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STABILITY ENGINEERING OF CAPACITIVE DEIONIZATION FOR WATER TREATMENT: DEGRADATION MECHANISMS, MATERIAL INNOVATIONS, REGENERATION STRATEGIES, AND CELL ARCHITECTURES

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Abstract

Capacitive deionization (CDI) has emerged as a promising electrochemical technology for low- and moderate-salinity water treatment, polishing, decentralized desalination, selective ion removal, water reuse, and resource recovery. Its mild operating conditions, modular design, and tunable ion-storage mechanisms make CDI attractive when moderate salt removal, flexible operation, or ion-selective treatment is required. However, practical deployment remains limited by long-term performance instability under repeated cycling and realistic feedwater conditions. This review develops a stability-engineering framework for understanding and advancing CDI systems, in which material design, operating conditions, regeneration protocols, and cell configurations are treated as interrelated factors that determine durability. First, key degradation mechanisms in classical CDI are analyzed, including asymmetric Faradaic reactions, electrostatic ion trapping, current-collector passivation, inorganic scaling, and synergistic organic fouling. These coupled processes narrow the effective operating window, increase resistance, block ion-transport pathways, and accelerate capacity decay. Second, recent material innovations are reviewed according to structural architectures, including one-dimensional, two-dimensional, three-dimensional, and hybrid electrode systems, with emphasis on their effects on salt adsorption capacity, salt adsorption rate, conductivity, ion accessibility, selectivity, cycling durability, and regeneration tolerance. Third, operational, antifouling, and regeneration strategies are critically evaluated in relation to voltage-window control, polarity reversal, hydrodynamic optimization, pretreatment, physical flushing, chemical cleaning, thermal treatment, and electrochemical regeneration. Finally, CDI cell architectures are discussed from the perspectives of charge efficiency, mass transfer, fouling distribution, scalability, energy use, regeneration compatibility, and long-term stability. By linking degradation mechanisms with material design, operational control, regeneration strategies, and cell architecture selection, this review identifies stability engineering as a key route toward durable and practically viable CDI systems for water treatment.

Keywords: capacitive deionization; stability engineering; degradation mechanisms; electrode material; electrode fouling; electrode regeneration; CDI cell architectures.

1. Introduction

Freshwater scarcity, increasing salinization of water resources, and growing demand for fit-for-purpose water reuse have intensified the need for desalination technologies that are energy-efficient,

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modular, and adaptable to decentralized applications. Conventional desalination processes, particularly reverse osmosis (RO), nanofiltration (NF), electrodialysis (ED), ion exchange, and adsorption, remain essential for many water-treatment applications. However, their suitability depends on feed salinity, target ions, fouling potential, brine management, chemical demand, and system scale. CDI is therefore considered in this review as a complementary electrochemical technology for selected water-treatment applications, not as a universal replacement for established desalination processes, especially for high-salinity desalination [1]. It is most relevant to low- to moderate-salinity brackish water, polishing treatment, decentralized systems, selective ion removal, resource recovery, and some water-reuse applications where moderate ion removal and flexible electrochemical operation are advantageous [2–4]. In conventional or traditional CDI, ion removal is primarily based on non-Faradaic electrosorption through electrical double-layer (EDL) formation at porous electrodes. This mechanism is extended in other CDI variants, where ion intercalation represents a specific Faradaic and redox-driven storage pathway, while redox-assisted storage is used as a broader term that includes intercalation and other reversible charge-transfer processes. Hybrid capacitive-Faradaic mechanisms combine EDL electrosorption with Faradaic ion storage in asymmetric or paired electrode systems. Over the past two decades, CDI has evolved from simple carbon-based electrosorption systems into a broad family of electrochemical deionization technologies, including membrane CDI, flow-through CDI, flow-electrode CDI, hybrid CDI, inverted CDI, and rocking-chair CDI [2, 3, 5].

Despite this progress, the practical implementation of CDI remains limited by long-term performance instability. Many studies report high initial salt adsorption capacity (SAC), rapid salt adsorption rate (SAR), improved charge efficiency, or enhanced ion selectivity under controlled laboratory conditions. However, these metrics often decline during extended cycling or when realistic feedwaters containing multivalent ions, dissolved organic matter, silica, iron, colloids, or biological components are used. This discrepancy indicates that CDI performance is not governed solely by initial surface area, capacitance, or redox activity. Instead, it is determined by the coupled evolution of electrode chemistry, pore accessibility, interfacial transport, current-collector stability, cell architecture, operating mode, and regeneration strategy. The degradation of classical CDI systems originates from several interconnected pathways. Asymmetric electrode polarization can trigger parasitic Faradaic reactions, including anodic carbon oxidation and cathodic oxygen reduction, which generate oxygen-containing surface groups, hydrogen peroxide, hydroxide ions, and local pH gradients [6, 7]. These reactions shift the potential of zero charge (E_{PZC}), narrow the effective operating window, increase charge leakage, and reduce charge efficiency [8]. Electrostatic ion trapping within micropores may prevent complete ion release during discharge, causing capacity inversion and inefficient energy utilization [9]. In addition, current-collector passivation can increase contact resistance and disrupt electron transport, converting part of the applied electrical energy into ohmic losses rather than useful ion electrosorption [10].

Feedwater complexity further amplifies CDI instability. Local pH shifts and ion concentration gradients may induce precipitation of iron, silica, calcium, or magnesium species near the electrode surface or inside porous structures [11–13]. Organic foulants such as humic substances, proteins, and polysaccharides can adsorb onto carbonaceous surfaces through hydrophobic, electrostatic, and hydrogen-bonding interactions [11]. More critically, multivalent cations can bridge organic macromolecules into dense gel-like layers that block ion-transport pathways and increase energy consumption. Consequently, stable CDI operation requires not only high-performance electrodes but also antifouling surfaces, appropriate pretreatment, optimized hydrodynamics, and effective regeneration protocols. Material chemistry innovation has become a major route for addressing these limitations.

Conventional porous carbons remain widely used due to their low cost, high surface area, and chemical robustness, but their performance can be limited by hydrophobicity, inaccessible micropores, weak selectivity, and oxidative instability. Strategies such as heteroatom doping, defect engineering, hierarchical pore construction, and carbon-based composite formation have been developed to improve wettability, conductivity, ion accessibility, and pseudocapacitive contribution [14, 15]. Meanwhile, Faradaic and intercalation materials, including Prussian blue analogues, transition-metal oxides, MXene-based hybrids, phosphate frameworks, and organic/polymeric electrodes, can exceed the capacity limits of purely capacitive carbons but may introduce new challenges such as lattice strain, dissolution, aggregation, and interfacial instability [16, 17]. Operational strategies and cell architectures are equally important. Applied voltage, current mode, cycle duration, polarity reversal, flow velocity, temperature, and pretreatment influence charge leakage, fouling accumulation, ion release, and energy efficiency [18, 19]. Physical flushing, chemical cleaning, thermal treatment, and electrochemical regeneration can recover CDI performance, but each involves trade-offs in cleaning efficiency, material compatibility, chemical use, and energy demand [18–22]. At the system level, cell architecture determines how ion transport, charge transfer, fouling, and regeneration are distributed throughout the CDI unit [2, 3, 5].

Although many reviews have discussed CDI electrode materials, membranes, modeling, reactor configurations, or selective ion removal, a stability-centered synthesis is still needed [23–25]. Degradation mechanisms, material innovation, fouling control, regeneration, and architecture design are often treated separately, yet they are strongly coupled. Faradaic reactions alter surface chemistry and pH; pH shifts accelerate scaling; fouling increases resistance and blocks ion transport; resistance redistributes local potentials; and cell architecture determines whether these processes are suppressed, amplified, or redistributed. In this review, stability engineering is defined as the integrated design, operation, monitoring, and regeneration of CDI systems to preserve ion-removal capacity, charge efficiency, energy performance, electrode integrity, hydraulic performance, and regenerability during extended cycling and realistic feedwater exposure. This definition treats degradation mechanisms, material development, regeneration, and cell configuration as interdependent aspects of CDI stability. It also provides a basis for linking fundamental degradation pathways with practical strategies for durable, regenerable, and scalable water-treatment systems.

