



HAL
open science

Exploring flow-electrode capacitive deionization: an overview and new insights

Myriam Tauk, Philippe Sizat, Roland Habchi, Marc Cretin, François Zaviska, Mikhael Bechelany

► **To cite this version:**

Myriam Tauk, Philippe Sizat, Roland Habchi, Marc Cretin, François Zaviska, et al.. Exploring flow-electrode capacitive deionization: an overview and new insights. *Desalination*, 2025, 597, pp.118392. <10.1016/j.desal.2024.118392>. <hal-05081633>

HAL Id: hal-05081633

<https://hal.science/hal-05081633v1>

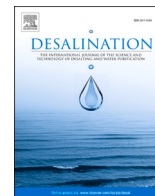
Submitted on 27 Jun 2025

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



Distributed under a Creative Commons CC BY 4.0 - Attribution - International License



Exploring flow-electrode capacitive deionization: An overview and new insights

Myriam Tauk^a, Philippe Sizat^a, Roland Habchi^b, Marc Cretin^a, Francois Zaviska^a, Mikhael Bechelany^{a,c,*}

^a Institut Européen des Membranes, IEM, UMR-5635, University of Montpellier, ENSCM, CNRS, Place Eugène Bataillon, CEDEX 5, 34095 Montpellier, France

^b EC2M, Faculty of Sciences, Campus Pierre Gemayel, Lebanese University, Fanar 90656, Lebanon

^c Functional Materials Group, Gulf University for Science and Technology (GUST), Masjid Al Aqsa Street, Mubarak Al-Abdullah 32093, Kuwait

HIGHLIGHTS

- FCDI facilitates continuous operation by enabling material regeneration external to the cell.
- The resistance and charge transport within the flow-electrode play crucial roles in determining its performance in FCDI.
- The charge transfer mechanism is contingent upon the composition of the electrode materials and the operational mode.
- Improving electronic conductivity in the flow electrode is key to enhancing FCDI performance.
- Desalination, heavy metal recovery, and nutrient retrieval are research areas in FCDI.

ARTICLE INFO

Keywords

Flow-electrode capacitive deionization
Desalination
Ion selectivity
Electric double-layer capacitors
Electrosorption
Charge transport

ABSTRACT

Amidst the escalating severity of global water scarcity, a diverse array of scientific undertakings is devoted to the advancement of technologies concerning water desalination. A notably promising advancement in this field is known as flow-electrode capacitive deionization (FCDI). This pioneering electrochemical methodology represents a derivation of capacitive deionization (CDI) technology, amalgamating ion-exchange membranes with flow particle-based electrodes. Over the preceding seven years, FCDI has garnered considerable interest due to its potential for energy-efficient, sustainable, and consistently high-quality production of fresh water. Additionally, it affords flexibility in the management of particle electrodes and concentrated streams. In this review, an all-encompassing survey of the recent strides in the realm of FCDI is provided. The focal point encompasses the fundamental principles of FCDI, diverse designs including cell architecture, operational modes, choices of flow electrodes and strategies for optimizing their performance in FCDI. This review thoroughly examines the complex mechanisms of mass and charge transfer within flow electrodes in FCDI. It also explores the impact of feed saline water and applied voltage, detailing their respective roles in the FCDI process. A detailed analysis of ion selectivity is provided, alongside discussions on the environmental applications of FCDI in areas such as water desalination, resource recovery, and the removal of waterborne pollutants. Clear definitions and performance metrics are established, and the review concludes with a forward-looking perspective on the future potential of FCDI technology, highlighting key areas requiring focused attention moving forward.

1. Introduction

Water scarcity is a critical global challenge, exacerbated by population growth, climate change, and the uneven distribution of freshwater resources. Addressing this issue is essential for both environmental

sustainability and social equity [1,2]. One of the primary approaches to mitigating water scarcity is water desalination [3], with reverse osmosis (RO) being the most efficient for high-salinity waters like seawater [4,5]. However, moderately saline waters, such as those in estuaries and inland lakes, highlight the need for innovative desalination methods.

* Corresponding author at: Institut Européen des Membranes, IEM, UMR-5635, University of Montpellier, ENSCM, CNRS, Place Eugène Bataillon, CEDEX 5, 34095 Montpellier, France.

E-mail address: mikhael.bechelany@umontpellier.fr (M. Bechelany).

<https://doi.org/10.1016/j.desal.2024.118392>

Received 23 July 2024; Received in revised form 25 November 2024; Accepted 30 November 2024

Available online 1 December 2024

0011-9164/© 2024 The Author(s). Published by Elsevier B.V. This is an open access article under the CC BY license (<http://creativecommons.org/licenses/by/4.0/>).

Electrochemical desalination techniques, including electrodialysis (ED) and capacitive deionization (CDI), are emerging as energy-efficient alternatives, particularly for low to moderate salinity waters (Fig. 1). Capacitive deionization technology, in particular, has gained significant research interest and popularity, especially for brackish water treatment [6–10]. It offers several advantages, including low energy consumption and the ability to regulate ion composition and concentration of the treated water through selective electrosorption, voltage and flow rate adjustments, and multi-stage processing. This makes CDI a versatile and energy-efficient solution for water treatment, especially in scenarios where fine control over ion removal and retention is required. Capacitive deionization was introduced as a conceptual notion in 1960 by Blair and Murphy [11], for the removal of charged entities from aqueous solutions, leveraging either electrostatic or electrochemical intercalation mechanisms. This technology operates based on the principle of ion adsorption, where porous carbon electrodes attract and temporarily trap charged species from the saline solution, generating desalted water [7,12,13]. This technology have been selected for various applications, including brackish water desalination [14], sea water desalination [15,16], wastewater remediation [17], and water softening [18–20]. It operates at low voltages (1–1.4 V) and pressure, resulting in energy-efficient and cost-effective process. Additionally, it can be powered by renewable energy sources, making it suitable for remote off-grid locations. The capacitive effect during ion sorption within the electrode double layers also presents the potential for energy recovery. Unlike traditional desalination methods, capacitive deionization technology with its distinct forms, offers inherent ion selectivity, enabling the removal of specific ions from water and providing customization options for various applications. Capacitive deionization comes in two main configurations: Conventional CDI and Flow-Electrode CDI (FCDI). CDI uses static carbon electrodes for ion adsorption via electrostatic or electrochemical mechanisms, operating intermittently. FCDI combines ion-selective membranes with flowable electrodes for continuous operation, offering more efficient desalination. Conventional CDI operates intermittently due to the need for solid electrode regeneration after saturation, while FCDI enables continuous operation, making it a focus of interest [21]. FCDI removes salt primarily through capacitive adsorption, where ions migrate under an electric field and accumulate in oppositely charged electrodes [22]. Additionally, FCDI benefits from fluidized electrodes containing active materials and conductive

additives [23]. Its continuous operation and efficient desalination are facilitated by introducing fresh or regenerated particle electrodes, with potential applications in resource recovery [24–26]. FCDI’s integration with renewable energy sources could further enable use in remote off-grid locations [22,27].

In the past decade, there has been a notable upswing in scholarly investigation concerning FCDI, with particular emphasis on the refinement of electrodes and cell design, as illustrated in Fig. 2. The studies primarily focus on improving desalination performance, advancing electrode and cell designs, process development, energy recovery, and characterizing FCDI as a reliable technology for desalination and resource recovery via the potential of ion selectivity. Since its inception by Jeon et al. in 2013 [21], FCDI has attracted significant academic interest. The diverse range of study areas, included optimizing cell configurations, components, and operational modes, cell architecture, electrodes/membranes, characterization/mechanisms, environmental applications, process and development, enhancing FCDI’s desalination

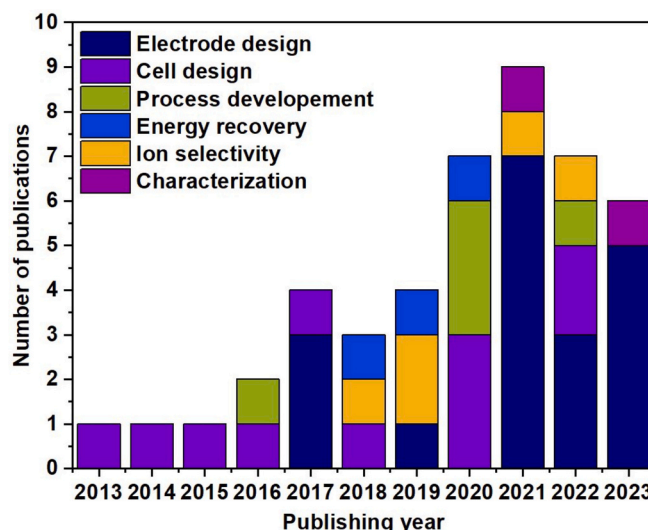


Fig. 2. Number of published papers on FCDI since 2013 categorized according to their main research topics. Data from the ISI Web of Science database.

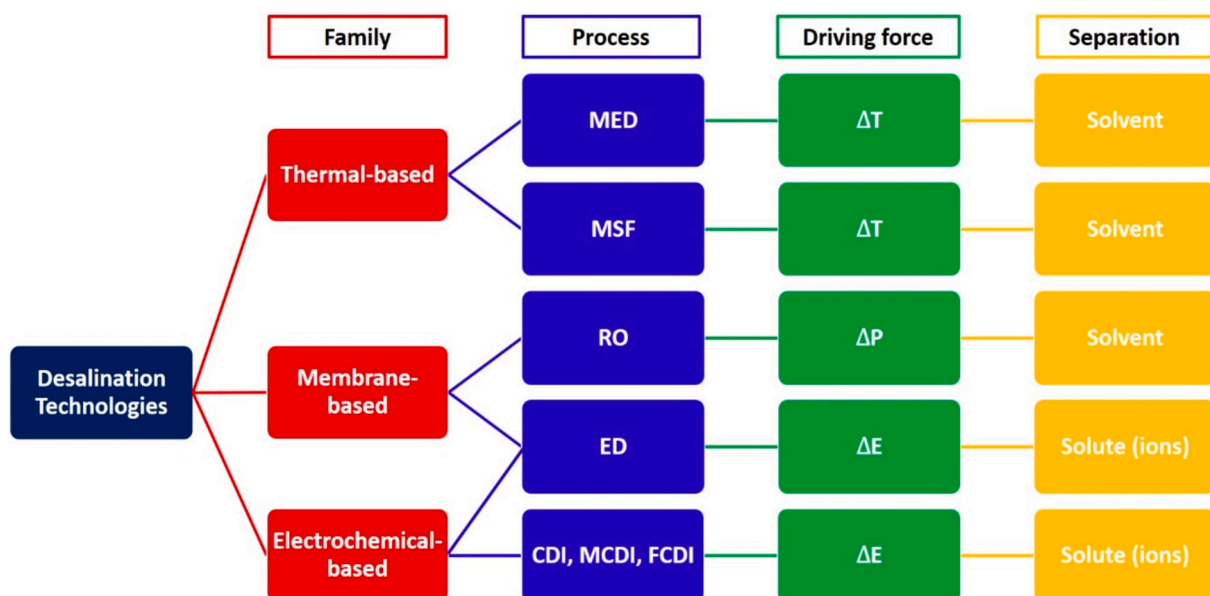


Fig. 1. Categorization of desalination technologies. Multi-effect desalination (MED); Multi-stage flash (MSF); Reverse osmosis (RO); Electrodialysis (ED); Capacitive deionization (CDI); Membrane-capacitive deionization (MCDI); Flow-electrode capacitive deionization (FCDI).

capabilities [22]. While desalination remains the prominent application area for FCDI, there is a growing number of studies since 2016, exploring its potential for resource recovery and contaminant removal through ion selectivity as shown in Fig. 2.

The principal objective of this review is to furnish an exhaustive exposition of FCDI technology, with specific emphasis on the following aspects: (i) Elaboration of FCDI design, encompassing cell architecture, electrode design, selection and arrangement of electrodes/membranes/current collectors, and operational procedures. (ii) Establishment of precise definitions and nomenclature for performance metrics associated with FCDI. (iii) Investigation of the role of feed saline solution and applied voltage in FCDI. (iv) Examination of the ion selectivity dimension within FCDI. (v) Exploration of prevailing and prospective applications of FCDI technology. (vi) Identification and assessment of potential challenges inherent to FCDI implementation. It is the aspiration of this review to expedite the seamless transition of this intriguing technology from laboratory experimentation to pragmatic real-world utilization.

2. An overview on the different capacitive deionization technologies: CDI, MCDI, and FCDI

The development of capacitive deionization (CDI) technologies dates back to the 1970s, when Johnson et al. first proposed the concept of using porous carbon electrodes for water desalination [28]. Early research primarily focused on demonstrating the feasibility of ion removal through this process (Johnson & Newman, 1971). However, practical applications were limited due to the lack of advanced electrode materials and the high energy costs of the process at the time. Significant progress occurred in the 1990s with the advent of modern carbon materials, such as activated carbon and carbon aerogels, which offered improved ion adsorption capacity and electrical conductivity [29]. In the CDI process, a small electrical potential difference (1 V to 1.4 V) is applied to the electrodes. This induces the migration of ions towards electrodes with opposite charges. These ions are then retained within the pores of the electrodes, as illustrated in Fig. 3. This adsorptive phenomenon persists until the pores of the electrodes reach their saturation point with ions. When an external direct voltage, typically below 1.4 V, is used, the electric field generated facilitates this electrostatic attraction and separation of ions. Upon reversing or discontinuing the external electric field, the electrodes undergo a short-circuiting process, inducing the release of ions from the interior of the electrode pores. This leads to the formation of a concentrated solution, as exemplified in Fig. 3. The desorption event enables the restoration of previously saturated electrode materials, rendering them available for subsequent operational cycles. Overall, CDI offers an energetically effective and

environmentally friendly approach to desalination, making it highly valuable for water treatment and related applications. CDI, the most conventional form of the technology presented in Fig. 4A, employs fixed carbon electrodes within the system. The integration of ion-exchange membranes in the 2000s marked a major milestone in CDI evolution, leading to the emergence of Membrane Capacitive Deionization (MCDI), which improved ion selectivity and reduced parasitic losses [30]. MCDI enhances ion control by integrating ion-exchange membranes (IEMs) between the stationary carbon electrodes and the spacer as described in Fig. 4B. These membranes enable the selective transport of the desired ions to the corresponding electrode, preventing co-ion adsorption and parasitic currents problems. Conventional CDI and MCDI operate intermittently, posing limitations on continuous functionality. In the past decade, Flow Capacitive Deionization (FCDI) has emerged as a transformative variant of CDI, utilizing flowable carbon slurries for continuous desalination and resource recovery. FCDI was introduced in 2013 by Jeon et al. [31] Their study presented a novel membrane capacitive deionization process utilizing flow-electrodes, enabling continuous desalination without the need for electrode regeneration. FCDI introduces a dynamic aspect by utilizing carbon electrodes in the form of a mobile slurry or suspension. The flowable electrodes composed of active materials, conductive additives, and electrolytes, enable both capacitive adsorption (where ions migrate under an electric field and get stored in oppositely charged electrodes) and electrolysis effects [23,32]. The carbon particles suspensions flow through specially designed channels or compartments carved on the current collectors as depicted in Fig. 4C. When an electric potential is applied to the FCDI cell, the charged carbon particles in the flow electrodes circulating through the channels, allow for continuous removal of ions. By continuously supplying the electrode chamber with regenerated carbon particle, FCDI achieves high desalination efficiency through continuous operation mode. FCDI offers the advantage of continuous desalination compared to both CDI and MCDI, since the desorption step to regenerate the electrode is completely dissociated to the desalination process. A comparative representation of the brackish water desalination character of diverse capacitive deionization configurations is represented in Table 1. In a common laboratory-scale FCDI system, the essential components include current collectors, separators or IEMs, spacers, and a pair of end plates that hold everything together. The spacer chamber, situated between the two IEMs, allows the feed water to flow through. By placing the IEMs on the current collectors, electrode chambers can be created, featuring flow-channels carved into them as shown in Fig. 4C. The flow-electrodes, which are responsible for ion removal, are typically recirculated between a reservoir and the electrode chambers using a pump (Fig. 4C).

Upon comparing the different capacitive deionization technologies

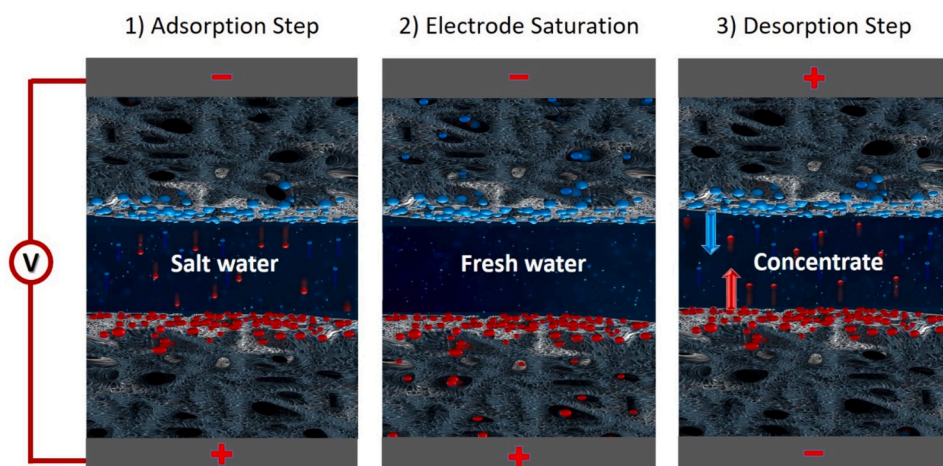


Fig. 3. Schematic representation on the different stages of the CDI process: Adsorption step, electrode saturation, and desorption step.

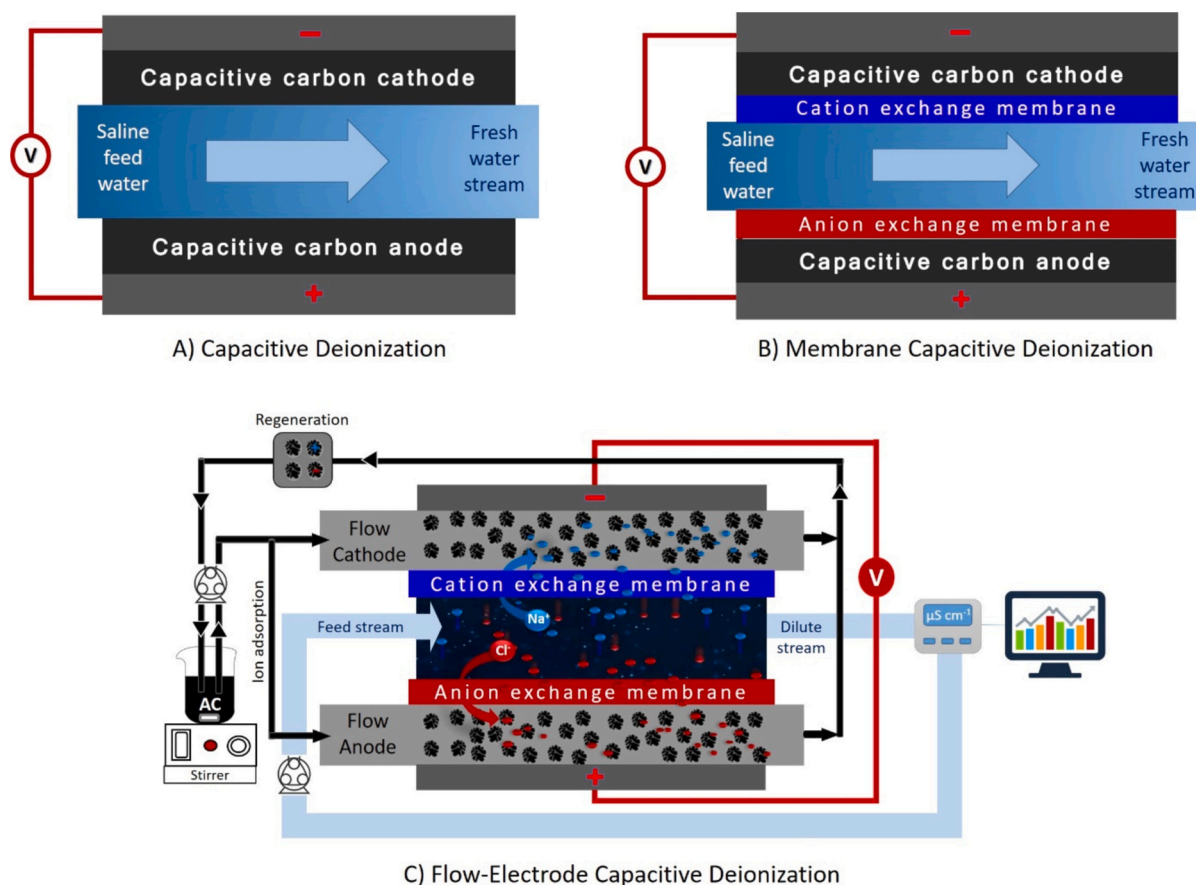


Fig. 4. Schematic illustrations of CDI (A), MCDI (B), and FCDI (C).

Table 1

Comparative representation of diverse capacitive deionization technologies efficiencies for brackish water desalination.

