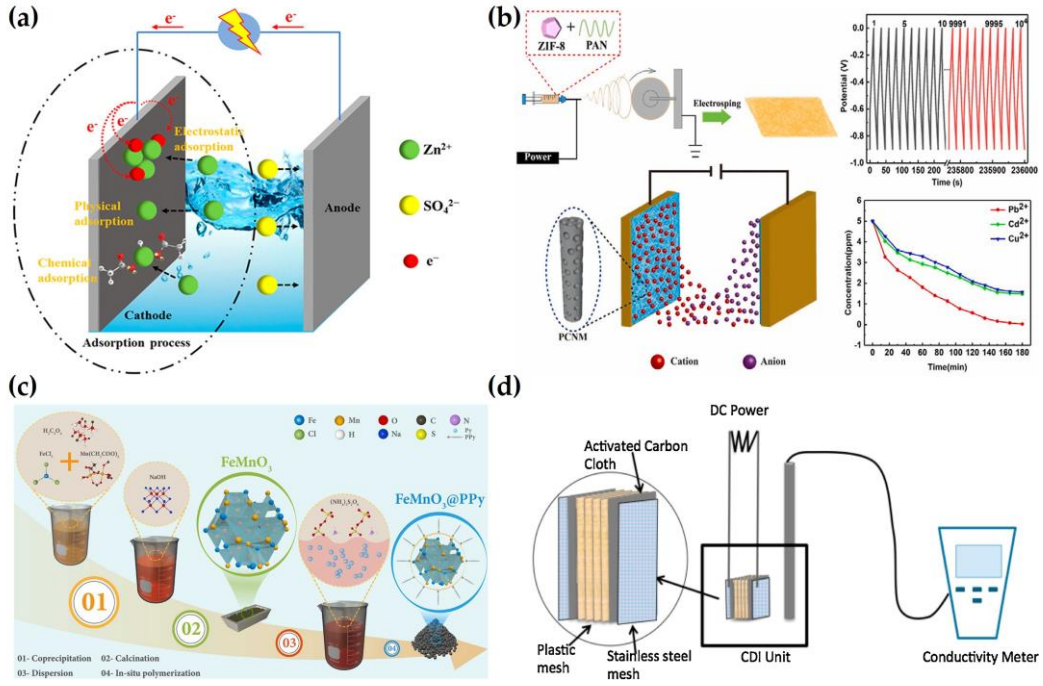

Figure 5. (**a**) The process of electro-assisted adsorption of Zn^{2+} from aqueous solution using activated carbon cloth as electrode in batch-flow mode [97]. Copyright 2019, American Chemical Society. (**b**) Controlled synthesis of ZnO-modified N-doped porous carbon nanofiber membrane for removal of heavy metal ions by CDI [98]. Copyright 2022, Elsevier. (**c**) Schematic illustration for preparation process of $FeMnO_3$ and FeMnO3@PPy [100]. Copyright 2022, Elsevier. (**d**) The schematic of CDI assembly for metal removal [105]. Copyright 2015, Elsevier.

Table 2. Removal efficiency of heavy metal ions by CDI.

Pollutant	Concentration (mg/L)	Electrodes	Flow Rate (mL/min)	Applied Voltage (V)	Removal Efficiency	Ref.
Cu^{2+}	96	Activated carbon	10	-	94.00%	[99]
Pb^{2+}	50	self-supporting porous carbon nanofiber membrane	10	1.8	99.42%	[98]
Cu^{2+}					68.46%	
Cd^{2+}					70.36%	
Zn^{2+}	40	Nitrogen-doped porous carbon	20	1.0	33.10 mg/g	[108]
As	0.02–0.1	Activated carbon	3000	1–1.5	>80%	[109]
Cd^{2+}	20	Perovskite oxide	20	1.2	144.60 mg/g	[100]
Pb^{2+}	100	Wood-converted Carbon	5	1.2	19.52 mg/g	[107]
Cr^{2+}	50				20.00 mg/g	
V^{5+}	1500	Composite materials based on ion-exchange resin and activated carbon	30	-	106.89 mg/g	[101]
V^{5+}	750	Composite materials based on porous carbon derived from ZIF-8 and activated carbon	30	1.2	77.27 mg/g	[102]
U^{6+}	100	highly porous phosphate-functionalized graphene	50	1.2	545.70 mg/g	[110]
Ni^{2+}	50	Multi-walled CNTs	50	1.5	145.73 mg/g	[111]