This perspective shifts the focus from maximizing initial desalination performance to understanding how CDI systems retain performance during extended operation, realistic feedwater exposure, and repeated regeneration. Material studies commonly emphasize initial SAC, SAR, or capacitance, whereas operational studies often focus on short-term removal efficiency and architectural studies examine charge efficiency or system productivity [2, 3, 5, 15, 20]. However, long-term CDI performance is governed by the interaction among these dimensions. Faradaic reactions alter surface chemistry and local pH [6–8]; pH shifts accelerate scaling and fouling [11–13]; fouling increases resistance and blocks ion transport [11]; and cell architecture determines whether these processes are suppressed, redistributed, or intensified [2, 3, 5].

The literature selection for this narrative review followed a structured strategy. Relevant publications were identified from Web of Science, Scopus, ScienceDirect, and Google Scholar using keyword combinations related to capacitive deionization, CDI stability, electrode degradation, Faradaic reactions, ion trapping, fouling, scaling, regeneration, and CDI cell architectures. Most publications considered were published between 2013 and 2026, while earlier foundational studies were included when necessary to explain CDI principles or historical development. The selection prioritized peer-reviewed experimental studies, review papers with clear synthesis value, and recent reports

on long-term operation, real-feedwater testing, pilot-scale systems, and stability-oriented electrode design. Studies were included when they addressed CDI mechanisms, degradation, material properties, operation, regeneration, or architecture-level performance relevant to long-term stability, and were excluded when they lacked direct relevance or sufficient experimental context.

2. Degradation mechanisms in classical CDI

2.1. Asymmetric Faradaic reactions and operating-window shifts

Ideal ion storage in classical CDI is based on capacitive electrical double-layer (EDL) formation. Because many classical CDI cells are operated in two-electrode mode, the potentials of the individual electrodes are not independently controlled, which can promote unwanted side reactions during cycling. Under these conditions, asymmetric polarization can drive one or both electrodes outside the stable electrochemical window of the electrolyte/electrode interface and promote parasitic Faradaic reactions [7]. The positive and negative electrodes may undergo different interfacial reactions, as illustrated in Fig. 1(a). At high positive potentials, carbon oxidation can introduce oxygen-containing groups and release CO_2, whereas the negative electrode can promote the oxygen reduction reaction (ORR), producing hydrogen peroxide and hydroxyl ions [6]. At the cell scale (Fig. 1(b)), these reactions can generate pronounced pH gradients, with cathodic pH differing substantially from anodic pH [7]. The accumulation of negatively charged surface groups shifts the E_{PZC} toward more positive potentials [7, 8]. These Faradaic reactions consume part of the useful charge and narrow the stable operating window, thereby contributing to reduced charge efficiency and capacity decay [6, 7].

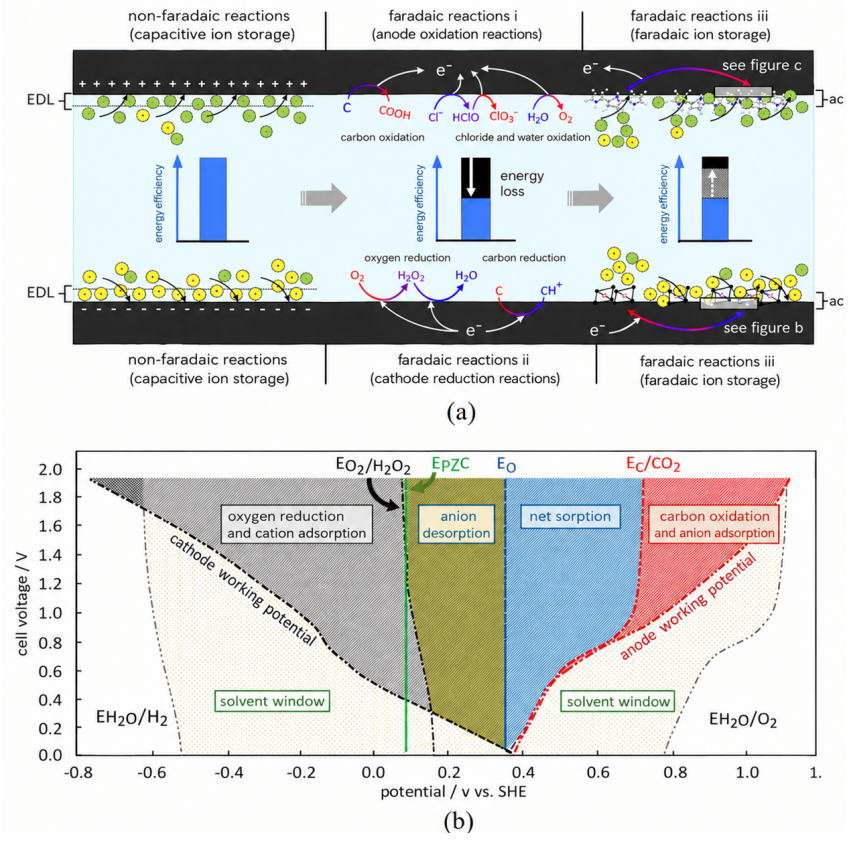


Figure 1. Mechanistic pathways and operating-window effects of Faradaic reactions in classical CDI systems [6, 7] (reproduced with permission)

2.2. *Electrostatic ion trapping and capacity inversion*

One consequence of the positive shift in the E_{PZC} is stronger electrostatic retention of salt ions within the micropore network. During the standard discharge phase at 0 V, cations (typically Na^+) may not be fully released into the bulk solution but can remain trapped near the inner Helmholtz plane (IHP). This retention is associated with the residual negative electrostatic field generated by carboxylate functional groups formed during anodic oxidation. At 0 V, the remaining electrostatic attraction may be insufficient to release these cations completely from the micropore surface; therefore, a reverse-polarity discharge potential (e.g., -1.0 V) may be required to provide sufficient electrostatic driving force for ion release [9].

As residual charges accumulate, the V-shaped E_{PZC} curve can shift into the active working potential window, producing a capacity inversion effect. During the initial charging phase of the subsequent cycle, the positive electrode can then operate partly within the cation-adsorption branch. In this situation, part of the electrical energy supplied from the external circuit may be consumed by co-ion expulsion rather than by useful counter-ion electrosorption. This parasitic energy dissipation is quantitatively captured by the voltage of null conductivity change (V_{ncc}) parameter. Reported correlations between SAC decay and increased V_{ncc} suggest that ion trapping can be a major contributor to CDI degradation. However, the relative importance of ion trapping depends on electrode chemistry, voltage window, dissolved oxygen, electrolyte composition, current-collector stability, and feedwater quality. Therefore, ion trapping is better interpreted as an important electrostatic degradation pathway rather than the sole root cause of capacity loss [8, 9].

2.3. *Current-collector passivation and electronic resistance*

Interfaces between porous carbon electrodes and current collectors are often less visible but important sources of CDI degradation. Electrochemical impedance spectroscopy (EIS) tracking (Fig. 2(a)) shows that the high-frequency Nyquist arc can increase over aging cycles, indicating a progressive rise in interfacial contact resistance (R_{ct}). Material-specific kinetic profiles (Fig. 2(b, c)) indicate that Au and graphite collectors are comparatively stable, whereas Ti foil can undergo substantial R_{ct} increases. Concurrently, the contact capacitance $C_{contact}$ maintains a stable plateau at ˜80 (Fig. 2(c)). These observations suggest that the TiO_2 passivation layer does not necessarily grow as a uniform dielectric film. Instead, localized increases in resistivity may develop due to defect propagation on the oxidized metal surface. This localized dielectric barrier can disrupt the electron percolation network, causing part of the external current to be dissipated as Ohmic loss rather than being used to establish an effective ion-adsorption electric field [10].