Type	Location	Capacity (m ³ /d)	Recovery (%)	Cell voltage (V)	SEC (kWh/m ³)	Membrane type	Ref
CDI	The Netherland	158	90	–	0.1–0.2	None	[41]
	Shandong, China	2400	69–75	1.5–2.0	1.33	None	[42]
	Shanghai, China	3600	78.5	1.5–2.0	0.55	None	[42]
	Taiwan	0.14	–	1.2	0.09	None	[43]
MCDI	Germany	0.14	–	1.0	0.29	None	[44]
	Taiwan	0.014	–	1.2	0.12	AEM/CEM (Beijing)	[45]
	The Netherland	0.043	50	1.4	0.17–3.45	Neosepta AMX/CMX	[46]
FCDI	Australia	4.3	46–87	0.4–0.8	0.28–0.37	Voltea AEM/CEM	[47]
	Korea	0.007	–	1.0	0.09	AEM/CEM	[48]
	Australia	0.0012	92	–	0.50–0.56	Fujifilm AEM/CEM	[49]
	Australia	0.0053	84.3	–	0.14–0.32	Fujifilm AEM/CEM	[50]

(CDI, MCDI, and FCDI), each has unique advantages and disadvantages. CDI is straightforward and cost-effective, but its intermittent operation limits scalability and efficiency for large-scale applications. MCDI addresses some of these limitations by integrating ion-exchange membranes, which enhance ion selectivity and reduce parasitic currents, but it comes at the expense of increased system complexity and cost. On the other hand, FCDI stands out for its ability to achieve continuous desalination and higher throughput by employing flowable electrodes. However, this comes with challenges related to maintaining the stability and uniformity of the slurry, as well as increased operational complexity due to the need for pumps and additional components. Therefore, the choice of technology depends on the specific requirements of the desalination application, such as scalability, cost, and operational continuity.

Among the different capacitive deionization variants, FCDI has garnered significant interest [21,33]. Its distinct dual-chamber configuration allows for exceptional flow efficiency of approximately 100 % as

the desalinated and brine streams are generated in separate chambers [34]. FCDI's manageable flow-electrode system presents opportunities for expanding its applications, such as resource concentration and recovery [25,35–38].

FCDI offers certain operational advantages over static capacitive deionization (CDI), particularly in terms of desalination efficiency. While it may not surpass reverse osmosis (RO) in overall energy consumption yet, FCDI's continuous flow system allows for more efficient ion removal over extended periods, reducing the downtime associated with electrode saturation in static CDI systems. This continuous operation mitigates the need for frequent regeneration of electrodes, enhancing energy efficiency compared to static CDI, where periodic halts for electrode regeneration increase energy consumption and reduce operational efficiency. Studies show that FCDI's flow-electrode design, where the carbon slurry continuously circulates, leads to higher salt removal rates and a more consistent desalination process

compared to static operation mode. This feature positions FCDI as a stronger competitor to other desalination technologies, particularly electro dialysis (ED), which also benefits from continuous ion removal but faces challenges when handling complex water chemistries [39]. In comparison to ED, FCDI has demonstrated potential in reducing energy consumption when dealing with brackish water. The ability to adjust the flow-electrode's composition and incorporate advanced ion-selective materials allows FCDI to target specific ions more effectively, leading to improved energy-normalized salt removal (ENRS). For instance, magnetic-assisted FCDI showed remarkable improvement in desalination performance, achieving a 78–205 % increase in the average salt removal rate (ASRR) compared to non-magnetic systems [39]. This design also yielded an energy-normalized removed salt (ENRS) of $8.77 \mu\text{mol J}^{-1}$, which can be competitive with RO but without the high pressure and membrane fouling issues associated. Further improvements in FCDI energy efficiency have been made through the optimization of ion exchange membranes (IEMs) and the introduction of dual-channel FCDI systems [40]. These systems enhance ion transport and minimize energy losses, particularly when dealing with feed water containing high concentrations of sodium chloride. In a dual-channel FCDI system, the arrangement of IEMs plays a crucial role in improving the current efficiency and reducing energy consumption. This setup has been shown to optimize the trade-off between desalination rate and energy consumption, addressing a key bottleneck in FCDI development.

Despite this, FCDI does not yet rival the lower energy footprints of large-scale RO systems, which benefit from advances in membrane technologies and energy recovery techniques. However, in scenarios requiring modular or decentralized water treatment, FCDI offers greater flexibility and operational efficiency compared to RO, which often requires significant infrastructure.

3. Ion storage mechanisms in capacitive deionization technologies

FCDI technology can involve both capacitive and Faradaic ion

storage, which is influenced by factors like electrode materials and the applied voltage. While capacitive ion storage is generally the dominant mechanism in capacitive deionization applications, there has been a noticeable rise in the occurrence of Faradaic ion storage in recent research within this field.

3.1. Capacitive ion storage

The capacitive deionization process capitalizes on electrical double layers (EDLs) for the transient adsorption of ions within the pores of the electrodes at the electrode-electrolyte interface. Hence, the mechanism of ion adsorption via the EDLs stands as an intrinsic cornerstone in capacitive deionization technologies. EDL was modelled for the first time in 1879 by Hermann Helmholtz who proposed the Helmholtz model. Later in 1910 and 1913, Louis Gouy and David Chapman respectively revised this model creating two EDL models that were later combined and widely adopted as Gouy–Chapman–Stern (GCS) model in 1924 [51]. Fig. 5 represents common EDL models, with a focus on a positively charged electrode. When an electrical driving force is applied, ions migrate towards electrodes with opposite polarities, resulting in the formation of EDLs on accessible surfaces. In accordance with the GCS (Gouy-Chapman-Stern) model, the EDL segregates into two distinct regions. The innermost layer, known as the Helmholtz layer, serves as the zone where ions of opposing charges accumulate in close proximity to the charged electrode's surface. Beyond this lies the diffusion region, referred to as the Gouy-Chapman layer, where ions disperse farther away from the electrode's surface. The distribution of electric charge within this layer is contingent upon the potential established at the electrode's surface. This electro sorption mechanism highlights the significance of materials with high surface areas, and well-developed porosity, facilitating efficient ion adsorption. One of the major advantages of CDI is the possibility of regenerating the electrodes (through physical desorption after inverting polarities) solely based on EDL formation, eliminating the need for chemical regeneration. Given that CDI predominantly utilizes microporous electrodes, the relevance of the GCS model diminishes. In response, the modified Donnan (mD) model, as

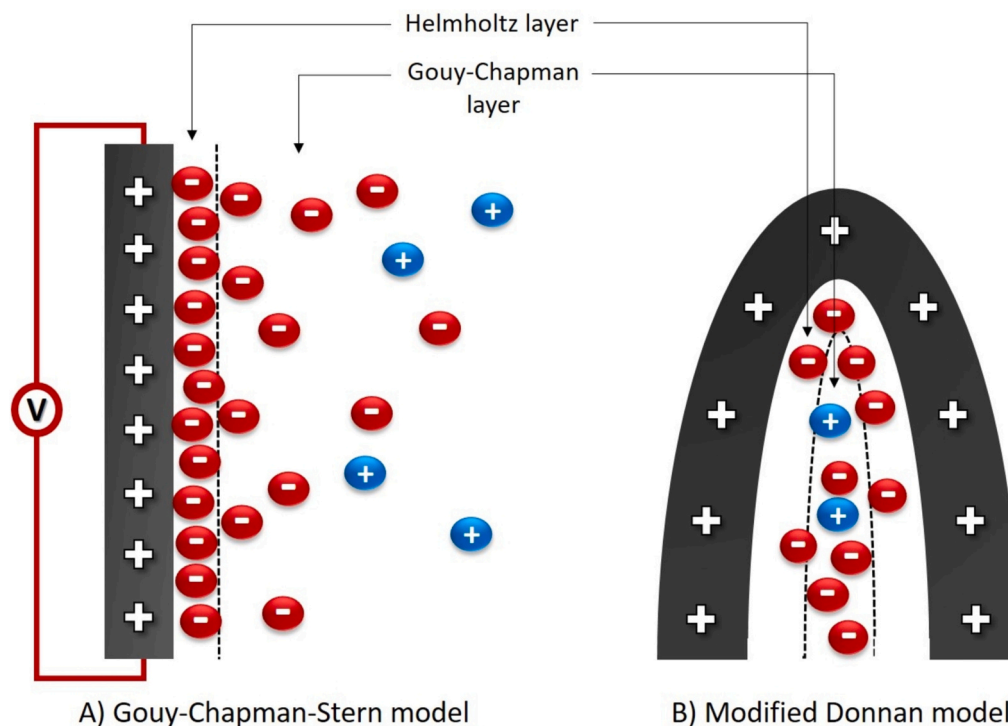


Fig. 5. The Gouy-Chapman-Stern model (A) and the Donnan modified model (B) both explaining the process of EDL formation and ion distribution on the electrode surface.

illustrated in Fig. 5B, becomes a pertinent framework. This model accommodates the potential for EDL overlap within finite pore structures, presuming a Debye length that exceeds the typical pore dimensions. Consequently, the mD model proves more apt for describing CDI processes involving microporous electrodes.

3.2. Pseudocapacitive or Faradaic ion storage

As previously discussed, capacitive ion adsorption takes place within the EDLs. On the other hand, an alternative approach called the desalting battery employs a different mechanism, where ions are absorbed through pseudocapacitive or Faradaic processes. By utilizing redox-active electrode materials for Faradaic ion storage, higher specific capacitance and deionization capacity can be achieved [52,53]. This enables the desalination of streams with higher salinity [54]. Pseudocapacitive mechanisms can be categorized into two types: redox pseudocapacitance, and intercalation pseudocapacitance. Redox pseudocapacitance surpasses mere surface adsorption by involving Faradaic ion storage through chemical reactions during the charging and discharging processes. By acting as a reservoir for both electrons and ions, the redox-active material greatly enhances the system's capacitance and overall deionization capacity [55,27]. The fundamental concept underpinning intercalation pseudocapacitance revolves around the incorporation of ions into the interlayer spaces or lattice structure of the electrode material. This process encompasses the introduction and removal of ions into and from the electrode, inducing volumetric expansion. Consequently, this expansion augments the available surface area for ion adsorption and desorption. This pivotal intercalation mechanism notably bolsters ion storage capacity by generating supplementary sites for ion retention. Consequently, the system's capacity is augmented, allowing it to accommodate a greater number of ions, which ultimately contributes to a remarkable improvement in the deionization system's overall performance [56].

Traditional electrode materials for pseudocapacitors include transition metal oxides (TMOs) such as ruthenium oxide (RuO_2), manganese oxide (MnO_2), and titanium oxide (TiO_2), MXenes, Prussian Blue Analogues (PBAs) [57], as well as conductive polymers like polyaniline and polypyrrole (as illustrated in Fig. 6) [53,58]. Conductive polymers, such as polypyrrole (PPy) and polyaniline, are crucial in advancing supercapacitor technologies due to their high pseudocapacitance and electrical conductivity. Polypyrrole, in particular, has demonstrated significant promise in improving the energy storage capacity of supercapacitors when combined with materials like carbon nanotubes or silver-incorporated composites. Electrochemical techniques, including cyclic voltammetry and electrochemical impedance spectroscopy (EIS), have been used to evaluate the performance of PPy when deposited on graphite electrodes, revealing its potential for high capacitance and excellent cyclic stability [59]. Further research by Gocki et al. showed that polypyrrole and its derivatives exhibit strong electrochemical properties, though challenges such as the durability of conductive layers on electrodes remain a limitation [60]. Additionally, Yuan et al. enhanced the performance of PPy combining it with materials like silver, which boosts charge transfer capabilities [61]. Moreover, studies have indicated that composites like PPy/single-walled carbon nanotubes offer improved capacitance by leveraging the strengths of both pseudocapacitive and double-layer mechanisms [62]. These findings emphasize the significant role of PPy in the development of supercapacitors, as it combines high energy density with good stability, making it a candidate for future advancements in energy storage.

On the other hand, Faradaic reactions can cause electrode degradation over time, reducing long-term durability and performance. Therefore, maintenance requirements are higher due to potential electrode degradation increasing operational costs. Complex and costly electrode materials are often necessary, additionally contributing to higher system costs. Large-scale implementation is challenging, limiting scalability. Nevertheless, ongoing research aims to address these issues and enhance

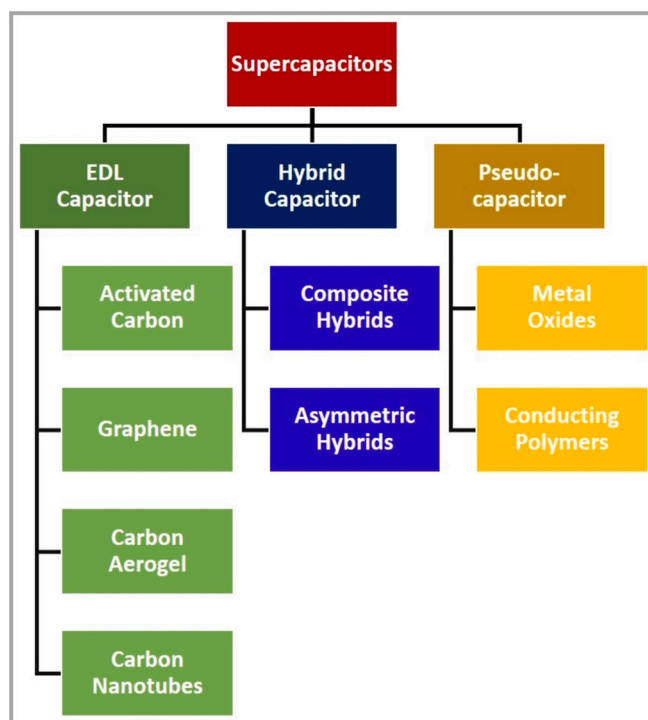


Fig. 6. A brief synopsis of supercapacitors and the electrode materials employed in each category.

the efficiency and performance of pseudocapacitive Faradaic CDI technologies.

3.3. Hybrid ion storage

A capacitor is an electronic component that stores and releases energy in the form of an electric field, typically used in electronic circuits for filtering or energy storage over short periods. A supercapacitor, stores much more energy than a regular capacitor due to its larger surface area and thinner dielectric, allowing it to bridge the gap between capacitors and batteries with fast charge/discharge cycles and higher energy density due to Faradaic reactions. Capacitive deionization systems can potentially utilize either capacitors or supercapacitors depending on their energy and performance requirements. A hybrid capacitive deionization (HCDI) cell incorporates both a capacitive electrode and a Faradaic electrode [63]. In this hybrid configuration, ion adsorption occurs simultaneously via capacitive mechanisms in one electrode and Faradaic mechanisms in the other integrating both the characteristics of electric double-layer capacitors (EDLCs) and pseudocapacitors. This offers enhanced energy and power densities. In these capacitors, one electrode is composed of a high surface area carbon material, while the other electrode is made from a pseudocapacitive material such as a metal oxide or conductive polymer (Fig. 6). By combining the advantages of both capacitive and Faradaic CDI systems, HCDI achieves remarkably high ion removal capacities and exhibits positive ion kinetics. This hybrid approach proves to be an innovative and effective solution for enhancing ion removal capabilities in deionization systems [63].

In summary, electrodes in capacitive deionization act as capacitors, storing energy through electrostatic charge separation. While traditional capacitors store energy through electrostatic charge separation, supercapacitors store energy via both electrostatic (double-layer capacitance) and electrochemical (pseudocapacitance) processes. In CDI, electrodes function similarly to supercapacitors by storing ions electrostatically, making it a specialized application of capacitive technology.

Capacitors are classified into two types: electrical double-layer

capacitors and pseudo-capacitors. EDLCs store charges through a non-faradaic process, while pseudo-capacitors use a combination of carbon material and transition metal oxides for a faradaic process with redox reactions. Supercapacitors exhibit significant capacitive capabilities, yet they encounter constraints linked to water electrolysis and electrode oxidation when subjected to elevated voltages, resulting in the generation of undesirable byproducts.

4. FCDI cell design

In the realm of FCDI research, the paramount importance of electrode and membrane designs is indisputable. Nevertheless, it is

imperative to recognize that system design holds substantial significance as well. Since the inception of FCDI in 2013, scholars have been committed to devising innovative cell arrangements. The primary goals of these advancements are to improve performance, streamline system management, and broaden the scope of applications for FCDI technology. In the subsequent section, we will explore some of the noteworthy breakthroughs in this area.

In the year 2013, Jeon et al. [21] introduced a significant innovation to capacitive deionization technology, by replacing the conventional stationary porous electrodes utilized in CDI cells with a dynamic carbon flow system. In this dynamic setup, carbon material circulates through specifically designed pathways on the current collectors, serving as the

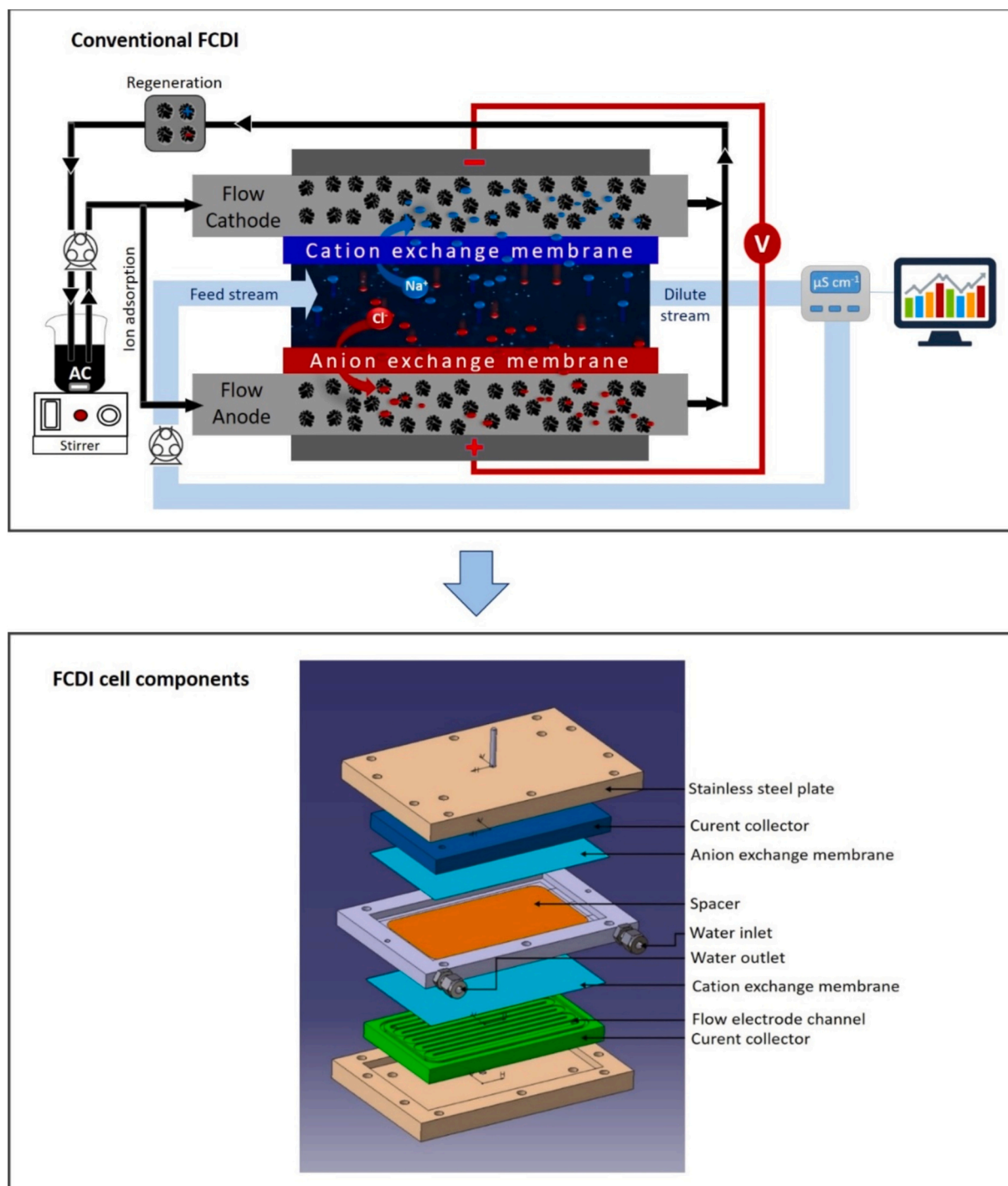


Fig. 7. diagrammatic portrayal of a standard laboratory-scale FCDI along with its key constituents.

flowing electrode. Fig. 7 illustrates the standard configuration of an FCDI module, comprising three compartments: two dedicated to housing the flow electrodes (one for the cathode and one for the anode), constructed from intricately sculpted current collectors positioned between endplates and ion-exchange membranes (IEMs). The third compartment accommodates a water chamber positioned between the areas occupied by the flow electrodes, incorporating a spacer located between the cationic membrane and the anionic membrane. Recent studies have explored the impact of the module's structure and material choices on the performance of FCDI cells. Herein, we delve into research advancements and the functions of each constituent within the FCDI system.