2.4. Removal of Phosphate and Nitrate

Products such as fertilizers and pesticides used by humans are rich in N and P. Excessive N and P are released into natural water bodies through industrial effluents, agricultural

runoff, and municipal wastewater systems [112] (Figure 6). Excessive enrichment of phosphate and nitrate in water will lead to eutrophication [113–118]. Algal blooms caused by water eutrophication seriously endanger the water supply system and ecosystem [119,120]. In addition, phosphate is an indispensable resource for industrial and agricultural production, but phosphate rock is a nonrenewable resource, so the world is facing a shortage of phosphorus resources [121,122]. Therefore, removing and recovering phosphate and nitrate from water is urgent. In recent years, some researchers have applied CDI technology to remove phosphate and nitrate in water, and some research progress has been made. Table 3 summarizes some research results of CDI application in phosphate and nitrate removal in recent years.

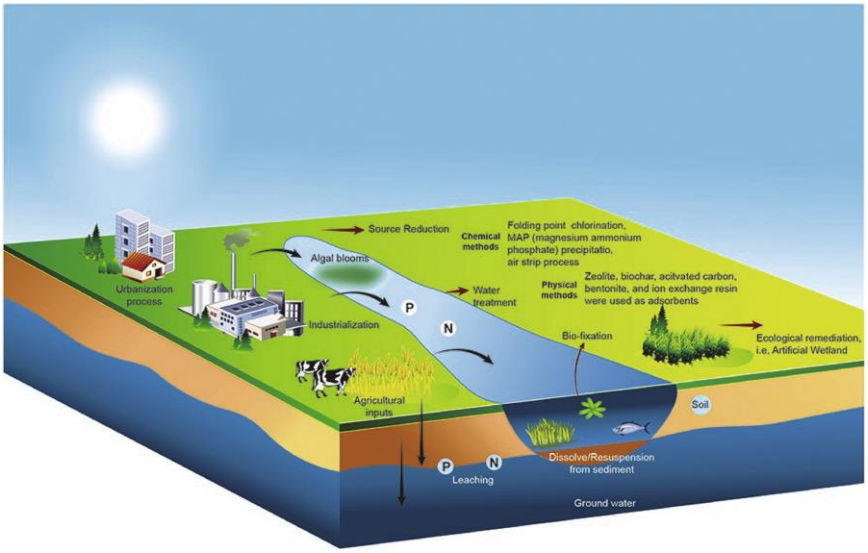

Figure 6. Source and control of nutrients (N and P) in the water environment [112]. Copyright 2020, Elsevier.

For example, Zhang et al. [123] developed a terephthalic acid intercalated carbon nanotube composite material (ZnZr-COOH/CNT) as an anode for CDI to evaluate its phosphate removal capacity (Figure 7a). For the wastewater containing 6 mg/L phosphate, the maximum adsorption capacity of 13.65 mg/g was obtained by applying 1.2 V voltage to the CDI device. The network structure of CNTs provides more space for phosphate adsorption and enhances selective phosphate adsorption. At the same time, there are two main reasons why the electrode material has a high affinity for phosphate. First, Zn and Zr in the electrode are strongly complexed with phosphate. Secondly, a hydrogen bond was formed between the hydroxyl group of phosphate and the carboxyl group of terephthalic acid.

Compared with CDI, FCDI uses flow electrodes, significantly improving desalination performance. The flow electrode provides a long-term continuous operation system for FCDI. Zhang et al. [124] used FCDI to remove and recover phosphorus from low-concentration wastewater in the SCC mode (Figure 7b). In the six cycles, the phosphorus removal rate and adsorption capacity remained stable at about 97% and 0.97 mg/g. This shows that the FCDI system has good cycle removal performance for phosphorus. Interestingly, when the pH value is 2.15–7.20, the phosphate is mainly $H_2PO_4^-$, and when the pH value is 5, the removal effect is the best. At pH 7.20–12.35, HPO_4^{2-} is the primary phosphate. The diffusion coefficient of $H_2PO_4^-$ is not only larger than HPO_4^{2-}, but also the hydration radius of $H_2PO_4^-$ is smaller than HPO_4^{2-}. This shows that $H_2PO_4^-$ can be more easily transferred from the spacing channel to the flow electrode. The above results show that the removal efficiency of phosphorus by FCDI is highly influenced by the initial pH value of feed water, and it is easier to remove phosphorus in a weak acid environment. They also found that the FCDI system could enrich phosphorus in solution

16 times after 11 h of operation. This shows that the mobile electrode can concentrate and enrich low-concentration phosphorus-containing wastewater, significantly promoting the recovery efficiency of phosphate.