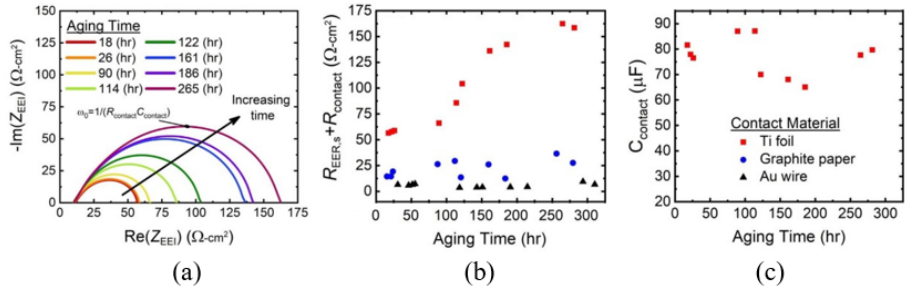


Figure 2. Electronic breakdown caused by current-collector passivation in CDI systems [10] (reproduced with permission)

2.4. Inorganic scaling and synergistic fouling

At the macroscopic interface, ion diffusion from the bulk solution into micropores can be substantially restricted by physical occlusion and mass-transfer barriers. Such blockage often develops from the combined effects of Faradaic side reactions and feedwater chemistry. As shown in Fig. 3(a), inorganic scaling may begin with the competitive electrosorption and exchange of multivalent cations (e.g., Ca^{2+}) within the electrical double layer. When local alkalinization caused by parasitic Faradaic reactions interacts with a Ca^{2+}-rich boundary layer, supersaturation can be triggered, promoting heterogeneous surface crystallization. The resulting non-conductive deposits can occlude micropore entrances and reduce accessible storage volume. In parallel, organic fouling is governed by interfacial interactions. Fig. 3(b) illustrates how hydrophobic effects, pH-dependent hydrogen bonding, and electrostatic interactions promote the attachment of natural organic matter, such as humic substances, to electrified surfaces. The consequences of organic fouling differ strongly depending on feedwater composition, as shown in Fig. 3(c). In the presence of organic macromolecules alone, size exclusion can limit their penetration into sub-nanometer micropores, resulting in relatively limited fouling where intrinsic electrostatic storage is largely preserved. However, when multivalent cations (e.g., Ca^{2+}) coexist with organic matter, these ions can cross-link organic macromolecules into dense, non-conductive gel-like layers. This synergistic gelation can impede interfacial ion diffusion, increase charge-transfer resistance, and cause reversible, partially reversible, or irreversible capacity loss depending on foulant strength, scale location, and the regeneration strategy applied [11].

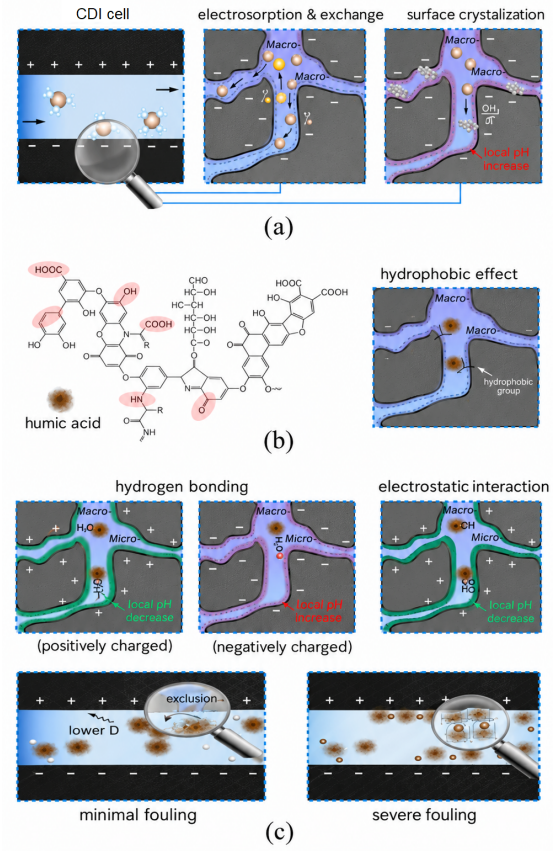


Figure 3. Interfacial mass-transfer barriers caused by inorganic scaling and synergistic organic fouling in CDI [11] (reproduced with permission)

3. Strategies for enhancement

3.1. Material chemistry innovations

Material chemistry innovation is a central pathway for improving both desalination performance and long-term stability in capacitive deionization (CDI). In early CDI systems, porous carbon materials were widely used because of their high specific surface area, low cost, chemical robustness, and relatively simple electrode fabrication [1, 5, 23]. However, the long-term operation of carbon-based CDI is often constrained by inaccessible micropores, limited ion selectivity, slow ion diffusion, hydrophobic surface chemistry, electrostatic ion trapping, and oxidative degradation during repeated cycling [8, 15]. These limitations indicate that high surface area alone is insufficient to ensure durable CDI performance. Instead, electrode materials are better designed to provide not only abundant ion-storage sites, but also fast ion-transport pathways, stable electronic conductivity, resistance to structural degradation, and compatibility with regeneration protocols. Recent material development has therefore shifted from single-function porous carbons toward structurally engineered and chemically functionalized electrodes. Depending on their dimensional configuration, CDI materials can be broadly classified into one-dimensional (1D), two-dimensional (2D), three-dimensional (3D), and hybrid architectures. Each architecture provides different advantages for ion adsorption, electron transfer, mechanical stability, and practical application. To establish a comparative basis, Table 1 summarizes representative CDI electrode materials according to structural architecture, SAC, SAR, and references. The corresponding SAC–SAR distribution is visualized in Fig. 4, which highlights the structure-dependent relationship between ion-storage capacity and desalination kinetics.

Table 1. Salt adsorption performance of representative CDI electrode materials by structural architecture

Name of the electrode materials	Structural architecture	SAC (mg g^{-1})	SAR (mg g^{-1} min^{-1})	Ref.
α-MnO_2	1D	22.1	0.1	[26]
AC Fiber	1D	4.6	0.02	[14]
CNTs-CNTs	1D	11	0.1	[14]
Carbon Nanotubes	1D	6.5	0.2	[27]
Carbon Nanofibers	1D	5.6	0.2	[27]
P-CNF	1D	30.4	1.0	[28]
Mn_2O_3/CNF composite	1D	27.4	2.7	[29]
PAN-based carbon fiber	1D	7.4	0.7	[30]
L-S-$Ti_3C_2T_x$ (MXene)	2D	72.0	1.3	[31]
BNCTs (B, N-doped)	2D	42.5	4.3	[32]
$MGFP_3$ (Graphene Foam)	2D	40.8	2.2	[33]
$Ti_3C_2T_x$ (MXene)	2D	45.0	2.4	[34]
MoS_2	2D	30.0	0.6	[34]
MnO_2 nanosheets	2D	24.1	2.3	[35]
CNT/$NaMnO_2$	2D	32.7	1.8	[35]
MoS_2/CNT composite	2D	25.0	1.2	[31]
Wire-shaped NVP-C	3D	98.0	2.4	[17]
α-MnO_2	3D	70.0	1.8	[34]
$NaTi_2(PO_4)_3$/rGO	3D	44.8	5.6	[36]
Flowerlike PB	3D	101.3	3.4	[32]
NiHCF@3DNC	3D	60.5	1.0	[32]
SPAC	3D	33.1	2.9	[15]
Prussian Blue Analog	3D	77.2	3.0	[37]