Current collectors are at the heart of the FCDI module, establishing potential disparities across the flow electrodes. The collectors' dimensions and material influence charge flow by managing the resistance. Common materials include graphite [64–66], epoxy-impregnated graphite [67], stainless steel [21], and gold-plated copper [68]. Carbon-based collectors are favored over stainless steel to prevent corrosion due to high Cl^- concentrations [67]. Innovative options include thin platinum wires within the IEMs and graphene layers connected to platinum wires, serving as current collectors [69].

The shape of the channels carved on the surface determines the contact surface area between the IEMs and the flow electrodes, which largely affects ions transport. Moreover, the size aspects - the length, width, and surface qualities - of these channels have a significant impact on both the performance and energy consumption. This effect is primarily influenced by the rheological properties of the carbon slurry. Usually, these channels are engraved on one side of the current collector. Nevertheless, it is important to highlight that a graphite current collector with dual-sided carving has also proven useful in FCDI cells stacking [66]. Innovative approaches, like Yang et al.'s use of a titanium mesh as a current collector, have been adopted. This mesh between the membrane and electrode flow channel shortens charge transport, boosting ion adsorption on carbon particles [70].

For the purpose of augmenting ion selectivity and mitigating co-ion migration, both the CEM and the AEM serve to isolate the spacer from the flow electrodes [71]. The resistance of these membranes is significantly influenced by their thickness and material composition. Additionally, a titanium mesh is interposed between the current collector and IEMs to enhance contact and safeguard against abrasion [72]. IEMs play an important role in ions selectivity in FCDI [19]. In this context, Zhang et al. demonstrated selective copper removal due to stronger attraction of Cu^{2+} ions to CEM compared to Na^+ ions [73]. Yang et al. demonstrated that the high ion exchange capacity and low electrical resistance of the ion exchange membranes (IEMs) were crucial in enabling efficient charge transfer from the spacer to the flow electrode, thereby improving desalination performance in FCDI [74]. They further demonstrated that higher crosslinking density increases endurance but raises electrical resistance [74].

The spacer, through which feed-water flows between the IEMs, affects FCDI performance by determining retention time. Recent materials and designs aim to optimize retention time and ion extraction efficiency. Materials such as nylon and polyester are mainly used in FCDI spacers [75,76]. A PU sponge spacer was employed by Choi et al. to extend the residence time of saline water, enhancing the effectiveness of ion extraction [77]. A porous spacer coated with CEM and AEM can be a solution to avoid costly freestanding IEMs [78]. While endplates do not impact FCDI performance directly, they affect module durability. Materials like polyvinyl chloride (PVC), polycarbonates and acrylic balance cost-effectiveness and robustness [21,64,66].

Since Jeon et al. transformed CDI cell architecture in 2013 by inventing FCDI, subsequent research focuses on refining each module component, aiming to enhance performance, durability, and cost-effectiveness of this technology. Scientists are as well exploring ways to improve performance and broaden the range of applications for FCDI technology by modifying its traditional setup. Subsequently, we will

examine diverse modifications that have been undertaken in the configuration of FCDI technology.

5. FCDI operational modes

This section delves into the examination of three distinct operational modes within the realm of FCDI: isolated closed-cycle (ICC), short-circuited closed-cycle (SCC), and single cycle (SC) modes [79].

In a typical ICC mode, a pair of separate flow-electrode streams circulate between their respective electrode chambers and an electrode reservoir chamber as depicted in Fig. 8A. The regeneration of electrodes and recovery of energy are achieved by periodically reversing electrode polarity under a constant current mode, similar to the intermittent process observed in conventional CDI [80]. By incorporating a secondary channel or module and manipulating the electrical circuit, advanced functionalities such as brine or nutrients separation and energy recovery can be attained [71,81,82].

Contrasting the ICC's complexity due to additional components, an alternative operational mode termed short-circuited closed-cycle (SCC) has been suggested [83]. In the SCC mode, shown in Fig. 8B, the two streams of electrodes converge in a shared reservoir chamber after departing from the electrode chambers [84]. Spent electrode particles spontaneously regenerate through charge neutralization during the mixing process, accompanied by the swift release of temporarily adsorbed ions into the electrolyte (as portrayed in Fig. 8B).

To further simplify the system, a new operational mode named single cycle (SC) was introduced by Ma et al. [85] In SC FCDI, unlike SCC operation, a singular closed flow loop is established, with flow-electrodes directly transitioning from one electrode chamber to the other as described in Fig. 8D.

Rommerskirchen et al. introduced a novel approach to supercapacitive flow-electrode capacitive deionization (SC FCDI) which entails a configuration where multiple discrete water compartments (e.g., dilute and brine compartments) are partitioned by an IEM [72]. In this arrangement, the flow-electrodes are systematically cycled between the electrode chambers. As described in Fig. 9, in one chamber (anode), ions (e.g., Cl^-) are extracted from the dilute compartment, adsorbed onto carbon particles, and subsequently desorbed in the opposing electrode chamber (cathode), accumulating in the brine chamber.

In comparison to the ICC mode where electrosorption and desorption happen consecutively, the SCC and SC modes enable timely desorption, renewing electrosorption capacity during desalination for enhanced performance and lower operating voltage. He et al. [80] demonstrated SCC mode's significantly reduced energy consumption (47 % of ICC mode) and lowered maximum voltage (1.0 V vs. 1.92 V) at the same current density. Ma et al. [85] found that the utilization of simultaneous charging and discharging in SC mode effectively taps into the accumulated electrostatic energy from desorption, resulting in decreased energy consumption. Similarly, Jeon et al. [86] employed a reverse series connection of two FCDI cells, similar to SC mode, where the positive electrode captures anions in the first cell, subsequently entering the negatively charged chamber of the second cell. Here, the anions are discharged and retained in the electrolyte, prompting increased cation migration into the anode chamber through charge neutralization.

In SCC or SC modes, ions gradually accumulate in the electrolyte, prompting the need for ion separation from flow-electrodes. SCC and SC modes offer significant advantages by effectively facilitating electrode regeneration, preventing ion migration to desalted water, and achieving high water recovery [87]. This approach overcomes the limited electrosorption capacity limitation of carbon particles, allowing desalination of both low and high concentration saltwater. Notably, Rommerskirchen et al. [88] accomplished successful desalination of high salinity water (60 g L^{-1} NaCl) using FCDI, achieving a water recovery of 77.6 %. It was considerably observed in the literature that operating FCDI in ICC mode triggers water dissociation or Faradaic reactions within the electrode chamber, leading to the production of OH^-

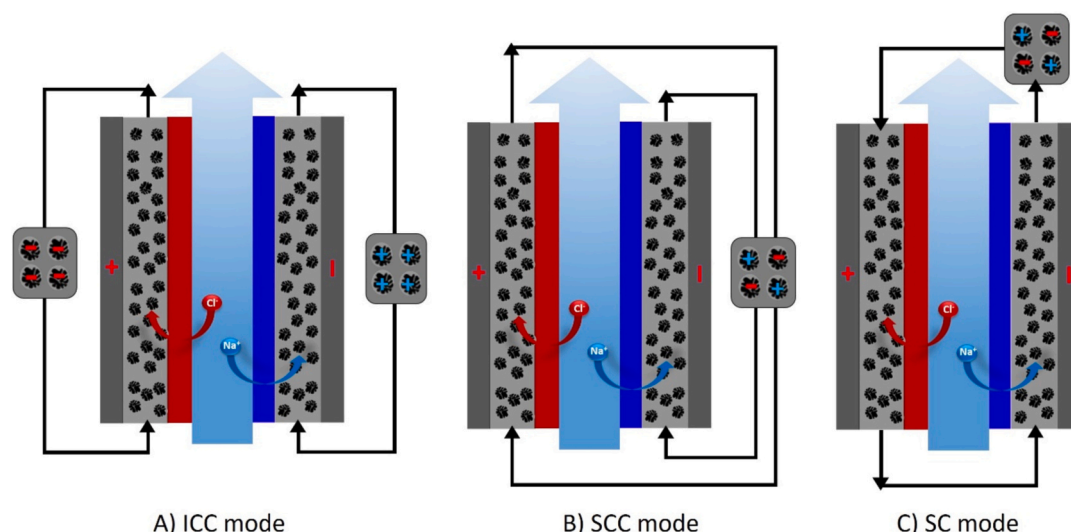


Fig. 8. Schematic diagram of (A) isolated closed cycle (ICC), (B) short-circuited closed-cycle (SCC), and (C) single cycle (SC) FCDI operational modes.

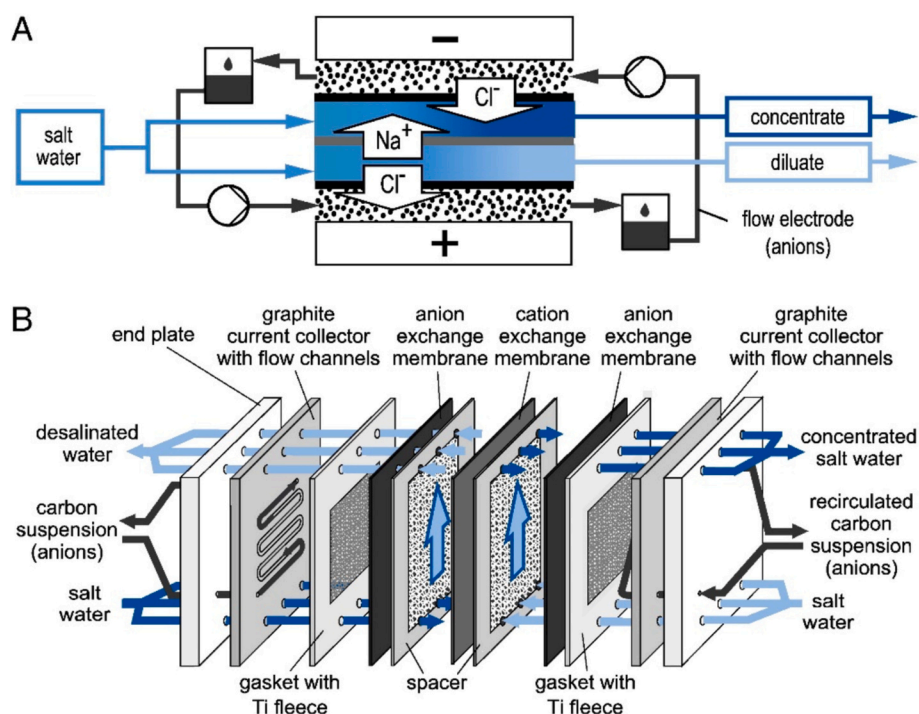


Fig. 9. The concept of single-module flow-electrode capacitive deionization (FCDI) is depicted in (A), while (B) illustrates the structure of the single FCDI module utilized in this study. Reproduced with permission from the reference ⁷².

and H^+ ions. This results in an increase in pH in the cathode chamber and a decrease in the anode chamber. A study conducted over an 11-h period using ICC mode reported significant changes in cathode pH (from 9 to 12.5) and anode pH (from 9 to 1.42) [32]. This pH shift in ICC mode can have advantageous effects, particularly for resource recovery. It can facilitate the conversion of certain substances, such as NH_4^+ to NH_3 in the cathode's alkaline solution, or the conversion of PO_4^{3-} to H_3PO_4 in the anode's acidic solution [26,89]. This conversion is beneficial for subsequent separation processes [26]. Zhang et al. utilized FCDI under ICC mode to effectively remove ammonium ions from dilute wastewater. By selectively concentrating ammonium ions in the cathode, they were transformed into dissolved NH_3 , which was subsequently stripped via a gas-permeable membrane and recovered as ammonium sulfate [89–91].

In summary, the operational modes of FCDI, including ICC, SCC, and SC, offer distinct advantages. ICC mode involves periodic electrode polarity reversal, while SCC and SC modes enable timely desorption, enhancing performance and water recovery. These modes facilitate electrode regeneration, ion separation, and resource recovery, making them promising for diverse applications, as demonstrated by various studies.

6. Flow electrodes in FCDI

The subsequent section delves into a crucial dimension of capacitive deionization, concentrating on the innovative and dynamic domain of flow-electrode systems. In the pursuit of augmenting the efficiency and efficacy of capacitive deionization processes, the incorporation of flow-

electrodes has surfaced as a promising avenue. This section systematically investigates the fundamental principles, progressions, and challenges associated with the application of flow-electrodes in capacitive deionization. As we traverse the complexities of flow-electrode systems, a holistic comprehension of their design, operation, and impact on deionization efficiency unfolds, offering valuable insights for both researchers and practitioners in the field.

6.1. Flow-electrode composition

Conventional CDI and MCDI systems primarily rely on capacitive processes to remove ions, but in FCDI systems, electro dialysis processes also have a significant impact. It is proposed that a portion of the ions transported to the electrode chambers in an FCDI system will remain in solution, leading to increased salinity of the electrolyte within those chambers [23]. The effectiveness of ion removal in FCDI depends on the properties of the flow-electrode. These properties determine the balance between electro dialytic and capacitive processes, thus influencing the ion removal efficacy. A flow electrode is generally composed of active material particles dispersed in a conductive liquid in the right proportions. The solid weight percentage (wt%) of flow electrodes is the proportion of solid materials present in the electrode mixture and is defined as

$$\text{Solid content (wt\%)} = \frac{\text{active material}}{\text{active material} + \text{electrolyte}}$$

Numerous studies have been conducted regarding the composition of flow-electrodes specifically for FCDI systems. In this section, we present a summary of the current approaches and materials utilized in the creation of high-performance flow-electrodes for FCDI application.

During the charging process, active materials play an important role both in electron transport and ion adsorption. In FCDI, carbon-based materials are widely employed as active materials, enabling ion storage EDLs [92].

Extensively studied as active materials for flowable electrodes, carbon-based materials such as AC [76,93], CA [94], CNTs [95–97], graphene [98–102], and MC [103] have garnered significant attention. These materials are favored due to their notable characteristics, including high specific surface area and porosity, good conductivity and ion mobility, high electrochemical stability, high capacitance, and cost-effectiveness. Furthermore, these carbon-based materials can undergo modifications through processes like chemical oxidation [104], nitrogen doping [105–107], or CO₂ activation [108,109]. Additionally, they can be coated with other components like TMOs, ion-selective polymers, or electron mediators [110–112]. These modifications bring about significant enhancements in the properties of flow-electrodes, such as improved wettability, increased capacitance, enhanced ion selectivity, and better suspension flowability and stability. Chemical oxidation of AC particles introduces oxygen-containing functional groups (COOH, C=O, C–OH), leading to an increase in carbon loading from 20 to 28 wt% and an increase in capacitance from 65 to 83 F g⁻¹ without compromising flowability and requiring additional energy [26]. Sulfonation of CNT-based flow electrodes reduces aggregation, facilitating effective contact between the active material and current collector, forming a conductive network [96].

Flow-electrodes used in FCDI usually have a mass loading of 5–20 wt% to achieve a balance between conductivity and viscosity. However, their conductivity is lower than static electrodes due to a high electrolyte content and a discontinuous electrical network [113]. Although increasing carbon loadings can enhance conductivity and performance, it comes with the risk of higher pumping energy and clogging [69]. Therefore, a carefully designed approach involving suitable active materials, conductive additives and supporting electrolytes is crucial for improving the performance of flowable electrodes.

Another important element in the composition of flow electrodes in FCDI is the conductive additive. Flow-electrode systems commonly use

highly conductive solids, including carbon black (CB), carbon nanotubes (CNTs), graphene/reduced graphene oxide (rGO), and molybdenum carbide (MXene).

CB is a popular conductive additive in both static and flowable electrodes, composed of dense carbon particles with high electrical conductivity but low specific surface area (<120 m² g⁻¹) [69]. Adding 1.5 wt% CB (average diameter: 12 μm) to flow-electrodes significantly boosts salt removal rate and charge efficiency in FCDI due to improved electron transfer and frequent collisions between AC particles and the current collector at higher loading [114]. It was found that graphite powder properties such as particle size distribution, particle shape, conductivity, and pores structure has an impact on FCDI performance when used as a conductive additive in the flow-electrodes [74]. CNTs act as a suspension bridge, enhancing connectivity and conductivity, doubling the salt removal rate with just 0.25 wt% addition [115]. rGO sheets also serve as effective conductive additives, facilitating rapid electron transport within the carbon slurry network [116]. While Mxene is effective for fixed electrodes, its use in flow-electrodes is yet unexplored [117–119]. Managing solid conductive additives is crucial to achieve high conductivity without particle aggregation and clogging [120].

In addition to the active materials and the conductive additives, the electrolyte in a crucial component of the flow-electrode. Flow-electrodes mainly consist of the electrolyte, comprising over 80 wt%. Introducing suitable salts like NaCl or Na₂SO₄ to the electrolyte can boost FCDI desalination efficiency (up to 25%) and decrease cell resistance [114]. However, altering the electrolyte salinity involves a trade-off, potentially affecting flow-electrode performance and slurry stability. Higher salinity levels might result in particle aggregation, increased slurry viscosity, reduced suspension stability, and higher energy consumption for electrode recycling [69]. Concerns also arise regarding counter-ion back diffusion, co-ion leakage, and water transport at higher salinities, which could compromise Coulombic efficiency and desalination performance [68,69].

6.2. Electric charge transport and resistance

FCDI relies on charge transport in the flow-electrode, involving crucial roles for both electrons and ions (Fig. 10). Electrons move

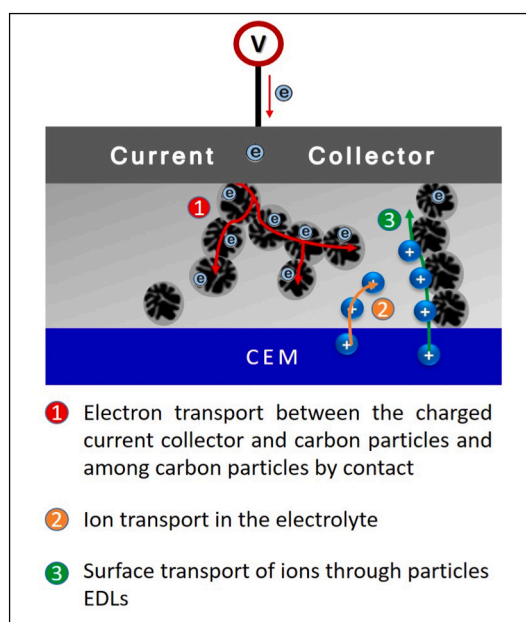


Fig. 10. Schematic diagram depicting the primary charge (electrons and ions) transport mechanisms in flow-electrodes in FCDI.

through direct contact between the current collector and activated carbon particles, as well as among the activated carbon particles themselves, forming a continuous network (indicated as 1 in Fig. 10). Additionally, redox reactions if present (pseudocapacitive FCDI) between activated carbon particles facilitate electron movement. Meanwhile, ions traverse through the electrolyte and EDLs of the particles tangentially to surfaces (marked as 2 and 3 in Fig. 10) [121].

A crucial aspect of optimizing FCDI performance is understanding and quantifying charge transport in flow-electrodes. The physical contact between particles significantly influences electron percolation within the flow-electrodes. Studies utilizing synchrotron radiation X-ray tomographic microscopy have unveiled percolating clusters based on solid content loading [122]. Furthermore, 3D reconstructed images provide valuable insights into percolation and interparticle voids, demonstrating that higher carbon content enhances the connected particulate network, leading to anisotropic electron transport and intricate ion transport paths [123]. Despite these valuable micron-level insights, challenges remain in measuring flow conditions in real application scenarios.

Electrochemical tests are employed to comprehend flow-electrodes by determining their resistance and capacitance. These in situ assessments enable a quantitative examination of charge transport mechanisms. The evaluations include various techniques such as electrochemical impedance spectroscopy (EIS) and direct current (DC) measurements. Researchers have previously used DC tests to obtain electron transport resistance [122,124]. EIS is particularly common, utilizing a sinusoidal signal with small amplitude to linearize the electrochemical system. The system's resistance and capacitance are then determined using an equivalent linear circuit model comprising interconnected circuit elements.

Further advancements are required to enhance the precision of charge transport simulation, with particular attention to ion transport. Back in the 1960s, a theoretical transmission line model for porous electrodes was introduced, facilitating the investigation of dynamic charge transport in fluidized bed electrodes operating under flowing conditions [125,126]. Drawing on this transmission line model and prior research on the electrode of an electrochemical flow capacitor, scientists have developed an FCDI-tailored equivalent circuit [76,127,128]. Within this circuit, the flow-electrodes carry both electrons and ions, and their respective currents are interconnected through a double layer capacitance (Fig. 11). The primary objective of this equivalent circuit represented in Fig. 11 is to better characterize the resistances in FCDI [128]. Specifically, R_s represents the series resistance, which encompasses the resistance of IEMs and the ionic resistance in the desalination chamber. Additionally, R_o corresponds to the resistance present in external circuit components, like current collectors. The contact resistance, R_{ct} , arises between carbon particles in the flow-electrodes and the current collector. Furthermore, the constant phase element, Q_{ct} , runs in parallel with R_{ct} , signifying the interfacial contact capacitance of current

collectors. Moreover, R_e/n , R_{ion}/n , and C/n denote the electronic resistance, ionic resistance, and EDL capacitance of the carbon particles in the flow-electrodes per unit volume/length, respectively, with 'n' being a discretization parameter dependent on the specific test system and computational requirements.