Bian et al. [125] also studied the removal of phosphorus by the FCDI system. They explored phosphate ions' existing form and migration behavior in the FCDI system (Figure 7c). When the pH value of the solution is 5, the phosphorus removal effect is the best, and the phosphorus removal rate is up to 38.3 mg/min. Compared with pH 9, the phosphorus removal efficiency at pH 5 increased by 84~104%. The main reason for the increase in phosphorus removal efficiency is that when the pH value is 5, P changes from HPO_4^{2-} to H_2PO^{4-}. The primary phosphate types in the feed solution and flow electrode will change with the change of pH value, and the reaction formula is shown in Equations (1)–(3) [126]. They also studied the energy consumption of the FCDI system for phosphorus removal. They also studied the energy consumption of the FCDI system for phosphorus removal. When the pH is reduced from 9 to 5, the energy consumption decreases by 42.9–56.1%. Based on the research results, they found that the removal rate and recovery rate of phosphate ions mainly depend on the pH value of the feed solution or electrolyte. The pH value of the solution is critical to the removal and recovery of phosphorus by FCDI. Therefore, proper pH values can improve phosphorus removal and recovery rates.

$$H_3PO_4 \rightleftharpoons H^+ + H_2PO_4^- \; pK_{a1} = 2.148 \quad (1)$$

$$H_2PO_4^- \rightleftharpoons H^+ + HPO_4^{2-} \; pK_{a2} = 7.198 \quad (2)$$

$$HPO_4^{2-} \rightleftharpoons H^+ + PO_4^{3-} \; pK_{a3} = 12.375 \quad (3)$$

Vivianite is the general name of a group of phosphate minerals with similar structures. Vivianite is considered slow available fertilizer and a possible reagent for manufacturing lithium iron phosphate [127,128]. Thus, some researchers have attempted to recover the phosphate from the wastewater and prepare it as a vivianite with some economic value. For example, Zhang et al. [129] pioneered the construction of a coupling system of FCDI and fluidized bed crystallization to recover phosphorus from wastewater. First, the flow electrode concentrates the phosphate, and then the concentrated phosphate forms the vivianite in the fluidized bed crystallization reactor. The experimental results show that the FCDI system can concentrate 63% phosphate into the electrode chamber. The fluidized bed crystallization system can convert approximately 80% phosphate to vivianite. This study provides a new avenue for effectively recovering phosphorus from wastewater as a value-added product.

Phosphate and nitrate need to be removed from the water, and the recovered phosphate can form products for secondary utilization. However, nitrate needs to be degraded without pollution after removal to avoid environmental pollution again. Recently, some researchers have applied CDI or MCDI technology to remove nitrate from wastewater. Fateminia et al. [130] prepared a ternary composite electrode for CDI to remove nitrate. An electro-adsorption capacity of 6.01 mg/g was obtained with a nitrate removal efficiency of 60.01% under 2.0 V applied to the CDI device. This technology mainly uses the synergistic effect of four materials (activated carbon/PVDF/polyyline/ZrO_2) to enhance the nitrate adsorption capacity in the feed solution. With the advantages of low cost and low energy consumption, MCDI is a promising alternative technology for water treatment. In another study, Cetinkaya [131] applied MCDI to remove nitrate from water. The method achieved a nitrate removal rate of 83.07% with an applied voltage of 0.8 V. The study also showed that the main factor affecting the environment is the materials used in the manufacturing process. Therefore, environmentally friendly materials should be used in the nitrate removal process to avoid contamination of the environment by the materials used.

Although CDI technology has been applied for the removal of nitrate, however, the treatment process will produce nitrate-concentrated waste liquid [132]. Therefore, the concentrated waste liquid needs to be further treated. Some researchers have studied the simultaneous removal and degradation of nitrate. This method makes CDI technology more

economical and applicable in nitrate removal. Hu et al. [133] doped Pd nanoparticles into Pd/NiAl-layered metal oxide by in-situ hydrothermal method and successfully prepared Pd-containing film electrodes. They used the electrode in the electro-adsorption/reduction solution containing nitrate (Figure 7d). The study successfully converted NO_3^- in solution to non-polluting N_2, reducing secondary waste generation.