Name of the electrode materials	Structural architecture	SAC (mg g^{-1})	SAR (mg g^{-1} min^{-1})	Ref.
$CNT/NaMnO_2$	3D	42.6	3.0	[35]
NiCoFe-PBA/CNT	Hybrid	127.6	4.3	[16]
AC/ZnO Nanorods	Hybrid	123.7	5.1	[38]
PB/PANI/PAN	Hybrid	133.9	7.2	[34]
$Na_{0.44}MnO_2$/BiOCl	Hybrid	140.0	4.8	[34]
$Fe_4[Fe(CN)_6]_3$/rGA//rGA	Hybrid	130.0	4.8	[34]
$Na_3V_2(PO_4)_3$@C//AC	Hybrid	130.0	4.2	[34]
$FeFe(CN)_6$@NPG//AC	Hybrid	125.0	6.6	[34]
NiHCF/CNT-5	Hybrid	116.3	7.2	[16]
VS_2/V_2CT_x	Hybrid	168.8	3.7	[39]
Fe_2O_3@CNFs	Hybrid	115.0	7.8	[28]

Note: The reported SAC and SAR values were collected from different literature sources and experimental conditions, including differences in initial salt concentration, applied voltage or current, flow rate, electrode loading, cycle duration, cell configuration, and calculation basis. Original experimental studies were prioritized where possible. Therefore, these values are presented as representative literature data and are not interpreted as directly comparable performance benchmarks. Entries without comparable SAR values were not included in the SAC–SAR distribution in Fig. 4.

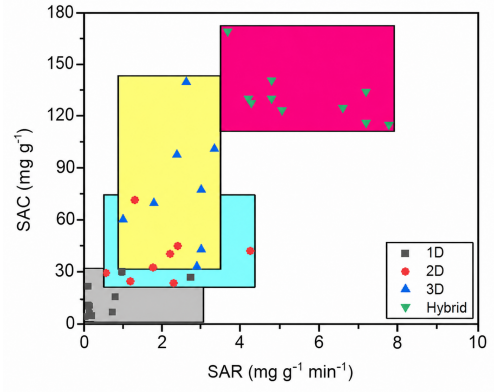


Figure 4. Structure-dependent salt adsorption performance of CDI electrode materials

a. Material structural architectures: 1D, 2D, and 3D materials

One-dimensional materials, including activated carbon fibers, carbon nanotubes, carbon nanofibers, electrospun carbon fibers, porous carbon fibers, and metal-oxide-decorated carbon fibers, are characterized by elongated conductive pathways and relatively direct electron-transport channels. This morphology is advantageous for electrode fabrication because 1D structures can form interconnected networks with good mechanical integrity and reduced electronic resistance. However, the data in Table 1 show that many conventional 1D carbon electrodes provide only moderate salt adsorption capacity. For example, activated carbon fibers, carbon nanotubes, and carbon nanofibers exhibit SAC values of 4.6, 6.5, and 5.6 mg g^{-1}, respectively, indicating that 1D morphology alone is insufficient when accessible pore volume and surface chemistry are limited [14, 27]. Performance can be substantially improved when 1D materials are combined with pore engineering, heteroatom doping, or redox-active decoration. P-doped carbon nanofibers reach a SAC value of 30.4 mg g^{-1}, while Mn_2O_3-decorated carbon nanofibers achieve 27.4 mg g^{-1} with a SAR of 2.7 mg g^{-1} min^{-1} [28–30]. These results suggest that 1D materials are particularly suitable as conductive backbones, flexible electrode frameworks, and mechanically stable supports for flow-by or flow-through CDI systems. Nevertheless, for long-term stability, their pore structure and surface chemistry are most effective when optimized to avoid ion trapping, incomplete desorption, and oxidative aging.

Two-dimensional materials, such as MXenes, graphene-based electrodes, MoS_2, MnO_2 nanosheets, and heteroatom-doped carbon nanosheets, provide large exposed surfaces, short ion-diffusion dis-

tances, and tunable interlayer channels. Their sheet-like morphology is beneficial for rapid ion access and surface-controlled electrosorption or intercalation. The performance trend in Table 1 confirms that 2D materials generally deliver higher SAC than many conventional 1D carbons. For instance, L-S-$Ti_3C_2T_x$ MXene shows a SAC of 72.0 mg g^{-1}, while $Ti_3C_2T_x$ MXene, B,N-doped carbon nanosheets, and mesopore-enhanced graphene foam reach 45.0, 42.5, and 40.8 mg g^{-1}, respectively. These materials are therefore attractive for high-rate desalination, membrane-electrode integration, and applications requiring fast interfacial ion transport. However, their practical stability is often constrained by nanosheet restacking, oxidation, and collapse of interlayer spacing during electrode fabrication or cycling. For MXene-based electrodes in particular, maintaining open interlayer pathways and suppressing aqueous oxidation are critical for preserving long-term SAC and SAR [31–34, 39]. Therefore, 2D architectures are most effective when combined with interlayer spacers, conductive scaffolds, or hybrid structures that prevent restacking and maintain stable ion-accessible channels.

Three-dimensional materials provide interconnected pores, open frameworks, and larger ion-storage domains, making them attractive for high-capacity CDI. Compared with 1D and 2D materials, 3D architectures can combine macropores as electrolyte reservoirs, mesopores as ion-transport channels, and micropores or redox-active sites as ion-storage regions. This multiscale structure explains why several 3D electrodes in Table 1 exhibit high SAC values. Wire-shaped NVP-C, flower-like Prussian blue, α-MnO_2, and Prussian blue analogues show SAC values of 98.0, 101.3, 70.0, and 77.2 mg g^{-1}, respectively [16, 17, 33, 34, 37]. Three-dimensional intercalation and Faradaic materials are therefore suitable for applications requiring higher ion-storage capacity, selective ion capture, or battery-type CDI operation. However, their stability challenges differ from those of purely capacitive carbons. Many 3D Faradaic materials suffer from low intrinsic conductivity, lattice strain, volume change, particle aggregation, and possible active-phase dissolution during repeated ion insertion/extraction. Therefore, 3D materials are promising for high-capacity CDI, HCDI, and RCDI systems, but they require conductivity enhancement, structural buffering, and stable electrode–electrolyte interfaces to maintain cycling durability.

b. Hybrid material architectures

Hybrid material architectures are an important direction in CDI electrode design because they integrate complementary functions that are difficult to achieve within a single material. In general, conductive carbon frameworks such as CNTs, carbon nanofibers, graphene, porous carbon, or MXene provide electron-transport pathways and mechanical support, while Faradaic or intercalation components such as Prussian blue analogues, metal oxides, sulfides, phosphates, and conducting polymers supply additional ion-storage sites. This synergistic design enables hybrid electrodes to address the low-capacity limitation of purely capacitive carbon materials and the poor-conductivity limitation of standalone Faradaic materials. As shown in Table 1 and Fig. 4, hybrid electrodes generally occupy the highest SAC–SAR region. NiCoFe-PBA/CNT achieves a SAC of 127.6 mg g^{-1} and a SAR of 4.3 mg g^{-1} min^{-1}, while NiHCF/CNT-5 reaches a high SAR of 7.2 mg g^{-1} min^{-1} [16]. Other hybrid systems, including PB/PANI/PAN, $Na_{0.44}MnO_2$/BiOCl, $Fe_4[Fe(CN)_6]_3$/rGA//rGA, $Na_3V_2(PO_4)_3$@C//AC, and $FeFe(CN)_6$@NPG//AC, also exhibit SAC values around 125.0–140.0 mg g^{-1} with relatively fast adsorption rates [34]. The highest SAC listed in Table 1 is obtained for VS_2/V_2CT_x, reaching 168.83 mg g^{-1}, while Fe_2O_3@CNFs exhibits a high SAR of 7.8 mg g^{-1} min^{-1} [28, 39]. These results indicate that conductive-redox coupling is effective for improving both ion-storage capacity and desalination kinetics. The practical significance of hybrid architectures lies not only in their high SAC and SAR, but also in their adaptability to different CDI applications. For brackish water desalination and water polishing, hybrid carbon-Faradaic electrodes can increase salt removal capacity without excessively