In the context of FCDI, the predominant resistance governing the flow-electrode primarily pertains to electronic resistance, which significantly surpasses the ionic resistance [128]. This finding suggests that electron transport plays a crucial role in limiting the charge transport within the electrode. Furthermore, the resistance is significantly affected by factors such as the AC content, salt concentration of the electrolyte, and flow rate, making them important parameters to consider.

The impact of augmented carbon content within flow-electrodes on different types of resistances was investigated. Notably, electronic resistance exhibits greater sensitivity to carbon content compared to ionic resistance [122]. When the carbon content is raised from 10 % to 20 %, the electronic resistance undergoes a substantial reduction by an order of magnitude [129]. This decline is likely due to the larger average size of the percolating cluster resulting from higher carbon loading, which promotes electron transport [122]. Similarly, the ionic resistance experiences a significant decrease from 7 Ω to 2.4 Ω as the carbon content increases from 5 % to 20 % [124]. These findings suggest that an increased concentration of solid carbon particles enhances charge transport. Ionic resistance was quantified DC tests [130]. The results reaffirm that flow-electrodes with higher carbon content exhibit reduced ionic resistance. This reduction is attributed to the short-circuiting of the ion conduction path through the particles. Nevertheless, it is noteworthy that an escalation in carbon content leads to an elevation in contact resistance (R_{ct}) [128].

Furthermore, the electrolyte concentration influences the resistance within the flow-electrodes. Increasing the concentration of electrolyte from 1 g L⁻¹ to 35 g L⁻¹ leads to a significant decrease in ionic resistance, reducing it from 7 Ω to 0.5 Ω . In contrast, the electronic resistance initially experiences a substantial drop as the salt concentration increases. However, this stabilization occurs once the salt concentration attains a specific threshold. As the ionic concentration of the electrolyte rises, the electrical double layers (EDLs) tend to compress, enveloping the carbon surfaces. As a result, the average distance between particles decreases leading to a more efficient path for electron transport between the particles [124,131,132].

Additionally, the electronic resistance exhibits a significant decrease at higher flow rates [133]. Higher flow rates promote flow disturbances, facilitating better contact between particles and thus reducing resistance. However, in a broader range of tests, it has been observed that resistance first decreases and then increases with faster flow rates applied [72,127]. This behavior is likely due to the influence of boundary layer effects.

In FCDI, electronic resistance plays a dominant role in controlling the flow-electrode's behavior, surpassing the influence of ionic resistance.

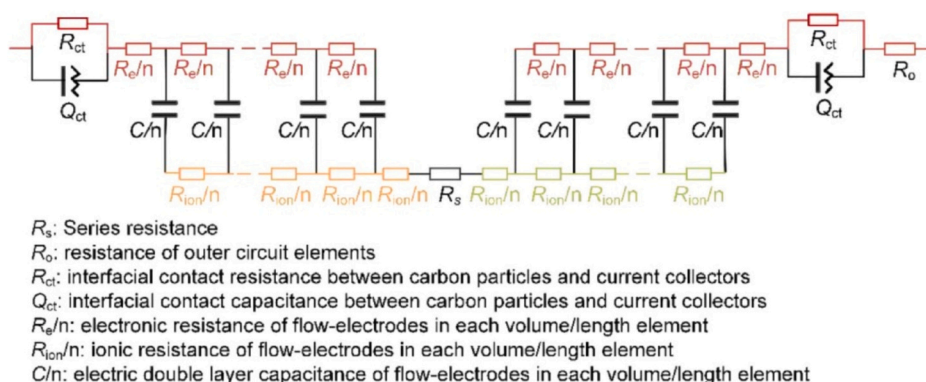


Fig. 11. Schematic showing an equivalent circuit of an FCDI cell based on a theoretical transmission line. Image taken from reference [128].

AC content, electrolyte concentration, and flow rate are critical factors affecting resistances and consequently impacting the system's overall performance. Higher carbon content improves both electron and ion transport, while increased electrolyte concentration reduces ionic resistance. Flow rates influence electronic resistance with intricate behavior.

6.3. The flow-electrode impact on the FCDI performance

The FCDI technology sets itself apart from other CDI methods due to its unique flowable characteristic, which simplifies the management of electrodes [104]. The ideal flow electrode for FCDI must possess both low resistance and high capacitance, making it an essential foundational component [65]. This electrode plays an indispensable foundational role within the FCDI cell, enabling the transport of ions from the spacer to the current collector. Composed of components like carbon slurry, electrolytes, and conductive agents, the rheological and electrochemical properties of the flow electrode depend on these constituents [134].

The transition from stationary to flow electrodes is designed to obviate the need for distinct desalination and regeneration phases. However, this transition introduces new avenues for charge transfer, including current collector to particles charge transfer, direct charge transfer, charge percolation, diffusion, convection, interactions occurring within the electrical double layer, and migration across ion-exchange membranes [135]. Unfortunately, these novel pathways can result in charge losses and less-than-optimal transfer efficiency, leading to a decrease in electrical conductivity to levels around $0.1\text{--}1\text{ mS cm}^{-2}$ [69,134,136]. In order to enhance the performance of FCDI cells, researchers continually concentrate on refining the composition and flow rate of the flow electrode to enhance charge transfer processes.

6.3.1. Influence of carbon in the flow electrode

Important factors affecting the flow electrode performance in FCDI include the weight percentage, the particle size and dimension [137], the chemical surface charge [138], and the morphology [76,139,140] of activated carbon within the slurry [8]. In contrast to traditional stationary electrode CDI where porous carbon content ranges from 85 % to 95 %, FCDI limits AC content to 5 % to 35 % due to hydrophobic interactions causing potential clogging [76]. Research by Rommerskirchen et al. [135] highlights the positive impact of mesopores on desalination performance, attributed to enhanced ion transport and greater outer particle dimensions relative to microporous structures. Tang et al. [141] explored metallic CNTs as a carbon source, yielding significantly higher salt removal capacity and charge efficiency compared to AC. However, their high cost and limited loading make their exclusive use unefficient [142]. A higher carbon content improves the charge transfer in the flow electrode, promoting efficient electron transfer through a 3D particle network [69,143]. However, excessive carbon content beyond 20 % raises viscosity and poses flowability problems, limiting practical use to around 20 % [64,69,122]. Researchers have also pursued surface modifications to boost carbon content. Hatzell et al. [104] achieved a 40 % carbon content increase (from 20 wt% to 28 wt%) through AC oxidation, reducing viscosity and pumping energy by 50 % while increasing electrical energy due to elevated operating voltage. Amination and sulfonation of AC particles reduced the viscosity of the slurry electrodes which allowed for a 35 % increase in carbon mass loading within the flow electrode [76]. While the surface modification resulted in reduced salt removal efficiency for activated carbon (AC) particles within the flow electrode, the elevated carbon loading contributed to an overall improvement in salt removal efficiency, elevating it from 8 % to 28 %. Xu et al. [144] combined MnO_2 , AC, and carbon black to create an asymmetric FCDI, broadening the potential window and enhancing desalination efficiency from 59 % to 78 %. The hydrophilic nature of AC/ MnO_2 could contribute to this improvement. In conclusion, to achieve peak performance in FCDI, it is crucial to consider and optimize parameters related to the carbon slurry,

such as activated carbon surface characteristics, outer particle diameter, material loading, viscosity, and suspension stability.

6.3.2. Influence of conductive additives in the flow electrodes

The process of AC surface modification involves additional labor and time. The application of polymer coatings for surface modification leads to a reduction in the effective surface area of activated carbon (AC) as a result of penetration into the internal pores, consequently diminishing the desalination performance [115]. Researchers primarily focus on improving the electrical conductivity within the flow electrode by enhancing the carbon weight percentage (CWP). To enhance electrical conductivity, one strategy is to introduce conductive additives that have a lower surface area but high electrical conductivity, facilitating efficient charge transfer within the flow electrode. In standard FCDI systems, electric double layers (EDLs) primarily form on activated carbon (AC) particles close to the current collector, leaving many AC particles that are not in contact with the collector in a neutral charge state due to limited charge percolation. Adding conductive materials like carbon black creates pathways for charge percolation among AC particles, as shown in Fig. 12, thereby promoting greater EDL formation [115]. This process enhances electronic charge transfer, thereby decreasing resistance within the flow electrode and leading to better desalination performance. Charge transfer primarily occurs due to particle collisions, influenced by Brownian motion as particles move within the flow channel. In an AC-only slurry, electronic charge transfer involves two steps: AC particles near the current collector collide with the collector first, then transfer the charge to another AC particle, as illustrated in Fig. 10. Introducing conductive additives changes this to a multi-step process, where collisions occur between the current collector and conductive additive particles, or between AC particles carrying charge to the conductive additive particles and then to other AC particles. The high electrical conductivity of these additives ensures efficient charge transfer during these collisions [114].

CB is a commonly used conductive additive in recent studies, though its hydrophobic surface can lead to flow channel clogging. Yang et al. [74] showed that the addition of CB to AC-based flow electrodes increased desalination performance by 1.5 times. However, clogging issues arose after 40 min due to CB aggregation caused by changes in surface charge. Minimizing clogging concerns can be addressed by using micro-sized or smaller conductive additives. Commercial conductive additives were compared based on their properties (surface area,

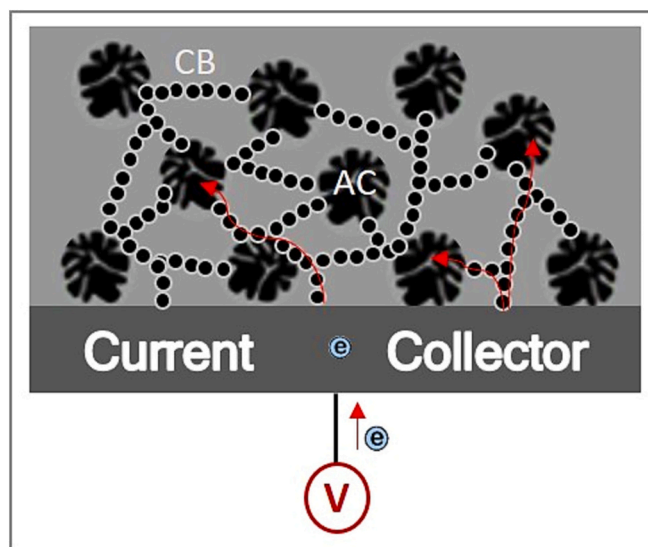


Fig. 12. Illustrative depiction of how carbon black functions as a conductive additive within flow-electrode systems. CB = Carbon Black; AC = Activated Carbon; e = electron.

powder conductivity, shape, and size), revealing that plate-shaped ones exhibited higher desalination efficiency and capacitance than spherical ones, likely due to better electron transfer [65,74]. CB also enhanced desalination rate at low electrolyte concentration but not at high concentration due to electrolyte diffusion [114].

Functionalized CNTs were introduced to improve aqueous dispersion and serve as conducting agents for bridging AC particles [82]. The introduction of functionalized carbon nanotubes (FCNTs) increased the desalination and current efficiencies. Although augmenting desalination performance by combining conductive additives with modified AC is possible, it necessitates cautious optimization owing to cost factors and the potential risk of channel clogging with slightly elevated carbon weight percentages.

6.3.3. Influence of the electrolyte in the flow electrode

Aiming to improve the efficiency of FCEDI systems, many research groups are developing active material suspensions with different electrolyte concentrations. Yang et al. [65] conducted an investigation into the influence of different electrolyte concentrations, specifically sodium chloride (NaCl), on the desalination efficiency of FCEDI cells. In cases where the flow electrode was devoid of electrolyte, ion diffusion exhibited inefficiency due to the extended path ions needed to traverse from the membrane to reach the activated carbon (AC) surfaces. The introduction of electrolyte, on the other hand, served to reduce the ion diffusion distance, thus facilitating charge transport. In the presence of electrolyte, migrating ions from across the membrane did not exclusively adhere to the AC surface; instead, they joined ions already present within the flow electrode. The impact of varying electrolyte concentrations within the flow electrode on desalination efficiency shows that the length of the diffusion path is less significant. Electrodes with higher electrolyte concentrations facilitate ion diffusion into the intra-pores of activated carbon (AC) compared to those with lower concentrations. Electrolytes with lower concentrations reduce ionic resistance and enhance the capacitance of AC particles, whereas higher concentrations further increase capacitance through enhanced ion diffusion into the intra-pores of AC [65,82,114]. However, an excessive ionic concentration in the electrolytes elevated polarization resistance, leading to the reverse diffusion of co-ions (leakage) [23]. Higher electrolyte concentrations resulted in a less viscous slurry, which in turn improved desalination performance [145]. Liang et al. [114] investigated varying electrolyte concentrations and voltages, finding that at lower voltages, higher electrolyte concentrations negatively impacted desalination performance due to dominant electrophoresis driven by ion concentration gradients causing high osmotic pressure. Elevated salt concentrations in the flow electrode were found to enhance energy recovery by reducing voltage drop and extending the discharging period due to lower resistance [82]. Moreno et al. [68] explored the effects of ionic concentration differences between the electrode and feed solution on ion movement, analyzing both diffusion and migration using the diffusion migration ratio (DMR). They found that higher concentration gradients across ion-exchange membranes (IEMs) increased ion diffusion, significantly influencing the regeneration capability and energy recovery of the flow electrode during regeneration. High concentration gradients during the charging phase resulted in a predominantly diffusion-driven process, which could not be entirely reversed by adjusting the electrode potential.

Recent studies emphasize that redox-active electrolytes like ferri-ferrocyanide and iron-based additives enhance the ion removal rate by improving charge transfer and electrode kinetics. For instance, Freire et al. [146] explored iron chloride and iron citrate as low-cost, environmentally friendly redox additives in FCEDI, noting a significant improvement in desalination rates with iron chloride outperforming other additives by doubling the salt removal rate. The composition of the electrolyte also influences pH stability and ion crossover, with iron citrate maintaining a neutral pH, which is preferable for potable water applications. Similarly, Mani et al. [147] investigated the use of ferri-

ferrocyanide redox couples in FCEDI and demonstrated an optimal electrolyte composition that significantly improved salt removal efficiency while minimizing energy consumption. Furthermore, Uyanik et al. [40] showed that the electrolyte's ionic composition and its interaction with ion exchange membranes influence ion selectivity and energy efficiency in dual-channel FCEDI setups, highlighting the importance of balancing electrolyte composition for enhanced performance. In summary, the electrolyte's properties, including redox capability, ionic strength, and pH stability, are key determinants of the operational efficiency and scalability of FCEDI systems.

6.3.4. Influence of the electrode flow rate

The flow rate of the feed electrode plays a substantial role in determining the deionization performance of the FCEDI module due to the dynamic interactions between particles, which result in varying charge percolation [8]. Rommerskirchen et al. [135] examined the influence of electrode flow rate on charge transport within the flow electrode, employing electrochemical impedance spectroscopy (EIS) for analysis. The effectiveness of deionization shows an improvement as the flow rate increases, up to a specific threshold, beyond which it starts to decline. This behavior is likely attributed to increased charge transfer (both to particles and the current collector) resulting from enhanced contact between them at higher flow rates. However, when flow rates become excessively high, charged particles are forcibly expelled from the FCEDI cell due to charge convection, leading to a decrease in deionization efficiency. Typically, the flow rate of the electrode is much higher than that of the feed water, which can further degrade performance. For instance, Fang et al. [24] noted a decline in ammonia removal efficiency when the electrode flow rate exceeded that of the feed solution, primarily due to the elongated ion diffusion pathways. It's worth mentioning that the energy requirement for pumping significantly increases with higher electrode flow rates. Consequently, while increasing the flow rate enhances desalination speed, it may not be the most energy-efficient strategy. Conversely, it was noted that the pumping energy needed for two distinct AC concentrations was identical. This implies that optimizing the AC concentration represents a more practical approach to achieving efficient desalination compared to merely increasing the flow rate. Recent research shows that increasing flow rates enhances the salt removal efficiency by promoting better ion transport and minimizing concentration polarization. For instance, Mani et al. [147] demonstrated that higher flow rates in the electrode chamber boosted the salt removal rate to $43.92 \mu\text{g cm}^2 \text{s}$, achieving a remarkable 76.38 % salt removal efficiency while optimizing energy consumption at 0.27 kWh/mol. Similarly, Shi et al. [148] explored how increasing the flow rate reduced energy consumption, particularly in the electrode compartment, where optimization could result in a 51.9 % reduction in overall energy use. Flow rate optimization not only improves ion adsorption kinetics but also reduces the resistances within the electrode slurry, as confirmed by Wang et al. [149] through their film-electrode model for FCEDI, where higher flow rates enhanced the overall deionization process by improving energy and desalination performance. Consequently, adjusting flow rates remains a key operational parameter for maximizing the efficiency of FCEDI systems while balancing energy use and desalination output.

6.4. Flow-electrode materials in FCEDI

Carbon materials play a crucial role as electrode sources in CDI, MCDI, and FCEDI due to their beneficial properties, including large specific surface area, high porosity, and cost-effectiveness. Although activated carbon (AC) [76,93,150–152] is the most commonly used electrode material in FCEDI, researchers have also explored other carbon forms like graphene [98,99,101,102], carbon aerogel (CA) [94], carbon nanotubes (CNTs) [96,115], carbon black (CB) [85,114,153,154], and others, gaining significant attention in the FCEDI field. Advanced synthesis and design strategies, such as microwave/ultrasound-assisted

hydrothermal methods, electrospinning, 3D printing, porous templates, surface modifications, and chemical activation, have resulted in improved carbon materials [155–159]. These improvements encompass a substantial specific surface area, a well-distributed hierarchical pore structure, excellent electrical conductivity, optimized wettability, and enhanced ion-selectivity. These factors collectively enhance the salt adsorption capacity and charge efficiency in CDI.

Additionally, Faradaic electrodes have attracted interest in desalination due to their superior ion storage, high selectivity, and reduced co-ion adsorption impact. This Faradaic approach opens new possibilities for enhancing desalination performance in high salinity water [160,161].

Researchers continue to strive for increased desalination capacity in both capacitive and Faradaic CDI electrodes, driving material development efforts. Recent advancements in electrode properties are highlighted in this section, aiming to enhance overall desalination performance. Various materials used in capacitive and faradaic electrodes are presented, drawing on published literature. Fig. 6 provides an overview of the diverse types of materials used as EDL capacitor, pseudocapacitor, and hybrid systems in CDI.

6.4.1. Capacitive carbon electrodes for EDL capacitors FCDI

The EDL capacitor operates through electrosorption on porous carbon-based electrodes, serving as an ion storage system in FCDI. Carbon-based electrodes are highly favored in FCDI due to their cost-effectiveness, corrosion resistance, high surface area, high electrical conductivity, and stability. Various carbon materials, such as activated carbon, carbon nanotubes, mesoporous carbons, carbon aerogel, and others, have been utilized as electrodes in FCDI.

This section highlights recent research efforts, techniques, and advancements related to selected electrodes based on findings from the literature. The goal is to provide valuable insights into ongoing efforts to improve the performance and capabilities of FCDI electrodes.

i. Activated carbon (AC)

The passage discusses the use of AC in FCDI and various applied strategies to enhance its performance. AC produced with different methods is favored for FCDI due to its large specific surface area (1000–2000 m² g⁻¹), well developed porosity, low cost, stability, and scalability. However, its limited hydrophilicity can be improved by incorporating hydrophilic groups on its surface.

In the first FCDI desalination study by Jeon et al., AC was employed as the flow-electrode [21]. Since then, AC caught attention for its high salt removal efficiency in continuous desalination. Flowability and rheological properties are essential factors in FCDI [69]. Preventing clogging in narrow channels is a significant concern addressed by the rheological property of the electrode suspension [76,162]. AC is typically used in flow-electrodes with a range of 5–35 wt% [64]. An increased carbon loading enhances conductivity, leading to higher current and improved salt removal. However, this also results in clogging problems due to the thick texture and particles sticking together and higher viscosity, necessitating more energy for pumping [64]. To tackle this issue, scientists have tried different methods, such as adding specific groups like nitrogen [163], ammonium [64], sulfonate [64], and oxygen-rich groups [101,104], on the surface of AC materials, improving its flowing properties, dispersion, and electrical conductivity. By using an oxidized active material, they achieved a higher mass density without increasing the energy requirements for pumping. Additionally, the rheology of carbon slurries can be significantly altered by surface heteroatoms. Functionalizing AC facilitates better particle separation, leading to improved flowability and dispersion with reduced aggregation [104]. In specific flow electrode applications, researchers have applied coatings of ionic head-groups to AC suspensions. This approach resulted in decreased viscosity, especially with a high carbon loading, as the ionic functional groups on the AC surface induced

electrostatic repulsion, promoting a desirable dispersion of AC particles without aggregation [76]. In this context, Park et al. modified the surface of a spherical activated carbon with trimethylammonium and sulfonate groups [64]. This allowed for an increase in AC loading to 35 wt% and simultaneously decreased the material viscosity. This substantial boost in carbon concentration enhanced the desalination efficiency by a 3.5-fold compared to the standard 10 wt% configuration. Similarly, Huang et al. enhanced the dispersion of AC through the synthesis of an AC, 0.25 wt% sulfosuccinic (SSA), and 0.13 wt% polyetherimide (PEI) composite via a straightforward mixing approach [164]. This composite, in contrast to chemical modification, yielded a noteworthy threefold improvement in salt removal performance compared to pristine AC slurry electrodes. The as prepared composite slurry electrode also exhibited robust cycling stability over 1000 cycles. Also on the same line, Hatzell et al. proposed an effective method to enhance the rheological characteristics of AC via strong surface acid treatment, resulting in oxidized carbon [104]. This technique introduces hydrophilic functional groups like hydroxyl, carboxyl, and carbonyl, enabling a substantial 40 % increase in mass loading within the flow-electrode. Consequently, this modification led to 25 % rise in specific gravimetric capacitance, contributing to elevated desalination efficiency while reducing energy consumption.