A similar study was done by Rogers et al. [134]. They used catalytic capacitive deionization (CCDI) technology for the removal and degradation of nitrate from wastewater and used commonly used porous carbon as the cathode, as shown in Figure 7e. The technology has the common advantages of catalytic water treatment and CDI technology. At 1.5 V, 91% NO_3^- can be reduced to N_2. They also analyzed the overall energy efficiency of the treatment process. CCDI may be considered an economical and applicable wastewater treatment method further to promote the development of the next-generation CDI system.

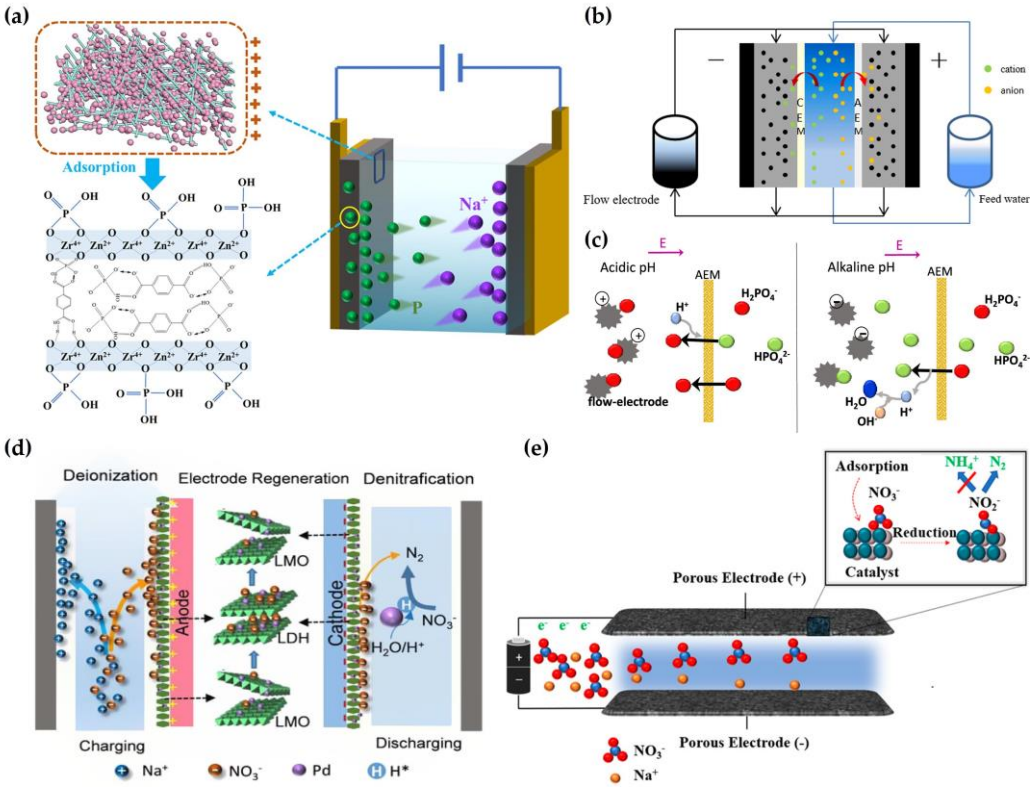

Figure 7. (**a**) Mechanism diagram of phosphate adsorption by ZnZr-COOH/CNT electrode [120]. Copyright 2022, Elsevier. (**b**) Schematic diagram of the FCDI cell operated in the SCC mode for phosphorus removal and recovery [124]. Copyright 2020, Elsevier. (**c**) The existing form and migration behavior of phosphate ions in FCDI at different pH values [125]. Copyright 2020, American Chemical Society. (**d**) The process of electro-sorption/reduction of NO_3^- by CDI with Pd/NiAl LMO electrode [133]. Copyright 2018, Elsevier. (**e**) The process of electro-sorption/reduction of NO_3^- by CDI with In-Pd NP electrode [134]. Copyright 2021, American Chemical Society.

Table 3. Removal efficiency of phosphate and nitrate by CDI.