increasing cell size. For selective ion removal and resource recovery, PBA-, metal oxide-, sulfide-, phosphate-, or polymer-based hybrid electrodes can provide ion-specific storage sites through redox or intercalation mechanisms. For high-rate CDI, carbon nanofiber, CNT, graphene, or MXene scaffolds can shorten electron/ion transport pathways and improve active-site utilization. These properties make hybrid materials especially relevant for HCDI, RCDI, membrane-assisted CDI, and advanced CDI systems requiring both capacity and stability. However, hybridization does not always improve stability or desalination performance. Excessive loading of Faradaic phases may block pores, increase charge-transfer resistance, reduce electrical percolation, or create mechanically weak interfaces. Poorly stabilized active phases may dissolve, aggregate, or undergo irreversible structural distortion during cycling or regeneration. Thus, a stability-oriented hybrid electrode requires a continuous conductive scaffold, well-anchored redox-active phases, open ion-transport pathways, and tolerance to the intended regeneration protocol.

Overall, the comparison in Table 1 and Fig. 4 shows a clear evolution from single-architecture materials toward multifunctional hybrid systems. One-dimensional materials are useful as conductive and mechanically robust frameworks; two-dimensional materials offer large exposed surfaces and fast ion-access pathways; three-dimensional materials provide high-capacity porous or intercalation frameworks; and hybrid architectures integrate these advantages into more balanced CDI electrodes. From the perspective of stability engineering, material innovation is better assessed beyond initial SAC or SAR. Electrode materials should also be evaluated using broader criteria, including charge efficiency, energy consumption, capacity retention, resistance evolution, fouling tolerance, regeneration compatibility, and structural durability. This view supports the main conclusion of this review: durable CDI systems require material architectures that combine high desalination performance with long-term electrochemical, mechanical, and interfacial stability.

3.2. CDI cell architecture

a. Evolution and types of CDI cell architectures

Cell architecture determines how ion transport, charge transfer, fouling, and regeneration are distributed within CDI systems. As illustrated by the representative architectures in Fig. 5 and further discussed below, CDI architectures have evolved from conventional flow-by cells to membrane-assisted, flow-through, flow-electrode, hybrid, inverted, and rocking-chair configurations.

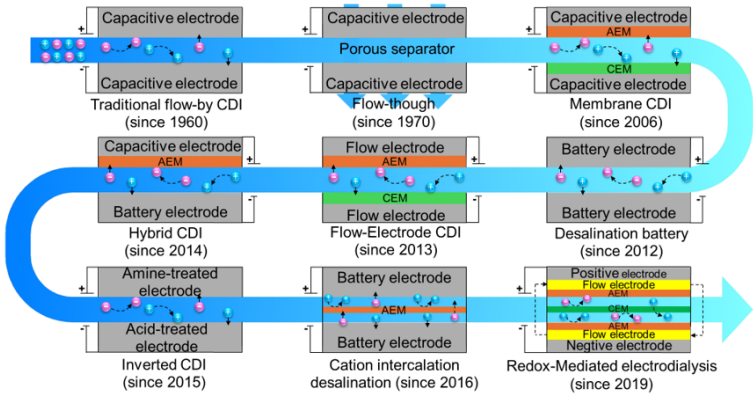


Figure 5. Evolution and classification of CDI architectures (schematic drawn by the authors based on the reviewed literature)

Conventional flow-by CDI is simple but limited by diffusion-controlled ion transport and co-ion expulsion. Membrane CDI incorporates ion-exchange membranes to suppress co-ion leakage and im-

prove charge efficiency, but it introduces membrane resistance, cost, and fouling risk. Flow-through CDI improves mass transfer by forcing water through porous electrodes, increasing adsorption rate but also raising hydraulic pressure drop and pore-clogging risk [3, 5]. Flow-electrode CDI uses carbon slurries as flowable electrodes, enabling continuous desalination and external regeneration, but its performance depends on slurry conductivity, rheology, pumping energy, and particle stability [2]. Hybrid CDI, inverted CDI, and rocking-chair CDI represent advanced architectures for improving capacity and stability. Hybrid CDI combines capacitive and Faradaic electrodes to increase ion-storage capacity. Inverted CDI modifies the adsorption/desorption sequence to reduce anodic oxidation. Rocking-chair CDI uses paired Faradaic electrodes to balance ion insertion and extraction, reducing capacity mismatch and improving cycling stability [3, 5, 16]. These architectures show that cell design can actively mitigate degradation rather than merely host electrode materials.

b. Electrode–cell integration and architecture-level material design

While Section 3.1 discusses electrode materials according to their intrinsic structural architectures, the translation of these materials into stable CDI performance depends strongly on how they are integrated into specific cell configurations. At the architecture level, material design is no longer limited to maximizing surface area, conductivity, or redox activity; it also considers electrode thickness, packing density, hydraulic permeability, current-collector contact, membrane compatibility, and regeneration accessibility. For example, highly microporous carbons may exhibit large theoretical ion-storage capacity, but their practical performance can be limited in thick electrodes if ion diffusion pathways are tortuous or if desorption is incomplete. Similarly, high-capacity Faradaic materials may improve SAC in hybrid or rocking-chair CDI, but their use requires careful electrode balancing, stable electrical contact, and compatible voltage windows to avoid capacity mismatch, lattice strain, or active-phase dissolution [15–17, 37]. The optimal structural design therefore depends on the CDI architecture. In flow-by CDI, porous carbon electrodes need to provide accessible mesopores and stable electrode–current collector interfaces because ion transport is mainly diffusion-controlled from the spacer channel into the electrode. In flow-through CDI, the electrode also needs to maintain hydraulic permeability and mechanical robustness, since feedwater is forced directly through the porous network and pore clogging can rapidly increase pressure drop. In MCDI, electrode structure needs to be compatible with ion-exchange membranes, because membrane resistance and fouling can offset the gain in charge efficiency. In FCDI, material design shifts from fixed porous electrodes toward flowable carbon suspensions, where slurry conductivity, particle dispersion, viscosity, and regeneration behavior become decisive factors [2, 3, 19]. For HCDI and RCDI, Faradaic or intercalation electrodes are selected not only for high ion-storage capacity but also for structural reversibility and charge balance between the two electrodes.