Additionally, AC particles play a pivotal role in charge (ion and electron) transport and resistance within flow electrodes in FCDI. Their arrangement influences ion adsorption, desorption dynamics, and electron transfer, significantly affecting the overall system efficiency. To enhance ion adsorption and reduce resistance within the inner AC flow-electrode, Lim et al. recently presented an approach integrating biochar and AC for desalination, and heavy ions removal [165]. Due to the increased electrical efficiency and the porous properties of carbon-rich material, this integration yields a significant 1.83-fold enhancement in desalination performance, surpassing the results achieved using only an AC electrode. Moreover, for the purpose of augmenting interparticle contact and mitigating Ohmic resistance in fluidized electrodes, Ma et al. integrated AC with magnetic material (MAC) [166]. No improvement in the salt removal rate was observed with the resulting MAC composite within the slurry electrode. However, MAC composite showcased significantly higher charge and energy efficiency (>100 %) in comparison to pristine AC of equivalent mass loading. Remarkably, the MAC-enhanced flow-electrode also extended the electrode's operational lifespan by a factor of 64.5. Additionally, Wang et al. studied the influence of electron transfer between AC and the current collector on the efficiency of FCDI desalination [167]. They compared two types of AC, W1200 and W900, where W1200 exhibited weak electron transfer, leading to a significant 95 % decrease in FCDI performance. Graphitization was the pivotal parameter for the electron transfer character of AC. Remarkably, enhancing AC graphitization degree from 0.69 to 1.03 led to a 20-fold improvement in FCDI desalination efficiency. This heightened graphitization also lowered the minimum energy required for electron escape from the AC surface, effectively boosting electron transfer capacity.

FCDI operates by extracting ions through electrosorption on the surface of charged particles in the flow electrode. Consequently, the surface area and porosity of AC play a pivotal role in influencing the desalination efficiency of FCDI.

ii. Carbon black (CB)

Carbon black (CB) is a finely divided form of carbon, typically produced by the incomplete combustion of hydrocarbons like those present in petroleum oil or natural gas or the thermal decomposition of gaseous hydrocarbons under controlled conditions [168]. It consists of partially fused colloidal spheroidal particles with a paracrystalline graphitic microstructure with a high surface area. It is widely used as reinforcing filler, and conductive additive in various applications, including electrical conductors. In the context of electrode materials for desalination

and energy storage, carbon black is often used to enhance electrical conductivity and facilitate charge transfer within the electrode structure.

To enhance desalination efficiency, CB is commonly used as a conductive additive in static or flowable electrodes in capacitive deionization [114,169]. Incorporating CB into flow-electrodes promotes electronic charge transfer between active materials and current collectors. Liang et al. demonstrated that adding just 1.5 wt% CB to a slurry electrode significantly increased salt removal rate by seven times and raised charge efficiency to 96.5 % at 2.4 V [114]. CB's role as both additive and active material has been explored in energy storage applications [170,171]. In FCDI desalination, Ma et al. compared AC and CB slurry electrodes (2.5 wt%) under SCC and SC modes [85]. Despite CB's lower capacitance, it achieved significantly higher average salt adsorption rates: $1.13 \mu\text{mol}/\text{cm}^2 \text{ min}$ (7.5 times higher) and $0.76 \mu\text{mol}/\text{cm}^2 \text{ min}$ (5 times higher) compared to AC under SC and SCC, respectively. CB flow electrodes suffer from instability problems arising from agglomeration and sedimentation of CB nanoparticles in aqueous electrolyte. This problem has been effectively addressed by Alsultan et al. [153] They introduced an innovative solution by altering the surface of CB with direct blue (DB) molecules, resulting in improved stability and reduced sedimentation. This modified CB slurry enables efficient and durable water desalination in flow-electrode capacitive deionization, showcasing high salt removal rates and minimal energy consumption.

In summary, CB serves as a versatile conductive additive and active material for desalination and energy storage applications. Its integration into electrodes enhances electronic charge transfer and conductivity. Challenges of CB instability have been addressed through innovative modifications, ensuring efficient and enduring water desalination. Future research could explore advanced surface modifications and hybrid materials to further optimize CB's performance in flow-electrode capacitive deionization.

iii. Carbon nanotubes (CNTs)

Since their identification in 1991 [172], CNTs have garnered significant attention because of their unique structural characteristics, which involve carbon atoms arranged in a hexagonal lattice featuring extensive sp^2 hybridized benzene rings. This unique arrangement bestows upon them exceptional mechanical, electrical, optical, and thermal properties, making them highly valuable in a wide range of applications, including water desalination [173,174].

In capacitive deionization systems, CNTs have demonstrated enhanced desalination capacity, leading to their successful incorporation as electrode materials [173,175–178]. Researchers have been actively exploring the synergistic potential of combining CNTs with other electrode materials to develop composites that maximize the advantages of both components. The addition of CNTs to the flow-electrodes in FCDI leads to enhanced salt-removal efficiency, attributed to their remarkable electrical conductivity and effective dispersion potential. Incorporating functionalized CNTs into pristine AC, as demonstrated by Cho et al., established conductive connections among isolated AC particles [115]. Adding just 0.5 wt% of functionalized CNTs within the AC flow-electrodes formed conducting bridges among activated carbon particles, significantly decreasing electron transport resistance. This modification led to a remarkable enhancement in the salt removal rate, increasing it from 8.7 to $34.32 \mu\text{mol cm}^{-2} \text{ min}^{-1}$. This innovation boosted the desalination performance ($5.72 \text{ mmol}/\text{m}^2 \text{ s}$), five times higher compared to pristine AC. Tang et al. probed CNTs as an active electrode material, revealing a high salt removal rate ($8.3 \text{ mg}/\text{m}^2 \text{ s}$) and notably stable desalination behavior [141]. A charge efficiency of 93.3 % was achieved which was superior to that of AC or mesoporous carbon. This was due to the increased electrical conductivity attributed again to the CNTs' bridging effect with low viscosity. Furthermore, Chen et al. recently applied CNTs to increase synergetic desalination and energy efficiency under isolated closed cycle (ICC) mode [179]. With an

average salt adsorption rate (ASRR) of $1.59 \mu\text{mol}/\text{cm}^2 \text{ s}$ (1.7 times greater than without CNTs), they achieved over 99 % charge efficiency and notably low energy consumption ($0.034 \text{ kWh mol}^{-1}$). This system was tested with real saline water, indicating promising practical utility with elevated desalination performance and minimal energy consumption. Additionally, Cai et al. explored the potential of sulfonated CNTs (CNT-S) as an active material, which diminished inter-tube aggregation and amplified network contact between the electrode material and current collector [96]. Utilizing 2.0 wt% CNT-S in the FCDI system showcased a desalination performance demonstrating a 1.6 times higher electrosorption rate and salt removal efficiency compared to unmodified CNTs. Wang et al. combined flow CNTs with AC to achieve an energy-efficient desalination process within FCDI [97]. The CNTs served a dual purpose as electron transporters and capacitive ion capturers alongside AC. The incorporation of 1 wt% CNTs into a 6.41 wt% activated carbon flow electrode led to an increase in the desalination rate with reduced energy consumption compared to pure AC flow electrodes. Rauer et al. investigated a poly(3,4-ethylenedioxythiophene) polystyrene sulfonate CNT (PEDOT:PSS-CNT)-based suspensions as an alternative to conventional carbon-based flow electrodes. The associated conductive hydrogel particles exhibit attributes such as softness, internal porosity, reduced density, hydrophilicity, and a mass-specific salt adsorption capacity surpassing that of AC by a factor of up to ten. Experimental results in FCDI demonstrate that, unlike AC, the intrinsic characteristics of PEDOT:PSS-CNT composite particles streamline the slurry preparation procedure and facilitate circulation of flow electrodes at markedly elevated particle volume fractions [180].

The utilization of CNTs has effectively boosted desalination performances in capacitive deionization systems. The integration of CNTs into FCDI electrodes not only enhances salt-removal effectiveness but also recent research highlights advancements in charge efficiency and energy conservation, driving their application in water desalination. Researchers have shifted their focus towards synthesizing CNTs with surface functionalization, hybrid composites with other nanomaterials, development of hierarchical structures, N-doping, and exploration of scalable and cost-effective methods. These advancements aim to enhance the electrode performance, ion adsorption capacity, and selectivity while addressing practicality and environmental concerns.

iv. Mesoporous carbon (MC)

Mesoporous carbon (MC) stands as a distinctive carbon variant, predominantly synthesized through the use of triblock copolymer templates. This involves combining tetraethyl orthosilicate (TEOS) with triblock copolymer P123, leading to the formation of ordered mesoporous carbon (OMC) upon carbonization at $850 \text{ }^\circ\text{C}$, followed by HF dissolution of silica [181]. Interestingly, resulting OMC displays substantially higher and faster electrosorption capacity at 1.2 V compared to normal AC in CDI experiments, despite having a lower specific surface area (SBET) of $844 \text{ m}^2 \text{ g}^{-1}$. However, OMC boasts a more significant mesopores fraction (82 %).

To address the challenges of limited electronic and ionic transport within FCDI's flow-electrode, Li et al. are the first to recently adopt hollow mesoporous carbon nanospheres (HMCNs) as a flow-electrode in FCDI (Fig. 13) [103]. Comparisons with AC and solid mesoporous carbon nanospheres (SMCNs) were conducted to understand FCDI performance dependence on flow-electrode structural properties. HMCNs flow-electrode exhibited favorable rheological behavior, dispersion, and increased viscosity. EIS analysis indicated reduced electrical resistance and enhanced ion diffusion in HMCNs. As expected, HMCNs also demonstrated superior desalination performance, outperforming AC and SMCNs. Particularly, at 1.2 V with 10 % mass loading, HMCNs achieved an average salt removal rate, charge efficiency, and energy consumption of $0.040 \text{ mg}/\text{cm}^2 \cdot \text{min}$, 92.9 %, and $0.035 \text{ kWh}/\text{mol}$, respectively. This promising desalination performance stems from HMCNs' structural advantages in electronic and ionic transport.

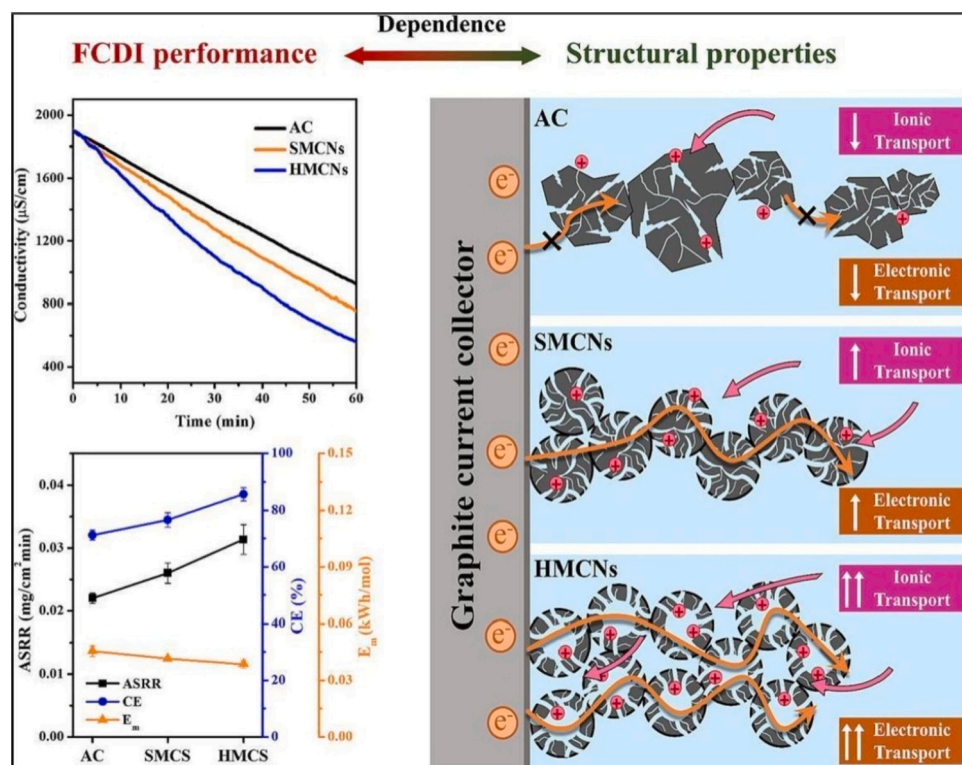


Fig. 13. Structural properties and FCDI performance of the hollow mesoporous carbon nanospheres (HMCNs) applied as flow electrodes by Li et al. [103] Image taken from reference 103.

Mesoporous carbons offer numerous advantages, such as tunable pore size, chemical stability, high specific surface area, and electrical conductivity, rendering them highly valuable in applications beyond electrochemical desalination, including gas separation and catalysis. Recent advancements involving doping and hybridization have introduced more tunable functionalities to MC. However, it is crucial to consider the impact of connectivity and pore sizes in MC, as they may influence material transfer to active sites and affect reactivity. Mesoporous carbon offers compelling attributes that make them highly suitable for FCDI desalination systems. Their distinctive characteristics such as enhanced ion transport, reduced electrical resistance, high conductivity, increased surface area, tailored pore size, and optimized flow dynamics, contribute to enhanced FCDI performance and efficiency. Consequently, there is a compelling case for increased utilization and advancement of mesoporous carbons as prevalent flow electrode materials within FCDI research.

v. Carbon – Transition metal oxides (TMO)

The application of electrode materials for desalination in supercapacitor technologies has garnered significant interest, especially with a focus on utilizing carbon and transition metal oxides (TMOs) [176,182]. When these materials are combined, they create a synergistic effect that enhances the specific capacitance of carbon electrodes [183]. Within capacitive deionization applications, researchers have explored the use of carbon-TMO composites. The incorporation of TMO onto carbon electrodes has a positive impact on CDI desalination efficiency due to their beneficial physicochemical properties. For instance, TMO increase the hydrophilicity of the electrode, resulting in improved ion removal rates [184]. Furthermore, they can modify the surface zeta-potential on the carbon particles, further enhancing ion removal [185].

To advance the practical use of carbon-TMO composites as electrodes in FCDI, it is crucial to carefully consider the stability of the metal oxide in aqueous solutions under varying potentials. Maintaining stability is

crucial for achieving optimal electrode performance. However, the use of such composites may also present challenges related to water electrolysis and parasitic redox reactions. These reactions, including dissolved oxygen reduction, carbon oxidation, and the oxidation of Cl^- ions into chlorine, are more prone to occur when employing carbon-TMO. This is due to the change in the electrode's potential of zero charge, which approaches the parasitic redox reaction potential. Consequently, it is of utmost importance to thoroughly assess potential contamination issues that could arise from eroded TMO entering the treated water during CDI desalination processes.

One example of TMO widely used in CDI is titanium dioxide (TiO_2). TiO_2 possesses unique properties that lead to its widespread utilization across diverse industries. Its photocatalytic capabilities make it valuable in water and air purification, effectively eliminating pollutants. Moreover, TiO_2 plays a crucial role in gas sensors, enabling the detection of gases and volatile organic compounds (VOCs) for applications in environmental monitoring, industrial safety, and automotive systems [186]. Folaranmi et al. improved flow capacitive deionization through the combination of electrospun titanium oxide nanofibers (TiO_2NF) and AC into a uniformly distributed flow electrode [187]. The composite with 1 wt% TiO_2NF exhibited superior capacitance and salt removal rate, attributed to the even distribution that hinders electrode passivation and agglomeration.

The application of AC- TiO_2 composite electrodes in capacitive deionization is essential for effective water treatment and desalination. These electrodes combine the outstanding surface area and adsorption capacity of AC with the beneficial electrochemical properties of TiO_2 . As a result, this composite maximizes ion removal during CDI, leading to improved efficiency, increased ion removal capacity, and prolonged electrode lifespan. TiO_2 , being a transition metal oxide (TMO), has garnered significant attention due to its environmentally friendly characteristics and the ability to control its particle size and structure using various methods, making it the most extensively studied additive in this context.

The utilization of TiO₂-carbon composites holds immense potential in advancing capacitive deionization technology, rendering water treatment and desalination more efficient and sustainable. Continued research and innovation in composite formation and structure regulation will play a crucial role in further improving their performance across various applications. To further advance TiO₂-carbon composites, researchers should focus on achieving uniform nanoparticle dispersion, particularly in their nanofibrous form, to prevent aggregation and agglomeration issues. Recent developments in material development introduced a modified sol-gel method that enhances both capacitance and electrosorption through uniform distribution and crystal structure regulation of TiO₂. However, this method requires high energy input [188].

Another important TMO frequently used in CDI is Zinc oxide (ZnO). The utilization of Carbon-ZnO composites in CDI has become highly significant. Integrating ZnO into the carbon matrix offers numerous advantages. ZnO nanoparticles introduce additional active sites for ion adsorption, greatly enhancing the overall capacity and hydrophilicity of the electrode. Moreover, the presence of ZnO improves electrical conductivity, enabling faster charge transfer. Notably, the carbon-ZnO composite demonstrates exceptional stability and durability during CDI, making it a promising material for long-term applications [189].

In FCDI, Mansoor et al. introduced a novel ZnO/N-GO/PANI composite, synthesized from zinc oxide, nitrogen-doped graphene oxide (N-GO), and polyaniline (PANI) [99]. Characterization techniques confirmed the composite's structural properties and crystallinity. Notably, the ZnO/N-GO/PANI composite exhibits enhanced surface area, greater specific capacitance, and improved electrosorption capacity compared to AC electrode. FCDI tests demonstrate high desalinating efficiency, establishing the composite as a favorable and promising electrode material for FCDI applications.

The incorporation of ZnO into carbon materials is beneficial due to the ZnO polar face, which is rich in oxygenated functional groups, becoming more exposed to the composite surface, thereby enhancing the hydrophilicity of the electrode. Additionally, ZnO acts as a semiconductor material, generating electron-hole pairs under an electric field. Despite the possibility of ZnO nanorods causing pore blockage of carbon due to their morphology, the interfacial electric field under potential reinforces charge transport, thereby promoting ion adsorption in CDI. Recent advancements in carbon-ZnO nanostructures focus on experimental analysis and numerical calculation studies to further enhance their desalination performance. These endeavors aim to unlock the full potential of carbon-ZnO composites in CDI and drive their adoption in water treatment and desalination technologies.

6.4.2. Redox-active electrodes (Faradaic electrodes) for pseudocapacitor and hybrid FCDI

While capacitive carbon materials like AC possess a large SSA that enables superior charge storage at the interface, it experiences a relatively low specific capacitance compared to its theoretical capacitance during electrochemical cycling [190]. To enhance the specific capacitance of AC electrodes, researchers have focused on introducing pseudocapacitive behavior through various approaches [191]. These include creating surface functional groups via chemical treatment, impregnation of metal oxides (such as MnO₂, RuO₂, V₂O₅, MgO, ZnO, etc.), and doping [111,192,193]. As explained before, the pseudocapacitive process involves reversible redox reactions or intercalation processes associated with charge transfer, offering charge storage through faradaic reactions on the electrode surface [194]. Functionalities like carboxylic, phenolic, and lactone groups contribute to extra capacitance through the pseudocapacitive mechanism [195]. Additionally, composite electrodes like MnO₂/AC and RuO₂/AC with mixed Capacitive-Faradaic behavior have been developed for hybrid CDI, demonstrating high performance attributed to the combined capacitive functionality of AC and the pseudocapacitive redox reactions of MnO₂ or RuO₂, respectively [196].