Pollutant	Concentration	CDI type	Applied Voltage (V)	Removal Efficiency	pH	Energy Consumption	Ref.
P	6 mg/L	CDI	1.2	13.65 mg/g	7	0.0075 kWh/g-P	[123]
P	2.5 mM	CDI	1.2	35 mg/g	6.5	-	[135]
P	62 mg/L	FCDI	-	164 mg/L per cycle	8.3	27.8 kWh/kg-P	[136]
P	8 mg/L	FCDI	-	61.9%	-	21.8 kWh/kg-P	[137]
P	100 mg/L	FCDI	1.2	97%	5	-	[124]
P	500 mg/L	FCDI	1.2	38.3 mg/min	5	0.59 kWh/kg-P and 0.043 kWh/m^3-water	[125]
N	0.5 mM	CDI	1.2	2.4×10^{-3} mmol/m^2	5.6	248.8 kJ/mol-NO^{3-}	[138]
N	50 mg/L	CCDI	1.5	91%	-	13.2 kWh·m^{-3} $order^{-1}$	[134]
N	42 mg/L	CCDI	1.0	-	6.5	-	[133]
N	200 mg/L	MCDI	1.8	53.3%	5.32	-	[139]
N	70 mg/L	CDI	2.0	60.01%	6	-	[130]
N	250 mg/L	MCDI	0.8	83.07%	-	-	[131]

2.5. Removal of Organic Contaminants

The coexistence of organic contaminants in wastewater will cause electrode pollution and deterioration, significantly inhibiting CDI's practical application. The organic contaminants in the solution will form scaling on the electrode and block the pores of the electrode material, seriously reducing the CDI operation efficiency and the electro-adsorption performance of the electrode material [140]. In order to ensure the continuous operation of the CDI system in practical application, effective methods are needed to remove organic contaminants from wastewater.

Some researchers combined CDI technology with other technologies to treat wastewater containing multiple pollutants. For example, Chen et al. [141] combined CDI and electro-oxidation (CDI-EO) to develop a hybrid system for the simultaneous removal of heavy metal ions and organics (Figure 8). At the same time, they have developed two electrodes that selectively remove heavy metals and organic contaminants. The anode of the CDI-EO cell is carbon-coated graphite paper for the removal of heavy metals, and the cathode is a RuO_2-IrO_2-protected titanium plate for the degradation of organic contaminants. CDI-EO battery is used to treat wastewater containing Cu^{2+} and Acid Orange 7. At the current density of 5 mA/cm^2, 80% Cu^{2+} and 89% Acid Orange 7 were removed, respectively. Interestingly, the degradation process of Acid Orange 7 and the removal process of Cu^{2+} will not affect and interfere with each other. The new technology should test the electro-adsorption performance and investigate its energy consumption because the technology with high energy consumption has no practical significance. The study achieved low energy consumption of 4.66 kWh/m^3-wastewater. This proves that the CDI-EO system not only removes metal ions and organic contaminants but also has the advantage of low energy consumption, making it economically suitable for wastewater treatment.

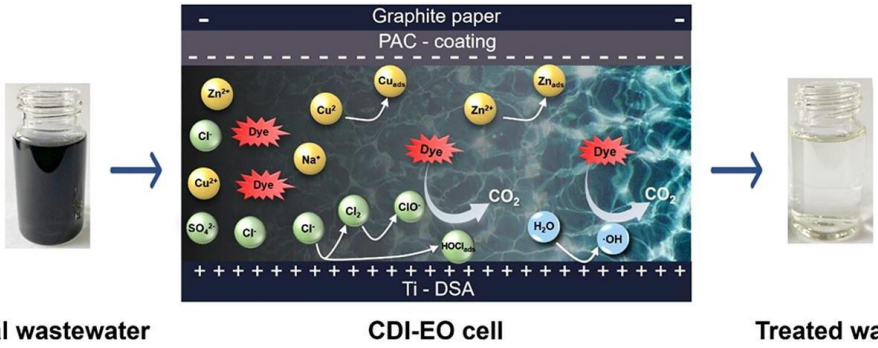

Figure 8. Removal process of Cu^{2+} and Acid Orange 7 by CDI-EO system [141]. Copyright 2022, Elsevier.