From the perspective of stability engineering, architecture-level material design therefore needs to connect electrode properties with cell hydrodynamics, charge distribution, and regeneration strategy. A material that performs well in a thin laboratory electrode may not retain the same SAC, SAR, or energy efficiency in a stacked module if electrical contact, ion transport, or hydraulic flow is poorly controlled. Likewise, an electrode that resists fouling under flow-by conditions may behave differently in flow-through or membrane-assisted configurations, where foulants can accumulate inside pores or at membrane interfaces. Thus, future CDI development can treat electrode material selection and cell architecture as a co-design problem. Stable CDI performance requires matching material dimensionality, pore structure, current-collector design, membrane arrangement, flow configuration, and regeneration protocol to the target feedwater and treatment objective.

c. Application-oriented selection of CDI cell architectures

The practical application of CDI depends strongly on matching cell architecture with feedwater characteristics, treatment objectives, fouling risk, and regeneration requirements. As illustrated in Fig. 5, different CDI configurations are not interchangeable; each architecture provides a distinct balance among ion-transport efficiency, charge efficiency, hydraulic complexity, regeneration mode, scalability, and long-term stability. Therefore, CDI cell selection should consider not only maximum salt adsorption capacity or laboratory-scale removal efficiency, but also the ability of the configuration to maintain stable performance under the intended water-treatment conditions. Conventional flow-by CDI remains attractive for low-salinity polishing, small-scale brackish water desalination, and decentralized water-treatment systems because of its simple structure, low hydraulic complexity, and relatively easy electrode fabrication. In this configuration, feedwater flows through the spacer channel between two porous electrodes, while ions diffuse into the electrode pores under the applied electric field. This simplicity makes flow-by CDI suitable for applications where moderate salt removal, low operating pressure, and compact system design are prioritized. However, its application is limited when high removal rate, high water recovery, or stable operation under complex feedwater is required. Because ion transport is mainly diffusion-controlled, flow-by CDI may suffer from slow adsorption kinetics, co-ion expulsion, charge leakage, and electrode asymmetry. Therefore, it is best suited for relatively clean, low- to moderate-salinity waters rather than highly fouling or high-salinity streams. Membrane capacitive deionization (MCDI) is more suitable when charge efficiency, ion retention, and effluent-quality stability are critical. By incorporating cation- and anion-exchange membranes in front of the electrodes, MCDI suppresses co-ion leakage and improves the fraction of electrical charge used for effective counter-ion adsorption [1, 2]. This makes MCDI attractive for water reclamation, brackish groundwater treatment, polishing after biological or membrane processes, and applications requiring more stable desalination performance. Pilot-scale MCDI studies also indicate its potential for water reuse when system commissioning, pretreatment, and long-term operation are properly controlled [4]. Nevertheless, membrane resistance, membrane fouling, scaling, and chemical-cleaning compatibility remain important limitations. Thus, MCDI is most appropriate when the benefit of improved charge efficiency outweighs the added cost and operational complexity of ion-exchange membranes.

Flow-through CDI is advantageous for applications requiring faster ion removal and higher mass-transfer rates. In this architecture, feedwater is forced directly through porous electrodes rather than flowing only through a spacer channel, thereby shortening diffusion pathways and improving access to internal ion-storage sites [1, 2]. This makes flow-through CDI suitable for high-throughput desalination, compact modules, or treatment scenarios where adsorption kinetics are more important than structural simplicity. However, its application is more sensitive to feedwater quality. Suspended solids, organic aggregates, iron precipitates, silica, or scaling precursors can penetrate the porous electrode network, increase pressure drop, and accelerate pore clogging. Therefore, flow-through CDI is preferably combined with effective pretreatment, such as filtration or activated media, especially for realistic waters containing colloids or organic matter [24]. Its practical value lies in high-rate operation, but this advantage can only be maintained if hydraulic resistance and internal fouling are controlled. Flow-electrode CDI (FCDI) is particularly relevant for continuous desalination, brine concentration, and higher-salinity feedwaters. Unlike fixed-electrode CDI, FCDI uses flowable carbon slurries as the electrode phase, allowing ion adsorption and electrode regeneration to be partially decoupled. This configuration is attractive for applications where continuous operation, external regeneration, and high salt loading are required, including industrial brine treatment

and zero-liquid-discharge-oriented processes [3, 18]. However, the practical application of FCDI is constrained by slurry conductivity, viscosity, pumping energy, particle aggregation, and flow-channel management. Therefore, FCDI is not necessarily the best option for simple low-salinity polishing, but it becomes more attractive when fixed electrodes are limited by finite adsorption capacity or when continuous operation is a major requirement. Hybrid CDI (HCDI) and rocking-chair CDI (RCDI) are more suitable for high-capacity desalination, selective ion capture, and resource recovery. These architectures incorporate Faradaic or intercalation electrodes, such as Prussian blue analogues, Na-ion intercalation compounds, and other battery-type materials, to overcome the capacity limitation of purely capacitive carbon electrodes [15, 16, 36]. HCDI typically combines one capacitive electrode with one Faradaic electrode, whereas RCDI uses paired Faradaic electrodes to balance ion insertion and extraction. These architectures are promising for applications where high SAC, ion selectivity, and stable redox-based ion storage are required. However, their practical use requires careful electrode balancing, voltage-window control, and structural stabilization to avoid capacity mismatch, lattice strain, active-material dissolution, and irreversible phase transformation. Therefore, HCDI and RCDI are most appropriate when the target application justifies the added material complexity, such as selective recovery of valuable ions, nutrient capture, or high-capacity brackish water desalination.

Inverted CDI provides a specialized stability-oriented configuration. Its main purpose is not necessarily to maximize salt adsorption capacity, but to mitigate carbon oxidation and extend electrode lifetime when anodic degradation is the dominant lifetime-limiting pathway [1, 2, 14]. By modifying the conventional adsorption/desorption sequence and using surface-modified electrodes, inverted CDI can reduce the exposure of carbon electrodes to oxidative conditions. This makes it attractive for systems where long-term carbon stability is more important than maximum short-term desalination capacity. However, inverted CDI requires precise control of electrode surface chemistry and operating sequence, and its performance may be sensitive to feedwater composition and electrode aging. Therefore, it is better considered as a targeted architecture for stability enhancement rather than a universal replacement for conventional CDI. Overall, the architecture-specific comparison indicates that CDI cell configurations differ not only in desalination performance, but also in stability-related trade-offs, including ion-transport efficiency, fouling distribution, regeneration compatibility, scale-up feasibility, and operational durability. Conventional flow-by CDI offers simplicity and low hydraulic complexity but is more vulnerable to diffusion limitation and surface fouling, whereas MCDI improves charge efficiency and effluent stability but may introduce membrane scaling, organic fouling, and cleaning-compatibility constraints. Flow-through CDI enhances mass transfer but increases the risk of internal pore blockage and pressure-drop growth under complex feedwaters. FCDI allows continuous operation and external electrode regeneration, but scale-up depends on slurry conductivity, viscosity, pumping energy, and particle stability [2, 3, 5, 11, 18]. HCDI and RCDI provide higher capacity and selectivity through Faradaic or intercalation electrodes, yet their durability depends on charge balance, voltage-window control, active-phase stability, and regeneration tolerance [16, 36]. Inverted CDI is most relevant when carbon oxidation limits lifetime, but its use requires careful control of electrode surface chemistry and operating sequence [14]. These architecture-specific trade-offs show that stable CDI design depends on matching the cell configuration with feedwater quality, fouling tendency, target ions, regeneration method, and module-scale operating conditions. For practical deployment, architecture selection also needs to be evaluated together with long-term operation, real-feedwater performance, module scale-up, and cost-related constraints. Laboratory-scale SAC and SAR values do not directly translate into stable field performance if the system suffers from flow maldistribution, pressure-drop growth, membrane fouling, slurry-handling difficulty, current-collector

degradation, or incomplete regeneration. Pilot-scale and real-feedwater studies are therefore important for assessing whether a CDI configuration can maintain stable ion removal, charge efficiency, hydraulic performance, and regeneration effectiveness under realistic water matrices [4, 40]. In addition, techno-economic and life-cycle considerations, including electrode lifetime, membrane replacement, pumping energy, cleaning chemical demand, brine management, and module maintenance, are important when comparing CDI with RO, NF, ED, ion exchange, or adsorption for specific applications. This application-based analysis supports the main conclusion of this review: durable CDI performance requires integrated design from material selection to cell configuration and operational control.