Polyaniline (PANI) is an organic intrinsically conducting polymer. It

is unique in that it can conduct electricity, similar to metals, while still retaining the properties of traditional polymers. Due to its electrical conductivity, chemical stability, and ease of synthesis, PANI has a wide range of applications such as water purification, electronics, photovoltaics, supercapacitors and energy storage. In FCDI application, this redox-active conductive polymer presents a compelling option for creating high-performance slurry electrodes. PANI has many advantages such as elevated electronic conductivity, high theoretical capacitance of 2000 F g⁻¹, straightforward and cost-effective synthesis methods, and good environmental stability. These attributes position PANI as a promising candidate for advancing the development of efficient slurry electrodes in FCDI. In the context of FCDI, PANI exhibits exceptionally high capacitance as an electrode material. Nevertheless, it faces challenges related to poor cyclic stability and susceptibility to decomposition. To address this issue, Singh et al. [197] took a proactive approach by creating a composite structure that combines PANI with AC. During the oxidative polymerization of PANI in the presence of AC, the pores within the AC material serve as sites for capturing and immobilizing PANI. This effect acts as a protective measure, preventing PANI from undergoing decomposition during repeated charging and discharging cycles. The as prepared composite slurry showed a pseudocapacitive behavior and achieved a significant 48 % increase in volumetric capacitance compared to the original AC slurry. Furthermore, the viscosity of the AC-PANI composite slurry was reduced compared to that of the pristine AC slurry at the same mass loading. Similarly, Xiong et al. [198] synthesized a hybrid AC/PANI material by coating polyaniline on activated carbon, aiming to retain conductivity and enhance wettability. Electrochemical assessments unveiled an increase in capacitance and a decrease in ion resistance, resulting in enhanced desalination capabilities. Notably, this approach achieved an adsorption capacity of 13.51 mg/g and a charge efficiency of 92.21 %. It also demonstrated a 39 % higher salt adsorption rate when compared to pristine activated carbon (AC). Similarly, Chen et al. [199] fabricated a composite material comprising PANI and AC using an ice-template in situ polymerization method, capitalizing on PANI's pseudocapacitance and the improved ion pathways within the AC pores. This led to substantial increases in specific surface area and capacitance, and resulted in reduced viscosity and improved conductivity. Notably, the desalination capacity of the FCDI system experienced a remarkable 70 % enhancement. PANI was also used by Chen et al. to address the problems of low conductivity in graphite channels and electrode sedimentation in FCDI [200]. They developed an electrochemically polymerized polyaniline-graphite (PGP) composite current collector. This innovation led to a 1.46 times increase in salt removal rate, reduced Ohmic resistance, improved charge transfer efficiency, modified graphite surface polarity, 15 % reduction in electrode sediment, and consistent performance across multiple cycles. These advancements significantly elevated the deionization effectiveness and stability of FCDI. Also on the same line of Faradaic FCDI, Sun et al. developed a novel sodium vanadium fluorophosphates-reduced graphene oxide (NVOPF-rGO) composite as a Faradaic flow electrode for FCDI application [98]. The structure facilitated rapid Na⁺ and electron transport, leading to improved dispersion, lower resistance, higher desalination rate, and reduced energy consumption compared to conventional AC capacitive electrodes. Optimal parameters achieved an ASAR of 5.32 μg cm⁻² min⁻¹, demonstrating the potential of this approach for enhanced FCDI performance.

Faradaic materials are vital in CDI due to their higher adsorption capacity for high saline solutions, offering promising prospects for electrochemical desalination and energy storage. However, several challenges pose major drawbacks in this emerging field. For instance, intercalation materials serving as electrodes may be limited to capturing specific ions, limiting their selectivity for other ions. Moreover, concerns arise about the viability of these electrodes for desalinating potable water, as metal ion leakage could occur after prolonged desalination cycles, posing health risks and compromising their suitability as potential CDI materials. Another challenge lies in the stability of Faradaic

materials, which is lower compared to carbon-based materials. This instability negatively impacts the long-term performance of Faradaic electrochemical systems.

6.4.3. New electrode materials used in FCDI

Recent research into new materials for electrodes in FCDI reveals several promising developments aimed at enhancing both efficiency and long-term stability. One significant breakthrough is the incorporation of iron-based redox additives, such as iron chloride and iron citrate, into activated carbon electrodes. These additives improve the desalination rate by up to 100 %, with iron chloride showing the highest increase while also maintaining a neutral pH, making it ideal for potable water applications. This method also ensures minimal iron crossover into the water streams, providing a safer alternative for large-scale desalination [146]. Furthermore, the development of porous polyaniline (pPANI) in combination with activated carbon electrodes has introduced pseudo-capacitive effects, increasing both surface area and conductivity. This innovation results in a 70 % improvement in desalination performance, a significant reduction in electrode viscosity, and a more efficient ion adsorption process, all of which are critical for scaling up FCDI technology [201]. Nano-scale carbon materials, such as multi-walled carbon nanotubes (MWCNTs) and graphene nanoplatelets (GNPs), also play a crucial role in enhancing FCDI performance. These materials, when blended with activated carbon, form stable conductive networks or “bridges,” leading to increased ion transport capacity and enhanced conductivity. This blending strategy outperforms traditional micron-scale materials, achieving greater energy efficiency and desalination rates, especially in continuous operation [202]. Additionally, the introduction of a two-stage FCDI system that integrates both free-standing and flow electrodes has shown remarkable potential in doubling desalination efficiency while reducing energy consumption and system size. This approach paves the way for more practical large-scale desalination solutions, making it an attractive option for addressing global water scarcity issues [203].

These advancements in electrode materials emphasize the importance of optimizing the physical and chemical properties of electrodes in FCDI systems. By enhancing conductivity, surface area, and ion selectivity, these new materials enable more energy-efficient, scalable, and

long-lasting desalination technologies.

Table 2 below summarizes various studies on FCDI systems, focusing on salt removal efficiency, energy consumption, electrode materials, Average Salt Adsorption Rate, and cell configurations.

The environmental implications FCDI technology involve concerns about the disposal of electrode materials and the management of electrolytes. The primary environmental challenge lies in the life cycle and disposal of the flowable particle electrodes, often composed of carbon-based materials. While these materials are efficient in ion removal, their large-scale application may lead to waste generation if not properly managed. Recycling or repurposing these carbon materials is essential to mitigate the environmental footprint [79]. Electrolyte management also poses a challenge, as the use of saline or chemically laden solutions can introduce contaminants into the environment if not treated or recovered efficiently. Advances in FCDI technology, such as closed-loop systems for electrode and electrolyte recovery, show promise in reducing environmental harm by minimizing waste and enabling resource recovery. However, the scaling up of these technologies requires careful consideration of operational costs, energy demands, and the environmental impact of continuous electrolyte cycling. Future research should focus on improving material efficiency and developing sustainable management strategies for both electrodes and electrolytes to ensure FCDI's environmental viability.

Having explored the various electrode materials and their impact on the performance of FCDI systems, it is essential to understand how these materials interact with key operational parameters. The efficiency and scalability of FCDI systems are not solely determined by the choice of electrode materials but are also significantly influenced by parameters such as flow rate, applied voltage, concentration of the electrolyte, and system configuration. These operational factors play a crucial role in optimizing ion removal and energy consumption. In the following two sections, we will examine these parameters in detail to provide a comprehensive understanding of FCDI system performance.

7. Feed saline water in FCDI

The efficiency of an FCDI cell is significantly impacted by the concentration, composition, and duration of water subjected to the

Table 2
Comparative representation of diverse flow electrode capacitive deionization technologies performance metrics.

Title	Desalination Efficiency	Energy Consumption	Electrode Material	Desalination rate	Configuration	Ref
Carbon aerogel electrode for excellent dephosphorization via flow capacitive deionization	-	1.74 kWh kg ⁻¹ p ⁻¹	Carbon Aerogel	1.20 μg·min ⁻¹ ·cm ⁻²	-	[204]
Enhancing the electronic and ionic transport of flow-electrode capacitive deionization by hollow mesoporous carbon nanospheres	-	0.035 kWh mol ⁻¹	Hollow Mesoporous Carbon Nanospheres	0.040 mg cm ⁻² min	-	[205]
New insights into the performance analysis of flow-electrode capacitive deionization using ferri/ferrocyanide redox couples for continuous water desalination	76.38 %	0.27 kWh mol ⁻¹	High Surface Area Activated Carbon and MWCNT	43.925 μg cm ⁻² s	Serpentine Flow	[147]
A hexagonal honeycomb-shaped flow channel for high-efficient desalination and flowability in flow electrode capacitive deionization	64.89 %	-	Carbon Slurry	2.05 μmol cm ⁻² min ⁻¹	Hexagonal Honeycomb	[206]
Development of Composite Nanostructured Electrodes for Water Purification	-	0.004 to 0.8 Wh mg ⁻¹	AC with PVA/PDMAAC	6 to 11.5 μg g ⁻¹ min ⁻¹	-	[207]
Capacitive Deionization Incorporating a Fluidic MOF-CNT Electrode for the High Selective Extraction of Lithium	89 %	-	ZIF-8/CNT	-	Three-stage system	[208]
Magnetic array for efficient and stable Flow-electrode capacitive deionization	>95 %	10 μmol J ⁻¹	Magnetic carbon (MC) and Activated Carbon (AC)	0.35 μmol cm ⁻² min ⁻¹	Magnetic array	[209]
A new static mixer concept for enhanced desalination performance in flow-electrode capacitive deionization (FCDI) systems	40 %	99 kJ mol ⁻¹	2 wt% Activated Carbon	4.424 μg NaCl cm ⁻² s ⁻¹	Membrane-assisted static mixer	[210]
Enhanced desalination performances by using porous polyaniline-activated carbon composite flow-electrodes in capacitive deionization system	85 %	0.70 kWh kg ⁻¹	pPANI-AC	1.73 μmol cm ⁻² min ⁻¹	pPANI-AC Flow Electrodes	[211]
Surface-treated carbon black for durable, efficient, continuous flow electrode capacitive deionization	78.5 %	1.83 J mg ⁻¹	DB-modified Carbon Black	53.43 μg cm ⁻² min ⁻¹	DB/CB Flow Electrodes	[212]

treatment process. Given that FCDI technology is still in its early developmental stage, much of the research has primarily focused on synthetic water compositions. There have been only a few investigations involving feed water containing multiple salts, where selectivity hinges on factors such as charge, size, and other operational parameters, as discussed in other studies. The impact of the remaining two variables, namely concentration and residence time, is further elucidated below.

7.1. Influence of feed saline water concentration

The focus on the parameter of feed water concentration has been a central aspect in the field of FCDI research since its inception. This emphasis is illustrated in the work conducted by Jeon et al. [21], who delved into the impact of varying NaCl concentrations in the feed water on the removal rate and efficiency of FCDI cells. It was observed that the removal rate exhibits an upward trend as the initial feed water concentration increases, owing to the heightened salt content in the spacer channel. This, in turn, results in an augmented ion permeation through the membrane into the flow electrode chamber, consequently reducing ionic resistance [21]. For instance, the Ohmic resistance value decreased from 15.55 Ω to 5.68 Ω as the ammonia concentration increased from 20 mg L⁻¹ to 100 mg L⁻¹ in the feed water [24]. This phenomenon can be attributed to the intensification of electrosorption of ions on microporous carbon particles, which is facilitated by the overlap of the EDL within the micropores of these carbon particles when the salt concentration rises [213,214]. Conversely, the effectiveness of salt removal experiences a decline as the feed water concentration increases [21]. This decrease primarily occurs because the quantity of salt introduced into the cell spacer exceeds the amount that is removed from it, as demonstrated by Choi et al. [215]. However, Choi et al. [215] also observed that the salt removal efficiency of the FCDI cell remains relatively stable over a wide concentration range, spanning from 4000 ppm to 35,000 ppm. Furthermore, as the concentration of salt in the spacer increases, there is an augmentation in both energy recovery and salt adsorption capacity, as documented by previous research [71]. Jeon et al. [84] investigated the impact of the feed water on energy recovery by utilizing both deionized water and saltwater during the discharging phase. Interestingly, they observed a 50 % increase in energy recovery when deionized water was substituted for saltwater during the discharge process. The increase in energy recovery can be attributed to ions migrating from the spacer to the flow electrode during discharge, counteracting the movement of ions from the flow electrode to the spacer. As a result, the utilization of deionized water enhances energy recovery in this scenario [84]. An increased concentration of feed water brings about a higher current density at an equivalent cell voltage. This phenomenon can be ascribed to the diminishing cell resistance as the feed water concentration rises, as demonstrated in prior studies [69]. Nevertheless, a higher feed water concentration leads to a decrease in current efficiency, primarily due to the declining selectivity of the IEM [216]. Additionally, as the difference in salinity between the feed and product streams increases, there is a concurrent rise in energy demand. This increase in energy demand arises as ion diffusion counteracts ion migration, resulting in an escalation in the energy required for the process, as indicated by previous research [71].

7.2. Influence of feed saline water flow rate

The flow rate of the feed water plays a pivotal role in directly affecting both the desalination rate and the overall performance of the system. Within a single FCDI cell unit, the feed water flow rate can vary from <1 mL min⁻¹ to 9 mL min⁻¹, and this range is essential to achieve effective and uninterrupted deionization [67]. It is worth noting that Yang et al. [66] managed to attain a flow rate of 15 mL min⁻¹ by employing parallel stacking of individual FCDI cells. The ASRR (Average Salt Removal Rate) experiences a slight increase as the flow rates are elevated. This phenomenon occurs because a higher feed water flow rate

facilitates the constant replenishment of ions in the spacer region, resulting in improved conductivity at a specific voltage and, consequently, a higher ASRR [69,72,217]. Gendel et al. [67] even demonstrated complete salt removal at a feed water flow rate of <1 mL min⁻¹ during continuous operation. However, it's essential to note that an increase in the flow rate leads to reduced deionization efficiency. This reduction is primarily attributed to the decreased retention time of ions within the spacer [80].

In summary, the effectiveness of FCDI is influenced by water concentration, composition, and retention time. Research primarily focuses on synthetic water, with limited multi-salt investigations. Concentration affects removal rate, efficiency, energy recovery, and selectivity. Feed water flow rate impacts desalination rate and performance, with higher rates improving conductivity but potentially reducing efficiency due to shorter ion retention times.

8. FCDI operation mode

The primary force propelling the deionization process, facilitating the migration of ions from the feed channel to the flow electrode through the IEMs, is the external energy source. This energy supply can manifest as either a constant voltage (CV) or a constant current (CC). The performance and characteristics of FCDI exhibit variations when shifting between these power supply modes, specifically between CV and CC. In most of the research conducted, the performance of FCDI is evaluated under CV operation. This is because CV mode offers the advantage of optimizing performance by investigating the impact of varying effluent concentrations while adjusting other parameters. However, in practical commercial applications of FCDI, maintaining a consistent effluent concentration is often essential, tailored to the specific application. When FCDI is operated in CC mode, the resulting effluent maintains a constant concentration, ensuring that the deionization process yields a predictable outcome. The precise control of the effluent concentration can be achieved by fine-tuning the value of the CC parameter [12,218].

To enhance the desalination rate of an FCDI cell, increasing the applied voltage proves effective. Applying a potential difference across an electrode alone can induce ion transport across membranes, similar to electrodialysis. However, to optimize FCDI cell performance, it is advisable to operate below 1.2 V, as water electrolysis initiates at 1.23 V. A potential difference >1.23 V would result in accelerated Faradic reaction [219,220]. Elevated voltages result in enhanced ion adsorption on carbon particles, leading to higher desalination rates but at the cost of increased energy consumption. Liang et al. proved that desalination rates notably rise with voltage increments [114]. For instance, desalination rate increased approximately seven times when the voltage was elevated from 0.6 V to 4.8 V. Achieving efficient desalination at low voltage involves strategies like minimizing membrane resistance (via thickness and material adjustments) and optimizing module design (flow electrode composition and spacer channel) to mitigate unwanted potential drops [217]. At higher voltages, charge efficiency diminishes due to Faradic reactions, particularly electrolysis [23,73,221]. Liang et al. [114] discovered that charge efficiency dropped from 91 % at 1.2 V to 69.5 % at 4.8 V. These faradic reactions not only impact performance but also expedite the aging of flow electrodes, particularly on the positive flow electrode's surface [222]. In cases where flow electrode regeneration is absent or flow electrode capacitance is limited (as in batch mode), the preference is for Constant Voltage (CV) mode. Operating in Constant Current (CC) mode can lead to unwanted reactions and potential system failure due to continuous voltage buildup [221].

Raising the current density yields a relatively linear increase in the deionization rate, but it concurrently results in a decline in current efficiency [69,80]. However, beyond a certain point of current density, a significant increase in voltage is needed due to the rise in internal resistance, which grows from 12.5 Ω to 25 Ω , dominated by polarization resistance. IEMs have an inclination for counter ions, causing local ion

concentrations to surpass those of the bulk solution ions. This concentration gradient induces a phenomenon known as back diffusion, which hinders the transport of ions at a specific voltage. This, in turn, gives rise to polarization resistance at the interface between the influent and the membrane [80,223]. In a system with five parallel-stacked FCDI cells, the current density and charge efficiency remain comparable to those of a single FCDI cell, while the total electrical current can increase by up to five times. This indicates a uniform distribution of ions across each unit cell. [66] However, as current density increases, the preferential removal of Ca²⁺ ions decreases. This is because Ca²⁺ ions, which have a higher affinity for the Ion Exchange Membrane (IEM), are more effectively removed at lower current densities. At these lower densities, divalent ions like Ca²⁺ migrate through the Cation Exchange Membrane (CEM) more readily than monovalent ions such as Na⁺. Additionally, at lower current densities, divalent ions replace monovalent ions on the activated carbon surfaces, aiding in the selective removal of Ca²⁺ ions [224,225]. The current density also affects the selectivity of carbon materials. Zhang et al. demonstrated that their system showed greater selectivity for NH₄⁺ ions over Na⁺ ions at lower current densities [90]. This difference in behavior is attributed to the disparities in the hydrated radii of NH₄⁺ (3.31 Å) and Na⁺ (3.58 Å), with smaller ions of the same valence charge being more quickly electro-adsorbed into the micropores of carbon particles.

The deionization process in FCDI is driven by external power supply (CV or CC), impacting its performance and behavior. CV optimizes performance through varied effluent concentrations, while CC maintains consistency for practical applications. Higher voltage enhances desalination but increases energy consumption, and results in Faradaic reactions, affecting charge efficiency and electrode aging. Current density affects deionization rate, selectivity, and resistance. Uniform segmentation and ion preferences contribute to the process.

9. FCDI performance indicators

To comprehensively assess and define the efficiency of any capacitive deionization system, it is crucial to examine desalination performance metrics. Achieving the highest level of optimization in the CDI process requires the identification and thorough comprehension of the key parameters that influence the separation of ions during the process. This process is largely affected by the system's design, the electrodes design, and the operating conditions.

9.1. Maximum salt adsorption capacity (mSAC)

This parameter signifies the amount of adsorbed ions relative to the weight of the adsorbent, expressed as milligrams of salt adsorbed per gram of electrode material. It is calculated using the eq. (1).

$$mSAC = \frac{(C_i - C_e) V}{m_{\text{electrode}}} \quad (1)$$

Where C_i is the initial salt concentration and C_e the final salt concentration at the equilibrium (when the electrode is saturated with ions) of the saline solution, V is the volume of the feed solution and $m_{\text{electrode}}$ is the mass of the electrode material. mSAC is predominantly influenced by the inherent characteristics of the electrode, including factors such as porosity, surface area, capacitance, physical and chemical properties, surface thickness, and design. Additionally, it is impacted by the electrochemical conditions, such as the applied potential, feed salinity, and hydrodynamics. mSAC is directly associated with the electrode's storage capacity and is regarded as one of the key parameters for assessing the performance of the electrodes.

9.2. Average salt adsorption rate (ASAR)

This parameter is related to the adsorption kinetics. In other words, it

provides an idea about ion migration rate to the pores of the electrode where they will be adsorbed and temporarily stored. It gives the amount of salt adsorbed per electrode mass per unit time (mg/g min⁻¹). It is calculated according to eq. (2).

$$ASAR = \frac{SAC}{t} = \frac{m_{\text{solutes}}}{m_{\text{electrode}} t} \quad (2)$$

Where SAC (mg.g⁻¹) is the salt adsorption capacity with $SAC = \frac{m_{\text{solutes}}}{m_{\text{electrode}}}$, m_{solutes} (mg) is the mass of salt removed, $m_{\text{electrode}}$ (g) is the mass of dry electrode material, and t is the adsorption time. ASAR is affected by different parameters like the initial feed water concentration, the flow rate, the cell configuration, but most importantly by the driving force (potential difference applied to the cell) which strongly affect the desalination kinetics. It depends also on the electrodes' physico-chemical properties and design. ASAR also has a large impact on the CAPEX and the OPEX of the desalination process.

9.3. Charge efficiency (CE)

The charge efficiency or Faradaic efficiency of a CDI system is the ratio of the electrical charge used specifically for ion adsorption to the total electrical charge applied to the system. The rest corresponds to the Ohm loss plus the current resulting from Faradaic reactions in case of Faradaic CDI. CE is calculated according to eq. (3).

$$CE = \frac{z (C_i - C_f) V F}{\int Idt} \quad (3)$$

Where z is the equivalent charge of the ions, C_i is the initial salt concentration of the feed solution, C_e the final salt concentration, V is the volume of the feed solution, F is the Faradaic constant (96,485C) and $\int Idt$ is the integrated quantity of charge passed to the system as a function of time. Current efficiency is an important indicator of the energy consumption of a CDI system. Higher charge efficiency means less charge loss and thus higher energy efficiency.

9.4. Specific energy consumption (SEC)

SEC quantifies the energy cost of the desalination process. SEC_V is expressed in kWh/m³ as the total electrical power used per hour to produce one cubic meter of desalinated water and is calculated according to eq. (4).

$$SEC_V = \frac{W}{V_{\text{perm}}} = \frac{\int P(t) dt}{V_{\text{perm}}} \quad (4)$$

Where W is the energy supplied, P is the applied power, t is the time of treatment, and V_{perm} is the volume of desalinated water. Since CDI process works in general at constant low voltage with changing current over time, eq. (4) can be replaced by eq. (5) as follow:

$$SEC_V = \frac{U \cdot \int I(t) \cdot dt}{V_{\text{perm}}} \quad (5)$$

Where U is the applied constant voltage, $I(t)$ is the current response to this applied voltage at a given time and V_{perm} is the volume of desalinated water.