In another study, Liang et al. [142] combined ultrafiltration with CDI. They invented a novel integrated ultrafiltration-capacitive-deionization (UCDI) process that can simultaneously remove organic contaminants and inorganic salts from wastewater. Firstly, the system uses an ultrafiltration component or electro-catalytic oxidation to remove organic contaminants. Secondly, the system uses CDI electrodes to adsorb and remove inorganic salts. The UCDI process thus achieves the simultaneous removal of organic contaminants and inorganic salts from wastewater by combining ultrafiltration and CDI. It was found that if organic contaminants, Ca^{2+} and Mg^{2+}, are present in the solution simultaneously, the organic contaminants may have complexation and promote the removal of Ca^{2+} and Mg^{2+}.

The electrochemical advanced oxidation process (EAOP) is a cleaning technology for removing organic contaminants. Some researchers have combined EAOP with CDI to simultaneously remove organic contaminants and inorganic salts from wastewater [143]. The electrode of the CDI battery is an activated carbon plate. To remove phenol and sodium chloride, a 2.0 V voltage battery with oxygen is applied. Under the optimal conditions, the removal rates of phenol, TOC, and salinity are 90%, 60%, and 20%, respectively. Organic contaminants in the solution can be removed by degradation through direct or indirect oxidation [144]. As expected, the phenol removal pathways in this study included both direct and indirect oxidation.

At present, there are still few studies based on CDI technology for the treatment of organic contaminants. The above research shows that CDI technology needs to combine with other technologies to form a new system to have the ability to remove organic contaminants. Even though the integrated approach based on CDI solves the problem of synergistic removal of organic contaminants and other ions in wastewater, the facilities and operation are complex in the later stage, and the maintenance cost is high.

2.6. Water Disinfection

The disinfection process is usually the final stage of water treatment. Water disinfection is an important guarantee to ensure that the water used by human beings is free from harmful bacteria and viruses. The main methods of water disinfection are chemical and physical [145]. The chemical method uses disinfectants for disinfection, such as chlorine, hydrochloric acid, ammonia, alum, ozone, and other disinfectants [146,147]. Although these disinfectants remove harmful pathogens, they can produce toxic by-products (DBP) during disinfection [146,148–150]. Physical methods mainly include UV radiation disinfection, but this method requires high clarity of the water, poor disinfection, and high cost and energy consumption [146]. Therefore, finding an effective, low-cost, and clean disinfection technology is essential.

In recent years, the disinfection performance of CDI systems has become increasingly prominent. Many studies have shown that CDI is an alternative disinfection technology with great potential. Usually, bacteria survive in an environment with a specific pH range, which gives them a negative electrostatic charge [151]. Therefore, bacteria are considered negative ions in an aqueous solution. Then, the bacteria move towards the anode electrode under the action of an external electric field. During the disinfection process, bacteria will be adsorbed to the anode surface by the electric field force. The bacteria will be killed when contacting the bactericidal materials on the anode surface [152]. In conclusion, CDI can be used to remove bacteria from aqueous solutions. Wang et al. [153] prepared Alkoxysilane octadecyl dimethyl-ammonium chloride (ODDMAC) functionalized graphene oxide (GO) hybrid electrodes for capacitive deionization. The technology mainly uses ODDMAC attached to the surface of GO material to kill bacteria. Based on the above killing principle, under the condition of applying 1.2 V voltage to the CDI battery, the water with a pollutant concentration of 104 CFU/mL was sterilized. The results showed that the disinfection efficiency of the hybrid electrode to *E. coli*, *P. aeruginosa*, and *S. aureus* was 99.65%, 98.64%, and 97.69%, respectively. Cao et al. [154] used chitosan to modify 3-D channel structured ordered mesoporous carbons (Ia3d). The altered activated carbon was used as a CDI electrode to disinfect seawater. The results showed that only 0.01% of *E. coli* survived

after 30 min when the brine was disinfected at 1.2 V. Chitosan has a bactericidal effect. At the same time, anionic Ia3d was modified by chitosan, and the modified Ia3d became cationic. Therefore, the electrode materials synthesized in this study have conductivity and bactericidal effects. The above research proves that CDI technology has excellent bactericidal performance. The ability of CDI technology to disinfect is mainly attributed to the bactericidal effect of the composite materials. When the composite material is used as the anode of the CDI battery, the bacteria with negative static charge pass through the CDI electrode will be absorbed and killed by the anode electrode.