3.3. Operational strategies and regeneration protocols

a. Operational strategies for fouling and aging mitigation

Operational control is one of the most effective approaches for mitigating fouling, electrode aging, and long-term performance decay in CDI. Among operational parameters, the applied voltage window is particularly critical because it determines whether the system operates predominantly through reversible ion electrosorption or enters a regime dominated by parasitic Faradaic reactions. Although increasing the applied voltage can enhance salt removal, excessive polarization, particularly beyond approximately 1.2–1.5 V, may promote water electrolysis, cathodic oxygen reduction, and anodic carbon oxidation. These side reactions consume charge without contributing to useful desalination, generate local pH gradients, and accelerate the formation of oxygen-containing functional groups on carbon electrodes. As a result, the electrode surface chemistry, micropore structure, and potential of zero charge may be progressively altered, leading to charge leakage, ion trapping, and irreversible loss of salt adsorption capacity [6, 8, 18]. Therefore, voltage optimization is considered a primary stability-control strategy rather than merely a means of increasing short-term salt removal. Cell configuration and electrical operating mode can further influence the rate of aging. Compared with conventional membrane-free CDI, membrane capacitive deionization (MCDI) can improve charge efficiency by suppressing co-ion leakage and limiting undesirable ion transport near the electrode interface [3, 5]. In long-term operation, the use of ion-exchange membranes may also contribute to more stable desalination performance by reducing the exposure of electrodes to species that accelerate oxidative degradation, although membrane resistance and fouling requires careful management. In addition, constant-current operation can provide more stable effluent quality than constant-voltage operation because the charge input is more directly controlled during each cycle. This is particularly useful in water reclamation or multi-stage systems, where maintaining predictable conductivity reduction is more important than maximizing instantaneous adsorption rate [3, 40]. Cycle duration is another key factor. Excessively long adsorption periods can increase charge leakage, ion trapping, and foulant accumulation, whereas shorter adsorption–desorption cycles or intermittent relaxation can delay the transition from reversible adsorption to irreversible fouling. Reverse voltage desorption and polarity reversal can further enhance ion release and weaken the attachment of charged foulants or early-stage inorganic scales, but overly frequent switching may increase energy consumption and impose additional stress on electrical components [11, 18, 19].

Hydrodynamic control provides a complementary physical pathway for reducing fouling. Increasing cross-flow velocity can enhance shear at the electrode or membrane surface and limit the deposition of weakly attached foulants. For example, moderate increases in cross-flow velocity have been associated with improved normalized flux, indicating more effective surface washing and reduced fouling accumulation. However, higher flow velocity is not always beneficial. Excessive hydrodynamic forcing may transport denatured protein aggregates, polysaccharides, colloids, or other foulant

precursors deeper into spacer channels and porous electrodes, thereby intensifying internal blockage and increasing hydraulic resistance. Temperature also affects fouling behavior. Elevated temperature may improve ion mobility, but it can also promote protein denaturation and the formation of dense organic–calcium or polysaccharide–calcium complexes, which generate high-resistance gel-like layers at the interface [11]. These observations suggest that hydrodynamic and thermal conditions are most effective when optimized according to feedwater composition rather than selected solely to increase mass transfer. For complex feedwaters, operational optimization alone is insufficient unless combined with pretreatment. Industrial brines, secondary effluents, and reclaimed waters often contain dissolved organic matter, colloids, hardness ions, silica, iron, and biological components that can accelerate fouling and scaling. Integrated pretreatment barriers such as biological activated carbon, ultrafiltration, granular activated carbon, or activated filter media can reduce organic loading, suspended solids, and silt density index before CDI treatment. Such pretreatment reduces the burden on the CDI cell and extends stable operating periods by limiting the entry of foulants into electrode pores and spacer channels. Nevertheless, even with pretreatment and optimized operation, gradual accumulation of mineral scales and organic deposits is difficult to avoid. Periodic mild chemical cleaning, including acidic cleaning for inorganic scaling and alkaline cleaning for organic build-up, may therefore be required to restore desalination performance [18, 19].

b. Regeneration protocols and cleaning methods

The long-term operation of CDI systems is strongly constrained by the progressive accumulation of reversible and irreversible degradation products, including weakly attached foulants, inorganic precipitates, trapped ions, and oxidized surface groups. Therefore, regeneration is not a secondary maintenance step; it is an integral component of CDI stability engineering. As illustrated in Fig. 6, four major regeneration pathways can be distinguished: physical flushing, chemical cleaning, thermal regeneration, and electrochemical regeneration. These approaches differ fundamentally in their recovery mechanisms and are selected according to the dominant degradation pathway.

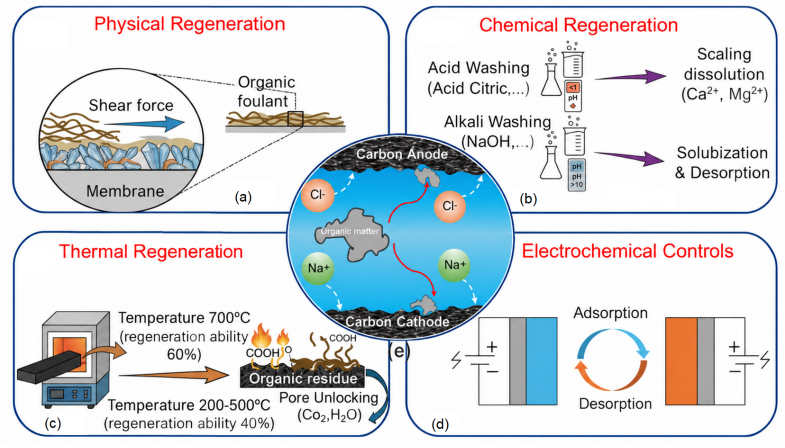


Figure 6. Schematic illustration of regeneration pathways in CDI systems (schematic drawn by the authors based on the reviewed literature)

In Fig. 6(a), physical flushing represents the most direct and least invasive regeneration method. It relies on hydraulic shear to detach loosely bound particles, weakly adsorbed organic matter, and surface deposits from the flow channel, spacer, and external electrode surface. Because no additional chemicals are required, this method is suitable for routine operation and early-stage fouling control. However, its recovery capacity is limited when foulants are strongly adsorbed, when inorganic

scales precipitate inside micropores, or when organic matter penetrates deeply into the porous electrode structure [18]. Thus, physical flushing is most effective as a preventive strategy for controlling reversible deposition, rather than as a complete recovery method for severely aged CDI electrodes. Chemical cleaning, shown in Fig. 6(b), is required when performance loss is dominated by persistent inorganic scaling or strongly attached organic foulants. Acidic cleaning can dissolve mineral precipitates associated with hardness ions, iron, or silica, while alkaline cleaning can increase the ionization and solubility of humic-like organic matter, thereby facilitating its detachment from the carbon matrix [18, 19, 22]. This pathway is therefore more powerful than physical flushing for restoring blocked ion-transport channels. Nevertheless, chemical cleaning introduces an important material-compatibility trade-off. Excessive acidity, alkalinity, reagent concentration, or exposure time may damage binders, ion-exchange membranes, current collectors, and redox-active phases. This issue is particularly critical for hybrid CDI, MXene-based electrodes, Prussian blue analogues, and other Faradaic materials, whose active phases may be sensitive to pH-induced dissolution or oxidation. Chemical cleaning is therefore applied selectively and optimized according to the type of foulant and electrode chemistry.