Since capacitive deionization unlike other desalination technologies targets the solutes and not the solvent, it is more accurate to express the specific energy consumption as SEC_m in kWh/kg of removed salt rather than SEC_V in kWh/m³. SEC_m in this case can be more accurately calculated as shown in eq. (6).

$$SEC_m = \frac{U \cdot \int I(t) \cdot dt}{(C_i - C_f) V_{\text{perm}}} \quad (6)$$

Where U is the applied constant voltage, $I(t)$ is the current response to

this applied voltage at a given time, C_i is the initial salt concentration of the feed solution, C_e the final salt concentration and V_{perm} is the volume of desalinated water.

Energy consumption is a critical parameter to consider when evaluating a CDI process making SEC one of the most important CDI performance indicators. SEC value is mostly affected by the feed salinity and the electrode material.

9.5. Electrochemical indicator for electrode material

The specific capacitance (C_s) is a measure of the electrostatic energy storage capacity per unit mass of an electrode material in an electrochemical system. Typically expressed in Farads per gram ($F g^{-1}$), it quantifies the ability of a material to store electrical charge at a given potential. The specific capacitance is calculated from a CV cycle and is derived from the relationship between the double-layer charging current (i), the scan rate (ν), and the applied potential. It can be expressed as follows:

$$C_s = \frac{\int i dV}{2 \nu \Delta V m} \quad (7)$$

where C_s is the specific capacitance ($F g^{-1}$), i is the double-layer response current (A) obtained from the CV cycle, ΔV is the potential window (V), ν is the potential scan rate ($V s^{-1}$), and m is the mass of the electro-active materials in the electrode (g).

To note that C_s can also be calculated according to eq. (8):

$$C_s = \frac{\int i dV}{2 \nu \Delta V A} \quad (8)$$

Where A is the effective area (cm^2) of the electrode material used in the CV test. In this case C_s is calculated in $F cm^{-2}$.

9.6. Technical indicators

Additional technical indicators pertaining to the fluidic behavior of flow electrodes within the FCDI system were computed. These metrics are established to assess process performance from a technical perspective, encompassing parameters such as Reynolds number, sedimentation velocity, and linear pressure drop.

9.6.1. Reynold's number

Reynolds number shows the flow nature of the electrode (collision pattern) in the flow channel. It was calculated according to the following equation:

$$Re = \frac{\rho \nu D}{\eta} \quad (9)$$

Where ρ is the density of flow-electrode, ν is the velocity of flow electrode in the flow channel calculated from the flow rate of the flow electrode ($0.0024 m^3 h^{-1}$) to the area of the flow cell ($5.4 \times 10^{-6} m^2$), D is the hydraulic diameter of the flow channel, 2.6 mm; η is the average coefficient of dynamic viscosity of flow-electrode as a function of shear rate at 25 °C.

9.6.2. Sedimentation velocity

Stokes' law of sedimentation shows the sedimentation velocity or the drag force resisting the fall of small particles through a fluid medium under the influence of gravity, and is described by eq. (10) as followed:

$$V = \frac{2(\rho_p - \rho_f)gR^2}{9\eta} \quad (10)$$

where ρ_p is the particle density, ρ_f is the fluid density, g is the acceleration of gravity, R is the particle radius, and η is the viscosity [226].

9.6.3. Linear pressure drop

The linear pressure drop is the loss of mechanical energy caused by friction on the common walls of the network (transformation into thermal energy). It is directly affected by the fluid viscosity, and is described the following equation:

$$\Delta P = \mu \frac{L}{D} \frac{1}{2} \rho \nu^2 \quad (11)$$

where μ is the friction coefficient with $\mu = 64/Re F = 64/Re$, L and D are the canal length and diameter, ρ is the fluid apparent density, and ν its velocity [227].

9.6.4. Pumping energy consumption

The consumption of additional pumping energy ($E_{pump-FE}$) due to the recirculation of slurry electrodes has been overlooked in FCDI earlier studies. The pumping energy can be quantified as follows:

$$E_{pump-FE} = n_{cell} \frac{\Delta P_{FE} V_{FE}}{n_{pump} V_d} \quad (12)$$

Where n_{cell} is the number of FCDI half cells, n_{pump} is the pump efficiency, ΔP_{FE} is the hydraulic pressure drop in a flow channel of an FCDI half-cell, V_{FE} is the flow rate of the flow electrode and V_d is the flow rate of the dilute water stream.

10. Ion selectivity in FCDI

The ion selectivity constitutes a pivotal application within the realm of FCDI technology, enabling the targeted removal of specific ions. The efficacy of this selectivity hinges upon diverse factors, including electrode materials, surface charge, and operational parameters. Notably, advancements have been made by researchers to tailor the ion selectivity of FCDI systems, meticulously designing and selecting electrode materials. The choice of electrode materials emerges as a key determinant for achieving ion selectivity in FCDI. Enhanced removal of distinct ions has been achieved through specialized substances or coatings applied to the electrodes. For instance, the utilization of graphene-based materials as electrodes permits the efficient removal of ions such as Ca^{2+} and Mg^+ , while allowing the passage of Na^+ and Cl^- . This selectivity arises from the differing sizes, hydration characteristics, and interactions of ions with electrode surfaces. Moreover, operational parameters have significant influence on ion selectivity during the deionization process. Parameters such as applied voltage, flow rate, and pH exert notable effects on ion selectivity within FCDI. The electric field strength further modulates ion transport, influencing the removal rates of different ions. Flow rate, a pivotal parameter, impacts ion selectivity by optimizing ion residence time in the electrochemical cell, thereby enhancing the targeted ion's efficient removal. Manipulating these parameters has enabled researchers to heighten removal efficiency for specific ions, engendering time-dependent selectivity.

In FCDI, ion selectivity is governed by multiple mechanisms, such as differences in ion size, charge density, and the interaction with functionalized membranes or electrodes. One primary method to enhance ion selectivity is the use of polymer multilayer-functionalized ion exchange membranes, which can switch their affinity between monovalent and divalent ions based on ion characteristics like charge density and size. This enables the selective recovery of ions such as Na^+ over Ca^{2+} , as demonstrated by Tran et al., who achieved a selectivity ratio of 3.26 for Na/Ca through membrane functionalization [228]. Moreover, the choice and optimization of carbon-based flow electrodes also contribute to ion selectivity, as nano-scale materials such as multi-walled carbon nanotubes (MWCNTs) and graphene nanoplatelets (GNPs) enhance desalination efficiency by providing more stable and conductive bridges between particles, as shown by Ratzel et al. [202] Altogether, optimizing the materials and operational parameters in FCDI, is crucial for targeting specific ions and improving the efficiency of ion recovery in

various applications.

By strategically adjusting the charging and discharging processes, Xu et al. [229] achieved selective separation of phosphorus (P) from synthetic urine solution rich in chloride (Cl^-) ions. During charging, both P and Cl^- ions migrated to the anode chamber and adhered to charged carbon particles. Faradaic chemical reactions led to the generation of hydrogen ions (H^+), converting charged P ions to uncharged H_3PO_4 , which were spontaneously released. Reversing electrode polarity predominantly pushed charged species like Cl^- back into the spacer chamber, while neutral H_3PO_4 remained in the anode chamber due to slow pH changes. Thus, a P-rich solution formed, facilitating easy P ion harvesting. Furthermore, Zhang et al. [230] used activated carbon-based FCDI to recover high-value ammonia solution. Electrosorption led to ammonium ion migration to the flowable activated carbon cathode via the ion exchange membrane. This resulted in localized pH increase, causing ammonium deprotonation into uncharged $\text{NH}_3(\text{aq})$. Reversal of polarity released constantly charged ions (Na^+ , K^+ , Ca^{2+}) back into the waste stream, while uncharged $\text{NH}_3(\text{aq})$ was trapped in the cathode, yielding an ammonia-rich solution. On the same line of ion selectivity, Jiang et al. [231] additionally examined the influence of FCDI parameters on selective fluoride (F^-) removal from brackish groundwater. Lower current and shorter residence times favored F^- removal over Cl^- . FCDI was also employed for selective continuous Cr(VI) removal from brackish wastewater, showcasing higher Cr(VI) removal than Cl^- [232]. The two ions exhibited distinct behaviors, with pH and coexisting carbonate anions affecting Cr(VI) removal. Additionally, FCDI offers an eco-friendly, chemical-free solution for NH_4^+ removal and recovery, though Na^+ coexistence presents challenges in this field. Lin et al. [233] proposed a potassium dititanate ($\text{K}_2\text{Ti}_2\text{O}_5$ or KTO) activated carbon flow-electrode for selective NH_4^+ removal over Na^+ . The presence of KTO enhanced NH_4^+ adsorption selectivity, boosting the adsorption and desorption efficiency. Moreover, a four-chamber FCDI configuration was used for phosphorus extraction as solid vivianite [234]. The setup generated dephosphorylated water and P-rich brine, demonstrating superior P selectivity over SO_4^{2-} with precipitated vivianite exhibiting high P content (12.4 %) and smaller volume.

In summation, optimizing electrode materials and operational parameters within FCDI is pivotal for achieving high ion selectivity, facilitating targeted ion removal and recovery. This knowledge paves the way for more efficient and selective FCDI systems for water treatment applications.

11. FCDI applications: exploring beyond desalination

Desalination holds the primary focus in FCDI technology research. FCDI demonstrates the capability to perform desalination effectively over a broad spectrum of salt ion concentrations owing to its exceptional ability to adsorb ions. Presently, efficient desalination is achievable primarily under brackish water conditions, considering the current cell configuration and electrode materials. Nonetheless, FCDI consistently outperforms traditional CDI across the entire spectrum of salt ion concentrations [235]. For instance, when considering a 25 mM influent, Hand et al. [236] observed an energy consumption of 0.64 kWh m^{-3} for conventional CDI, whereas FCDI exhibited a lower energy consumption of 0.42 kWh m^{-3} . This difference becomes even more pronounced when dealing with a 50 mM influent, where energy consumption increases fivefold for conventional CDI and only twofold for FCDI. While still in its initial stages of development, FCDI shows promise to outperform current commercial desalination methods, including those applied to seawater desalination. Earlier sections of this review delve into diverse research aspects, including electrode materials, cell design, and operational conditions relevant to desalination.

Beyond this, the concept of ion selectivity in FCDI has opened up a realm of possibilities beyond its conventional application in desalination. The ability to tailor the removal of specific ions has broadened the horizons for diverse applications of FCDI technology. This section sheds

light on various innovative applications of FCDI, leveraging its ion selectivity feature.

11.1. Nutrients recovery

Untreated municipal and agricultural wastewater contain substantial nitrogen and phosphorus nutrients, posing issues like eutrophication if released untreated. Traditionally, complex and energy-intensive biological processes remove these nutrients in wastewater treatment plants [237,238]. With fertilizer demand rising due to increasing food production needs, the energy-intensive Haber-Bosch process contributes 1.5–2.5 % of global energy consumption, discharging ammonia waste [239]. Recovering these nutrients from municipal and agricultural wastewater could alleviate the fertilizer demand issue [240]. Various studies explore the potential of low-energy FCDI for nutrient removal and recovery from wastewater treatment, as discussed below.

Bian et al. [26] conducted a study on the simultaneous extraction and reclamation of nitrogen and phosphorus nutrients (specifically $\text{PO}_4^{3-}\text{-P}$, $\text{NH}_4^+\text{-N}$, $\text{NO}_3^-\text{-N}$) using a conventional FCDI module. They observed an important increase in the removal efficiency of $\text{NH}_4^+\text{-N}$ and $\text{NO}_3^-\text{-N}$, achieving up to 99 %, as the carbon loading increased from 5 wt% to 15 wt%. Conversely, the removal efficiency of $\text{PO}_4^{3-}\text{-P}$ displayed heightened sensitivity to carbon loading, with an increase in efficiency from 50 % to 91 % as the carbon loading rose from 5 wt% to 15 wt%. This heightened sensitivity was attributed to the physical adsorption of phosphates on adsorption sites, which became more pronounced with higher carbon loading. The relatively low removal efficiency of phosphate was attributed to its larger hydrated radius and the lower concentration of phosphate ions in the source water. Nutrient recovery was positively correlated with discharge time, resulting in peak recovery efficiencies of 72.5 %, 82.9 %, and 89.4 % for $\text{PO}_4^{3-}\text{-P}$, $\text{NH}_4^+\text{-N}$, and $\text{NO}_3^-\text{-N}$, respectively. In another innovative approach, Zhang et al. [90] invented an ammonia recovery technique from synthetic wastewater by combining FCDI with a hydrophobic gas-permeable membrane contractor. NH_4^+ and NaCl solution was used as the feed water. During desalination, NH_4^+ ions migrated to the cathode chamber, where the alkaline environment favored the formation of NH_3 species. Given that NH_4^+ species predominate at pH levels below 9.3, while NH_3 species become dominant at pH levels above 9.3 (with the pKa of the $\text{NH}_4^+\text{-NH}_3$ acid-base pair being 9.3), the dissolved NH_3 was then extracted through a membrane contractor and was retrieved as an acidic solution ($(\text{NH}_4)_2\text{SO}_4$). This method resulted in removal and recovery efficiencies of 90 % and 60 %, respectively. In a follow-up study, Zhang et al. [91] replaced the hydrophobic gas-permeable membrane with a flat sheet gas-permeable membrane (FGM), positioned adjacent to the cathode. On the opposite side of the FGM, an additional spacer containing an acid solution was placed. This configuration yielded a consistent recovery efficiency of 80 %. Crucially, the natural Faradaic reactions occurring within the flow electrode chamber of the FCDI cell enabled the necessary pH adjustment, setting it apart from alternative ammonia recovery techniques that rely on external chemicals for pH regulation. As cited in the previous section, Zhang et al. [89] also documented the chemical-free recovery of a valuable ammonia-rich solution from wastewater using a conventional FCDI cell. During the charging phase, NH_4^+ and Na^+ ions migrated to the cathode chamber through the CEM. Faradaic reactions led to an increase in pH, prompting the conversion of NH_4^+ ions into uncharged NH_3 , which subsequently adhered to the carbon surface as discussed earlier. During the discharge phase, the uncharged NH_3 was repelled by the Cation Exchange Membrane (CEM), whereas Na^+ ions moved through the CEM back into the desalination chamber, leading to a notable ammonia concentration of up to 927 mg L^{-1} in the cathode chamber. The considerable cell wall permeability (CWP) played a crucial role in dampening Faradaic reactions, thereby minimizing pH fluctuations. When using a CWP of 5, the pH within the flow electrode chamber rose from 7 to 11.9 during charging and then decreased to 9.2 during discharging, establishing an ideal pH environment for NH_4^+ .

Conversely, in scenarios where a CWP of 0 was employed, the pH plummeted to 4.2 during discharging, impeding the trapping of NH_3 due to excessive Faradic reactions. Fang et al. [24] investigated the retrieval of pre-concentrated ammonia from municipal wastewater using FCDI under various operational settings. They also studied the effect of feed water pH on ammonia recovery. The most favorable ammonia recovery was obtained at pH 4. Under alkaline conditions, NH_4^+ was transformed to NH_3 , rendering it unresponsive to deionization. In contrast, under acidic conditions (pH < 4), carbon particles exhibited a preference for adsorbing H^+ ions over NH_4^+ , attributed to the smaller hydrated radius of H^+ ions. When operating under the optimal parameters, they achieved a concentrated solution of ammonium salt (NH_4^+) at 20 times the original concentration, albeit with a reduced current efficiency. This reduction was attributed to back diffusion, resulting from a flow electrode concentration 127 times higher than that of the source water, driven by high osmotic pressure. In a study conducted by Song et al. [75], they investigated the non-electrostatic influence of different components in the FCDI process for nitrate removal. Nitrate's non-electrostatic adsorption onto AC particles posed challenges in regeneration. While non-electrostatic adsorption initially played a significant role in nitrate removal, the diminished adsorption capacity did not severely impact overall performance. Remarkably, the cost-effectiveness of nitrate removal was found to be higher when reducing levels from 10 mg $\text{NO}_3\text{-N L}^{-1}$ to the standard level (<1 mg $\text{NO}_3\text{-N L}^{-1}$) compared to starting at 50 mg $\text{NO}_3\text{-N L}^{-1}$. This approach led to a remarkable water recovery of 91.4 % by periodically replacing the electrolyte during continuous deionization in the SCC mode.

11.2. Recovery of valuable ions

FCDI emerges as a sustainable innovation, embodying ion selectivity, low energy consumption, and eco-friendliness. FCDI is capable of selectively harvesting valuable ions, requiring minimal energy input. This eco-conscious approach, coupled with continuous flow operation, marks a pivotal advancement in responsible water treatment and resource recovery.

With the expansion of the battery industry and escalating waste battery accumulation, the retrieval of lithium (Li) has garnered notable significance. Progress in electrochemically mediated Li recovery is underscored by the emergence of Li-selective electrodes, exemplified by lithium manganese oxide (LMO) [241]. Yoon et al. [241] introduced a hybrid FCDI strategy employing LMO in conjunction with silver-coated activated carbon (Ag-AC) electrodes. The implementation of Ag coating culminates in a substantial 32 % augmentation of Li uptake efficiency as compared to the pristine AC electrode system. Furthermore, the employment of the "stop-flow operation" technique engenders an impressive enhancement, elevating the Li selectivity coefficient to 171, thereby FCDI affords substantial contributions to the progression of electrochemical Li recovery technology [241]. Advanced oxidation processes (AOPs) offer potent organic compound degradation, but persistent carboxylates hinder breakdown, leading to chemical waste and carbon depletion. Extracting minor carboxylates is crucial [242]. Traditional electrochemical techniques pose risks of elevated energy consumption and organic substance deterioration. Yu et al. [242] utilized an FCDI system, operating via the SCC mode, to extract and recuperate obstinate acetate and oxalate. Carbonaceous flow-electrodes were pivotal, reducing energy consumption and augmenting recuperation efficiency. Under optimized parameters, >80 % of acetate and oxalate were efficiently reclaimed, with minimal energy expenditure (0.45 kWh/m³). The utilization of FCDI under SCC mode emerged as a promising avenue for unceasing carboxylate recovery. Utilizing wastewater for CO_2 mineralization is a prospective avenue, tackling environmental issues while yielding economic advantages. Han et al. [243] presented a novel FCDI system to effectively extract bicarbonate ions and enrich ammonium bicarbonate from synthetic swine wastewater. A parametric exploration of the FCDI system was undertaken, involving

adjustments to operational variables including applied voltage, feed solution flow rate, and feed solution pH, with the aim of proposing optimal operational settings. Under optimized parameters, high-purity NH_4HCO_3 with improved removal kinetics and lowered energy consumption was attained [243]. Iodine (I) is an essential, non-renewable trace element with scarcity. The nuclear industry generates low-concentration radioactive iodine, posing environmental and human risks. Liu et al. [35] explored FCDI for sustainable iodine recovery and removal from wastewater. FCDI parameters influenced iodine enrichment. I^- was preferentially removed over Cl^- , exhibiting distinct behaviors. Favorable selectivity and removal (>96 %) persisted through 100 cycles under optimal FCDI conditions. Extended batch experiments concentrated low-concentration iodine solution (100 mg L^{-1}) 18-fold in 24 h [35].

Within the domain of chemical processes, double displacement reactions, commonly recognized as metathesis reactions, have established a historical track record of successful utilization across diverse applications encompassing potassium nitrate synthesis, acid-base neutralization, and silver chloride production. Thampy et al. [244], utilized the electrochemical capabilities of salt metathesis through the application of electrodialysis techniques to produce potassium carbonate. Concurrently, Linnartz et al. [245] exhibited innovative synergy by coupling FCDI with electrochemical metathesis, introducing the potential for recuperation of valuable solutions through the salt metathesis process. This strategy entailed the sequential generation of concentrated magnesium sulfate (MgSO_4) solutions via a series of staged decomposition and recombination actions involving sodium sulfate (Na_2SO_4), magnesium chloride (MgCl_2), and potassium sulfate (K_2SO_4) solutions. During the decomposition phase, voltage-driven deionization transpired within discrete FCDI modules housing MgCl_2 and K_2SO_4 solutions. This led to the extraction of Mg^{2+} , K^+ , Cl^- , and SO_4^{2-} ions from their respective flow electrodes. Subsequent to this, the recombination stage entailed the amalgamation of these deionized constituents from their corresponding FCDI modules, resulting in the generation of concentrated solutions comprising potassium chloride (KCl) and MgSO_4 upon the application of reversed voltage. The investigation of the metathesis process was conducted via both a bifurcated two-step mode and a continuous semi-continuous mode. The purity of the recuperated solution was affected by the chosen mode and the precise operational parameters. Notably, the attained purity of the generated MgSO_4 solution showcased variability, spanning from 81.5 % to 99.9 % within the semi-continuous mode and from 82.0 % to 99.9 % in the two-step mode, wherein fluctuations in the conversion factor exerted influence. This underscores the inherent potential of FCDI technology in the establishment of concentrated chemical streams derived from initially dilute reagent solutions [245].