3. Energy Recovery and Energy Consumption Assessment

Energy recovery is a significant advantage of CDI in the field of water treatment because energy recovery can reduce energy consumption. Energy consumption is not only an essential indicator for evaluating the performance of CDI, but it is also a key indicator of superiority over other technologies [14,155]. More importantly, the low energy consumption makes CDI technology occupy a larger market in water treatment. The essence of CDI is to realize the migration of solution ions through the action of the electric field force. The migration of ions on the electrode surface and between solutions will promote the formation and disappearance of the double electric layer. This is very similar to the principle of an electric double-layer capacitor, which shows that CDI also has the characteristics of energy storage and recovery. If the CDI battery is regarded as an energy storage element, the adsorption and desorption stages can be regarded as the charging and discharging stages of the energy storage element.

More and more people have recently built energy recovery devices for CDI technology and use the devices to recover energy from water treatment processes. Chen et al. [156] successfully constructed an energy recovery system based on a four-switch buck-boost converter, which can transfer electric energy from the CDI module to the supercapacitor (Figure 9). The study's results showed that the system achieved an excellent energy recovery rate of 49.6%. It has been found that higher NaCl concentrations and shorter distances between electrodes are more conducive to energy recovery. This is because electronic resistances and ionic resistances produce low energy dissipation. In other studies, by changing the circuit connection sequence of the CDI system, not only can 70% of the energy be effectively recovered, the recovered energy can also be used in another ion adsorption process [157].

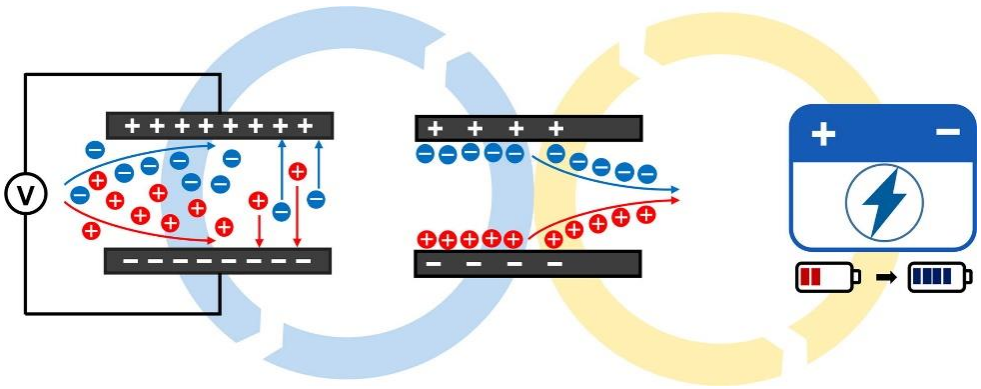

Figure 9. Schematic diagram of energy recovery system and charging based on our-switch buck-boost converter [156]. Copyright 2019, Elsevier.

The two most common charging modes are constant voltage (CV) and constant current (CC) operation. Chen et al. [158] found that the energy recovery rate decreased with increasing charge current, as did the voltage. The research shows that 46.6% of the energy can be recovered by connecting external loads. In addition, the energy recovery rate also showed a significant downward trend with an increasing discharge current. The energy

recovery rate was generally higher in the CC mode than in the CV mode. The main reason for this phenomenon is the severe energy loss caused by ohmic and non-ohmic resistance at higher charging currents.

Zhao et al. [159] and Choi [160] also found that the energy consumption of CC mode is lower than that of CV mode. Another study found that the resistance dissipation of CC mode was smaller than that of CV mode, so the energy consumption was lower [161], although, most studies show that CC mode is more energy-saving than CV mode. However, the CV mode is still widely studied and applied in academic and commercial fields. This is mainly because the CC mode has a certain degree of operational difficulty [155]. This is because it is difficult to control the voltage level of CDI cells in CC mode [155]. More importantly, the most energy-efficient CDI operation mode may not be the best solution. Although a model can achieve energy efficiency, it does not necessarily have a high removal efficiency. For example, although CV mode has higher energy consumption than CC mode, CC mode cannot achieve the required removal efficiency in producing ultrapure water [162].

Recent issues regarding the energy consumption of MCDI have focused on comparative studies of different modes of operation, while the development of new charging modes has been neglected. Choi et al. [163] proposed an advanced staged voltage (SV) mode for the first time. The voltage applied in this mode gradually increases with the operation of the MCDI battery (Figure 10). In flexible operation, the SV mode meets the product water quality requirements and achieves the required removal efficiency and minimum energy consumption, although, the CC mode of operation shows the highest energy efficiency compared to other modes. However, the SV mode may be a promising operating strategy to achieve the required product quality.