Thermal regeneration, presented in Fig. 6(c), targets a different degradation mechanism. Instead of removing external foulants, it is mainly used to recover aged carbon electrodes whose performance decline is associated with oxidative surface modification. Heat treatment can decompose oxygen-containing functional groups, reopen blocked pores, reduce electrical resistance, and partially restore electrosorption capacity [18]. This makes thermal regeneration especially relevant when anodic carbon oxidation and surface functionalization are the dominant causes of capacity loss. However, thermal regeneration is energy-intensive, usually conducted ex situ, and difficult to integrate into continuous CDI operation. For this reason, it is more appropriate as a periodic deep-regeneration method for severely degraded carbon electrodes, rather than a routine cleaning protocol. Electrochemical regeneration, illustrated in Fig. 6(d), is the most operationally integrated strategy because it can be incorporated directly into the CDI charge–discharge cycle. Short-circuiting, reverse voltage desorption, and polarity reversal can promote ion release, reduce electrostatic ion trapping, and weaken the attachment of charged foulants to electrode surfaces [18, 19]. This pathway is particularly valuable when capacity loss is associated with trapped ions, incomplete desorption, or weakly attached charged species. However, electrochemical regeneration also requires careful control. Excessive reverse voltage or overly frequent polarity reversal may trigger additional Faradaic reactions, water electrolysis, carbon corrosion, or active-material degradation [6, 18]. Therefore, the effectiveness of electrochemical regeneration depends on the optimization of voltage amplitude, regeneration duration, and switching frequency.

Overall, the four pathways in Fig. 6 are best viewed as complementary rather than interchangeable. A rational CDI regeneration strategy can be organized in stages: physical flushing for routine removal of reversible deposits, electrochemical regeneration for trapped ions and weakly attached charged foulants, chemical cleaning for persistent scaling or organic fouling, and thermal regeneration for deeply oxidized carbon electrodes. In practice, regeneration can be triggered by early diagnostic indicators such as increasing cell resistance, declining charge efficiency, delayed desorption, rising pressure drop, or unstable effluent conductivity [11, 18, 32, 40]. These indicators help identify whether the dominant degradation mode is hydraulic fouling, electrostatic ion trapping, inorganic scaling, or electrode aging.

c. Antifouling material design for regeneration-compatible CDI operation

Antifouling material design can be viewed as a preventive strategy that complements operational control and regeneration protocols. While Section 3.3.2 focuses on restoring performance after fouling, scaling, ion trapping, or electrode aging has occurred, antifouling design aims to delay the onset of these degradation processes during long-term CDI operation. This distinction is important because practical CDI systems are often exposed to complex feedwaters containing dissolved organic matter, multivalent ions, colloids, silica, iron, and microorganisms. Under such conditions, performance decay is not caused only by insufficient salt adsorption capacity, but also by progressive interfacial blockage, increased charge-transfer resistance, delayed desorption, and reduced regeneration efficiency. Therefore, antifouling materials are developed not merely to enhance initial SAC or SAR, but to preserve ion-transport pathways, reduce foulant adhesion, and remain compatible with repeated cleaning or electrochemical regeneration. A major direction is the regulation of electrode surface chemistry to reduce organic adhesion. Pristine porous carbon surfaces may interact with humic substances, proteins, and polysaccharides through hydrophobic attraction, hydrogen bonding, and electrostatic interactions, which can initiate organic fouling and promote subsequent organic-inorganic complexation [11]. Hydrophilic coatings, zwitterionic polymer brushes, and PEG/TiO_2-based surface layers can form hydration barriers that weaken the attachment of organic macromolecules and reduce protein adsorption [22]. However, these protective layers need to be carefully controlled. If the coating is too thick or poorly conductive, it may increase mass-transfer resistance, reduce accessible surface area, and lower electrosorption efficiency. Thus, surface modification for CDI requires a balance between antifouling protection, ionic permeability, electrical conductivity, and mechanical durability.

A second strategy is to design pore structures that remain functional under partial fouling. In practical operation, complete prevention of fouling is unlikely; therefore, the electrode needs to maintain alternative ion-transport pathways even when part of the pore network is blocked. Hierarchical porous structures are particularly relevant because macropores can serve as electrolyte reservoirs, mesopores can provide diffusion channels, and micropores can contribute to ion storage [20]. This multiscale structure helps reduce the sensitivity of the electrode to localized pore blockage and supports more effective flushing or electrochemical desorption. From a stability-engineering perspective, hierarchical porosity is therefore viewed not only as a route to improve desalination kinetics, but also as a structural buffer against fouling-induced mass-transfer limitation. Electrochemical stability is also an important consideration in antifouling electrode design. Defect-engineered and nitrogen-doped carbons can improve wettability, tune surface charge, and suppress undesirable oxygen reduction pathways, thereby reducing carbon corrosion and the formation of unstable oxygen-containing groups during cycling [15]. This is important because oxidative surface aging can promote charge leakage, potential-of-zero-charge shifts, ion trapping, and capacity inversion. Nevertheless, surface functionalization is most effective when carefully optimized. Moderate heteroatom doping or defect engineering can improve electrolyte accessibility and stability, whereas excessive oxygen-rich functional groups may intensify parasitic Faradaic reactions and accelerate electrode aging. Therefore, antifouling design aims to avoid creating surfaces that are hydrophilic but electrochemically unstable.

Biofouling and mixed organic-inorganic fouling also require attention in long-term CDI operation. MXene/carbon nanofiber composites, for example, have been reported to provide both high conductivity and antifouling or antibacterial functionality, offering a potential route for reducing biological attachment while maintaining charge transport. Such multifunctional electrodes are attractive because realistic feedwaters rarely contain only one foulant type. However, MXene-based and other

hybrid electrodes need careful evaluation under repeated cycling and regeneration conditions, since oxidation, restacking, or active-phase instability may reduce long-term performance [21]. Thus, antifouling functionality is most useful when introduced without compromising structural stability or regeneration tolerance. Overall, antifouling material design is most effective when integrated with operating mode and regeneration strategy. A surface that resists organic adhesion but cannot tolerate acidic, alkaline, or electrochemical regeneration may still become unsuitable for practical operation. Conversely, an electrode with high SAC may lose its advantage rapidly if foulants block ion pathways or if the active phase degrades during cleaning. This leads to four practical design guidelines for future antifouling CDI electrodes: low foulant adhesion, stable ion-accessible porosity, electrochemical resistance to oxidative aging, and compatibility with physical, chemical, thermal, or electrochemical regeneration. In this sense, antifouling material design complements regeneration protocols and reinforces the main conclusion of this review: durable CDI performance requires the co-design of material surface chemistry, pore architecture, operating conditions, and regeneration-compatible cell operation.

4. Conclusions

This review frames CDI stability as a system-level challenge governed by coupled degradation, material, operational, regeneration, and architectural factors rather than by initial desalination capacity alone. Faradaic reactions, carbon oxidation, oxygen reduction, ion trapping, current-collector passivation, inorganic scaling, and synergistic organic fouling can interact to narrow the operating window, increase resistance, block ion transport, and accelerate capacity decay. Stability engineering therefore requires integrated design across materials, operation, regeneration, and cell architecture. Material innovations, including 1D, 2D, 3D, and hybrid electrodes, can improve SAC, SAR, conductivity, and ion accessibility, but their practical value depends on capacity retention, resistance evolution, fouling tolerance, active-phase stability, and regeneration compatibility. CDI architectures and operating strategies are also most effective when selected according to feedwater characteristics, degradation risks, and treatment objectives. Voltage-window control, hydrodynamic optimization, pretreatment, antifouling design, and physical, chemical, thermal, or electrochemical regeneration are essential for maintaining long-term performance. Overall, future CDI research should move beyond short-term SAC and SAR toward stability-oriented evaluation under realistic feedwaters and extended cycling. By integrating degradation understanding with material design, operational control, regeneration strategies, and architecture selection, CDI can advance toward more reliable, regenerable, and scalable water-treatment applications.

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Declaration of generative AI and AI-assisted technologies

The author(s) declare that no generative AI or AI-assisted technologies were used in the preparation of this manuscript.

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