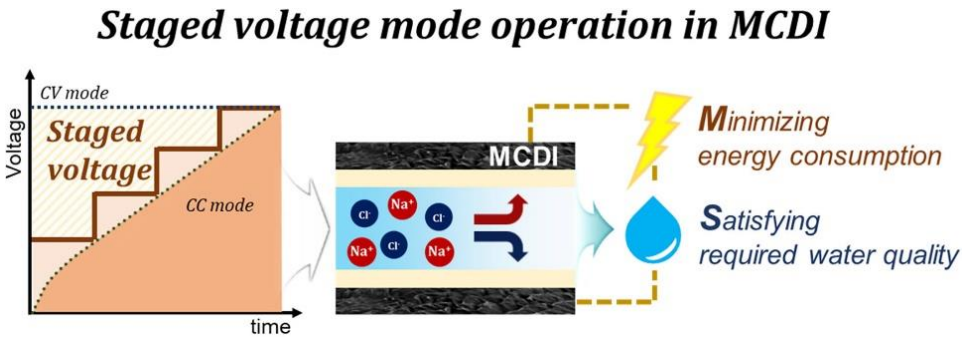

Figure 10. Schematic diagram of SV mode in MCDI [163]. Copyright 2020, Elsevier.

4. Challenges and Future Perspectives

Although CDI technology has made significant progress in water treatment, there are still some key challenges to be solved in the multi-functional application of CDI. Currently, most of the CDI technology application research is only based on laboratory research on simulated water. The composition of synthetic water is simple, while that of actual water is complex. Actual water contains a variety of non-target treatment objects, such as organic, inorganic, and impurity ions, which may hurt the application of CDI and reduce the treatment performance. Therefore, future research should be conducted on the actual water environment to promote CDI technology's commercial application further.

The sustainability of CDI is challenging because of the scaling during operation. Scaling affects the removal of CDI and increases energy consumption. In the future, the collaborative combination of CDI and other technologies should be deeply studied to minimize the impact of scaling on the CDI operation process. Only by promoting the sustainable operation of CDI can the application of expanded CDI technology be further expanded.

The primary research of CDI technology has been extensively studied, and the results have been rich enough. However, there are still few studies on the pilot scale. Therefore,

CDI technology needs more pilot-scale research in seawater desalination, hard water softening, heavy metal removal, organic contaminants removal, phosphate and nitrate removal, water disinfection, energy collection, and other fields.

Current research suggests that energy recovery is feasible for CDI/MCDI systems, but more research still needs to be done. Researchers should do more work to improve energy recovery rates further and promote energy recovery commercialization. With the aggravation of global warming and resource shortage, CDI technology should make further progress in energy conservation and resource recovery.

Electrode materials should be developed for single or multiple target pollutants to achieve efficient removal. Electrode regeneration is also noteworthy as it can reduce energy consumption and economic costs.

5. Conclusions

With the increasing population of the world and the rapid development of industry, the shortage of water resources has become increasingly prominent. Based on the advantages of energy conservation, high efficiency, low cost, green, and pollution-free CDI technology, CDI has been widely used in many fields. This paper introduces the principle of CDI technology and CDI device configuration and comprehensively summarizes the latest research progress and existing problems of CDI technology in water treatment, including seawater desalination, hard water softening, heavy metal removal, organic contaminants removal, phosphate and nitrate removal, water disinfection, and energy collection. The current challenges of the application of CDI technology in water treatment are discussed, and future research directions and prospects of CDI are proposed. Recent research shows that compared with other technologies, CDI technology has a great application prospect in the field of water treatment. This work has significant reference value for promoting the development of CDI technology. CDI is a challenging but potential water treatment technology. Even after decades of development, it is still considered a new water treatment technology.

Author Contributions: For research articles with several authors, Conceptualization, C.X. and S.B.; methodology, C.X., S.B. and B.C.; software, W.D.; validation, Y.L.; formal analysis, C.X.; investigation, C.X.; resources, S.B. and Y.Z.; data curation, C.X.; writing—original draft preparation, C.X.; writing—review and editing, C.X. and S.B.; visualization, C.X.; supervision, Y.Z.; project administration, S.B.; funding acquisition, S.B. All authors have read and agreed to the published version of the manuscript.

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