11.3. Contaminants removal

The recalcitrance and high toxicity of heavy metals make them a significant concern in water pollution [246]. Contemporary strategies consider these contaminants as potential valuable assets, amenable to recovery via technologies such as ion exchange, despite challenges linked to energy-intensive operations and intricate regeneration protocols [247]. For instance, Ma et al. [248] elucidated the efficacy of FCDI technology in successfully mitigating uranium contamination, achieving a remarkable reduction of uranium levels to below 10 $\mu\text{g L}^{-1}$ from an initial concentration of 40 $\mu\text{g L}^{-1}$. However, the efficacy of uranium removal was found to be compromised under specific operational conditions due to inherent complexities. Similarly, Zhang et al. [249] also utilized FCDI for the selective extraction of copper from saline solutions, yielding heightened removal efficiency for sodium ions in comparison to copper ions at elevated applied potentials. This phenomenon was attributed to the preferential electrosorption of copper ions onto distinct particles, facilitating their separation from the saline medium. Notably, the pronounced desorption of sodium ions played a

pivotal role in governing this distinctive separation mechanism.

Heavy metals resilience and toxicity drive water pollution concerns. New approaches treat them as valuable resources, recoverable through ion exchange, despite energy-demanding processes. FCDI proved effective and promising for removing uranium and copper, involving preferential electrosorption and notable sodium desorption.

12. Commercialization status and prospects of FCDI

Given the nascent stage of FCDI, the absence of pilot or full-scale testing and comprehensive industry data impedes the precise and dependable assessment of capital costs. This section presents preliminary insights on this matter, previously discussed in details by Zhang et al. [79]; however, extensive large-scale investigations are essential before this technology can be deemed ready for commercialization.

12.1. Economic feasibility

The economic feasibility of FCDI is influenced by several core components, each contributing to the overall system cost and operational efficiency. The primary cost driver is the Ion Exchange Membranes (IEMs), which account for >50 % of the total capital cost [250]. Recent advancements in membrane technology, driven by innovations in fuel cells and energy harvesting, are expected to lower the costs of IEMs from the current range of \$20 to \$100 per square meter to below \$10 per square meter, enhancing the economic viability of FCDI systems [251]. Another significant component is the current collectors, which, along with flowable electrodes, are currently expensive but offer room for cost reduction through optimized design and material selection [252]. The use of carbon nanomaterials for flow electrodes has shown promise in improving desalination performance while potentially reducing costs [253]. Additionally, the FCDI system includes costs associated with pretreatment and post-treatment facilities. Pretreatment costs, which involve components similar to those used in Electrodialysis brackish water desalination plants, typically account for about 10 % of the total capital cost. These components are essential for preparing the feed water for the deionization process, ensuring the removal of large particulates and conditioning the water to optimal pH and conductivity levels [79]. This cost includes equipment for filtration, pH adjustment, and chemical dosing systems necessary to protect the FCDI system from fouling and scaling, which can affect performance and longevity. Ancillary facilities, such as pumps, electrical infrastructure, piping, tanks, and sensors, also contribute significantly to the total investment [254]. Special pumps are necessary for the recirculation of slurry electrodes, adding to the costs [255]. These components must be robust and efficient to handle the specific requirements of FCDI systems [256]. For example, some studies present a dynamic reconfigurable charge pump that improves the overall power saving in multi-channel electrical stimulation [256]. It highlights the efficiency gains from using multiple charge pumps controlled by low-power frequency modulation. The energy consumption of FCDI systems is another critical factor [254]. The introduction of multiple cell pairs within the FCDI module can significantly reduce the energy demand by lowering the voltage drop per cell pair and the specific pumping energy required. This design not only enhances energy efficiency but also reduces operational costs. Given the complexity and significant costs associated with the components of FCDI systems, it is anticipated that the capital expenditure for FCDI treatment of brackish water will be substantially higher than that for ED or RO systems with comparable throughput [257]. Therefore, it is imperative to focus on simplifying the engineering design, developing cost-effective and durable materials such as IEMs and electrodes, and importantly, identifying specialized application areas like selective contaminant removal and element recovery to enhance the competitiveness of this technology compared to existing alternatives [258].

12.2. Current commercialization and scale-up status

The FCDI technology is currently in a developmental and optimization phase, with several pilot projects and small-scale implementations focusing on improving its feasibility and scalability. Most of the existing devices reported have limited treatment capacity ($<100 \text{ L d}^{-1}$) [79]. Scaling up is a pivotal step to facilitate the practical application of FCDI technology. Currently, two-dimensional CDI systems are predominantly utilized [259]; however, the exploration of innovative tubular FCDI designs is also being considered [260]. Recent studies have shown promising results in terms of efficiency and operational stability, indicating strong potential for broader commercial adoption in the near future. Frequent clogging of carbon particles within the flow channels has been anticipated, potentially compromising the long-term stability of the FCDI system [261]. To address this issue, Computational Fluid Dynamics (CFD) simulations can be employed to aid in the design and optimization of the unit's hydraulic conditions [262]. This approach aims to reduce instances of slurry electrode clogging by improving the flow dynamics of the electrodes and feed streams. From the manufacturing point of view, the use of 3D printing has emerged as a cost-effective and efficient method for the rapid, consistent, and well-documented production of reactor components [263]. For instance, 3D-printed electrodes with highly specific designs facilitate enhanced mass transport by providing high surface areas and promoting convective flow, which is crucial for improving the efficiency of electrochemical reactors [263]. Additionally, 3D-printing is particularly advantageous for creating complex parts such as spacers, current collectors, and metallic electrodes, which traditionally require lengthy and intricate manufacturing processes. Going further, the advent of 4D printing technology offers the potential for components to react to stimuli or evolve over time by integrating smart materials capable of sensing, adapting, or self-repairing under specific conditions [264].

Scaling up FCDI from laboratory settings to industrial applications faces several key challenges. One significant issue is the need for robust electrode materials that can maintain long-term stability and performance under industrial conditions. As the system scales, the flow-electrode design becomes critical for preventing clogging, ensuring uniform ion transport, and avoiding efficiency losses, all while handling large volumes. Additionally, the energy consumption increases significantly in industrial applications, requiring optimization of energy input while maintaining desalination efficiency. Innovations in flow management, such as optimizing flow rates and designing advanced electrode structures, are crucial for overcoming these bottlenecks. Furthermore, industrial applications necessitate scalable and cost-effective materials for electrodes, membranes, and other system components, which are not always commercially available or affordable. Another challenge is related to the power density and ion adsorption kinetics, which need to be managed carefully to maintain performance at scale.

13. Challenges and future perspectives

FCDI technologies hold immense potential, yet they also come with significant hurdles. Addressing these challenges are crucial prior to the successful integration of these technologies as efficient processes in the market.

Faradaic processes, encompassing anodic oxidation and cathodic reduction reactions, are inherent to the operation of CDI systems. These reactions are acknowledged for their detrimental impact on electrode efficacy, energy utilization efficiency, electrode durability, and the potential to induce pH fluctuations in the treated water. In contrast, in the context of FCDI, these electrochemical reactions are regarded with reduced concern. In pseudo-capacitive FCDI, carbon particles experience mild oxidative transformations during their transit through the anodic chamber, followed by compensatory reduction processes as they traverse the cathodic chamber [265,266]. Empirical observations over a

14-day period indicate minimal introduction of oxygen-containing functional groups, affirming the resilience of carbon particle electrodes in FCDI, with no discernible adverse impact on ion removal efficacy [265].

Presently, there exists a limited corpus of knowledge regarding flow channel configurations, carbon morphologies, and their impact on carbon charging phenomena within the domain of FCDI technology [79]. Significant challenges encompass the modest electronic conductivity inherent in flow electrodes, the propensity for channel obstructions, and the supplementary energy requisites during electrode slurry pumping and circulation. Nevertheless, a number of investigations have been documented, addressing these challenges in flow electrodes through strategies entailing Controlled Wetting Properties (CWP), Carbon Additives (CAs), and electrolyte concentration. Furthermore, the utilization of Computational Fluid Dynamics (CFD) simulations shows potential for refining unit design and optimizing the hydrodynamics of the flow electrode. Particular attention is given to mitigating the formation of stagnant areas. Moreover, certain studies have begun to quantify the influence of pH modulation within flow electrodes, scrutinizing its implications for the continuous functionality of the FCDI process, yet warranting further in-depth inquiry.

FCDI is still in its nascent stages, and the absence of pilot and full-scale tests, as well as industry data, hampers a reliable assessment of its capital costs. Extensive large-scale studies are imperative before contemplating commercialization. The comprehensive capital cost of an FCDI system encompasses expenses related to the FCDI device, including components like membranes, electrodes, current collectors, spacers, and casings. Notably, IEMs constitute a significant portion (>50 %) of the FCDI device cost, primarily dictated by the required membrane area for specific desalination tasks [267,268]. Ongoing advancements in fuel cells and reverse electrodialysis for energy harvesting have renewed investigations into IEMs, with expectations of cost reductions from the current range of \$20–100 per square meter to below \$10 per square meter in the near future [267–269]. Graphite current collectors and flowable electrodes, although costly at present, hold potential for cost reduction through optimized design and material selection. Additionally, components like pumps and tanks could represent significant fractions of the overall investment, especially when specialized equipment is needed, such as pumps for recirculating slurry electrodes in an FCDI plant. Immediate attention is needed to streamline engineering designs, develop cost-effective and durable materials (e.g., IEMs and electrodes), and, crucially, identify specialized application areas (e.g., selective contaminant removal and element recovery) to enhance the competitiveness of FCDI technology in comparison to alternative options [270,271].

While FCDI is still in its early stages, with a relatively brief history of less than eight years, the majority of devices discussed in existing literature exhibit limited treatment capacities, typically below 100 l per day. The crucial challenge lies in scaling up the technology for practical deployment. Presently, flat-sheet 2D systems predominate [272], although there is ongoing exploration of innovative tubular FCDI configurations [273]. A potential issue on the horizon is the anticipated occurrence of carbon particle clogging within the flow channels, potentially limiting the long-term stability of FCDI systems. Computational Fluid Dynamics (CFD) simulations can play a pivotal role in unit design and optimizing hydraulic conditions within flow-electrode systems. This includes minimizing the formation of dead zones (attributable, for instance, to the baffling effects of parallel ion exchange membranes) and mitigating the risk of slurry electrode clogs. In terms of manufacturing, recent advancements have made 3D printing an accessible and cost-effective method, particularly for fabricating cell components with high complexity, such as spacers, current collectors, and metallic electrodes. This shift to 3D printing is expected to significantly reduce costs and material wastage [274,275]. Ultimately, fostering robust research collaboration between industry and academia is of paramount importance and will be instrumental in expediting the scale-

up and commercialization of FCDI technology.

Concerning its possible uses, FCDI technology presents encouraging opportunities as an alternative method for water treatment, complementing desalination processes. FCDI holds relevance across various industrial domains, including the extraction of heavy metals, the recovery of nutrients, water softening, and the focused retrieval of valuable chemicals. This adaptability stems from FCDI's ability to selectively separate charged species. However, further strides are needed to enhance specificity towards particular ions of interest. This outlook capitalizes on the substantial innovations within materials, notably electrodes and IEMs. With the attainment of an elevated degree of ion selectivity, substantial reduction in volumetric energy consumption becomes a foreseeable outcome. Our envisagement extends to the emergence of large-scale FCDI implementations, facilitating the concentration of valuable resources such as ammonia, phosphate, and lithium, while concurrently affording the preferential elimination of perilous, ultra-dilute contaminants like heavy metals, fluoride, and perchlorate from water and wastewater matrices.

FCDI presents several challenges that must be addressed to enhance its development and practical application. A primary concern is the stability and uniformity of the flowable electrode slurry [276]. Maintaining consistent dispersion of carbon particles is crucial for optimal ion adsorption and system performance. Additionally, the integration of IEMs introduces complexities related to membrane fouling and degradation, which can impair ion selectivity and overall efficiency. The energy consumption of FCDI systems remains a significant challenge, particularly when compared to established desalination technologies like reverse osmosis. Achieving energy-efficient operation necessitates advancements in electrode materials and system design. Furthermore, scaling up FCDI for industrial applications poses challenges in maintaining performance consistency and managing operational costs. Addressing these issues requires comprehensive research into electrode slurry dynamics, membrane technology, energy optimization, and scalable system architectures.

14. Conclusion

FCDI emerges as a potent electro-driven desalination technology, showcasing inherent merits such as versatility across salinity gradients due to its nearly limitless electrosorption capacity, potential for continuous and stable operation, facile management of electrodes, and remarkable flow efficiency. Our all-encompassing review delves into contemporary advancements of FCDI, accentuating its principles, different configurations, operational modalities, characteristics of its flow-electrodes, characterizations, influencing factors, and ongoing applications with a special focus on ion selectivity. Essential definitions and performance metrics are introduced to enable unbiased assessment and contrast among diverse systems and separation scenarios. However, it is pertinent to note that this technology remains in its incipient stages, often marked by constrained treatment capacities (<100 L d⁻¹) in existing devices. To facilitate the transition of this promising technology into pragmatic utilization, conscientious attention is merited for some focal points. Therefore, challenges related to capital costs, scaling up, and electrode clogging need to be addressed. As the FCDI domain matures, the collaboration of intensive research efforts between industry and academia stands as a pivotal determinant, crucial for the successful transmutation of this captivating technology into tangible and sustainable applications.

Declaration of competing interest

We declare no interest.

Data availability

Data will be made available on request.

References

- [1] M.A. Shannon, et al., Science and technology for water purification in the coming decades, *Nature* 452 (2008) 301–310.
- [2] M. Elimelech, W.A. Phillip, The future of seawater desalination: energy, technology, and the environment, *Science* 333 (2011) 712–717.
- [3] M.W. Shahzad, M. Burhan, L. Ang, K.C. Ng, Energy-water-environment nexus underpinning future desalination sustainability, *Desalination* 413 (2017) 52–64.
- [4] C. Fritzmann, J. Löwenberg, T. Wintgens, T. Melin, State-of-the-art of reverse osmosis desalination, *Desalination* 216 (2007) 1–76.
- [5] D. Zarzo, D. Prats, Desalination and energy consumption. What can we expect in the near future? *Desalination* 427 (2018) 1–9.
- [6] M.E. Suss, et al., Water desalination via capacitive deionization: what is it and what can we expect from it? *Energ. Environ. Sci.* 8 (2015) 2296–2319.
- [7] G. Folaranmi, M. Bechelany, P. Sístat, M. Cretin, F. Zaviska, Towards electrochemical water desalination techniques: a review on capacitive deionization, membrane capacitive deionization and flow capacitive deionization, *Membranes* 10 (2020) 96.
- [8] S. Porada, R. Zhao, A. van der Wal, V. Presser, P.M. Biesheuvel, Review on the science and technology of water desalination by capacitive deionization, *Prog. Mater. Sci.* 58 (2013) 1388–1442.
- [9] J.G. Gamaethiralalage, et al., Recent advances in ion selectivity with capacitive deionization, *Energ. Environ. Sci.* 14 (2021) 1095–1120.
- [10] F.A. AlMarzooqi, A.A. Al Ghaferi, I. Saadat, N. Hilal, Application of capacitive deionisation in water desalination: a review, *Desalination* 342 (2014) 3–15.
- [11] E.N. Wood, et al., Electrochemical demineralization of water with carbon electrodes, in: U.S. Dept. of the Interior: For Sale by the Supt. Of Docs., U.S. Govt. Print. Off, Washington, D.C., 1965.
- [12] S. Porada, et al., Direct prediction of the desalination performance of porous carbon electrodes for capacitive deionization, *Energ. Environ. Sci.* 6 (2013) 3700–3712.
- [13] C. Zhang, D. He, J. Ma, W. Tang, T.D. Waite, Faradaic reactions in capacitive deionization (CDI) - problems and possibilities: a review, *Water Res.* 128 (2018) 314–330.
- [14] C.J. Gabelich, T.D. Tran, L.H.M. Suffet, Electrosorption of inorganic salts from aqueous solution using carbon aerogels, *Environ. Sci. Technol.* 36 (2002) 3010–3019.
- [15] M. Dahanayaka, et al., Corrugated graphene layers for sea water desalination using capacitive deionization, *Phys. Chem. Chem. Phys.* 19 (2017) 8552–8562.
- [16] H. Liu, J. Zhang, X. Xu, Q. Wang, A Polyoxometalate-based binder-free capacitive deionization electrode for highly Efficient Sea water desalination. *Chemistry – a European Journal* 26 (2020) 4403–4409.
- [17] Bharath, G. *Et al.* the fabrication of activated carbon and metal-carbide 2D framework-based asymmetric electrodes for the capacitive deionization of Cr(VI) ions toward industrial wastewater remediation. *Environ. Sci.: Water Res. Technol.* 6, 351–361 (2020).
- [18] Z.Y. Leong, H.Y. Yang, Capacitive deionization of divalent cations for water softening using functionalized carbon electrodes, *ACS Omega* 5 (2020) 2097–2106.
- [19] L. Wang, S. Lin, Mechanism of selective ion removal in membrane capacitive deionization for water softening, *Environ. Sci. Technol.* 53 (2019) 5797–5804.
- [20] P. Nie, et al., Highly efficient water softening by mordenite modified cathode in asymmetric capacitive deionization, *Sep. Purif. Technol.* 250 (2020) 117240.
- [21] S. Jeon, et al., Desalination via a new membrane capacitive deionization process utilizing flow-electrodes, *Energ. Environ. Sci.* 6 (2013) 1471–1475.
- [22] J. Ma, C. Zhai, F. Yu, Review of flow electrode capacitive deionization technology: research progress and future challenges, *Desalination* 564 (2023) 116701.
- [23] J. Ma, C. He, D. He, C. Zhang, T.D. Waite, Analysis of capacitive and electro-dialytic contributions to water desalination by flow-electrode CDI, *Water Res.* 144 (2018) 296–303.
- [24] K. Fang, et al., Recovering ammonia from municipal wastewater by flow-electrode capacitive deionization, *Chem. Eng. J.* 348 (2018) 301–309.
- [25] Y. Ha, et al., Continuous Lithium extraction from aqueous solution using flow-electrode capacitive deionization, *Energies* 12 (2019) 2913.
- [26] Y. Bian, X. Chen, L. Lu, P. Liang, Z.J. Ren, Concurrent nitrogen and phosphorus recovery using flow-electrode capacitive deionization, *ACS Sustainable Chem. Eng.* 7 (2019) 7844–7850.
- [27] K. Ramalingam, et al., Achieving high-quality freshwater from a self-sustainable integrated solar redox-flow desalination device, *Small* 17 (2021) 2100490.
- [28] A.M. Johnson, J. Newman, Desalting by means of porous carbon electrodes, *J. Electrochem. Soc.* 118 (1971) 510.
- [29] J.C. Farmer, D.V. Fix, G.V. Mack, R.W. Pekala, J.F. Poco, Capacitive deionization of NH₄ClO₄ solutions with carbon aerogel electrodes, *J. Appl. Electrochem.* 26 (1996) 1007–1018.
- [30] J.-B. Lee, K.-K. Park, H.-M. Eum, C.-W. Lee, Desalination of a thermal power plant wastewater by membrane capacitive deionization, *Desalination* 196 (2006) 125–134.
- [31] S. Jeon, et al., Desalination via a new membrane capacitive deionization process utilizing flow-electrodes, *Energ. Environ. Sci.* 6 (2013) 1471–1475.
- [32] P. Nativ, Y. Badash, Y. Gendel, New insights into the mechanism of flow-electrode capacitive deionization, *Electrochem. Commun.* 76 (2017) 24–28.
- [33] S. Dahiya, B.K. Mishra, Enhancing understandability and performance of flow electrode capacitive deionisation by optimizing configurational and operational parameters: a review on recent progress, *Sep. Purif. Technol.* 240 (2020) 116660.
- [34] J. Ma, et al., Water recovery rate in short-circuited closed-cycle operation of flow-electrode capacitive deionization (FCDI), *Environ. Sci. Technol.* 53 (2019) 13859–13867.
- [35] Q. Liu, B. Xie, D. Xiao, High efficient and continuous recovery of iodine in saline wastewater by flow-electrode capacitive deionization, *Sep. Purif. Technol.* 296 (2022) 121419.
- [36] L. Xu, et al., Selective recovery of phosphorus from synthetic urine using flow-electrode capacitive deionization (FCDI)-based technology, *ACS EST Water* 1 (2021) 175–184.
- [37] H. Deng, et al., Potential-mediated recycling of copper from brackish water by an electrochemical copper pump, *Adv. Sci.* 9 (2022) 2203189.
- [38] L. Baudino, C. Santos, C.F. Pirri, F. La Mantia, A. Lamberti, Recent advances in the Lithium recovery from water resources: from passive to electrochemical methods, *Advanced Science* 9 (2022) 2201380.
- [39] L. Xu, et al., Enhancing brackish water desalination using magnetic flow-electrode capacitive deionization, *Water Res.* 216 (2022) 118290.
- [40] H. Zhang, et al., Influence of ion exchange membrane arrangement on dual-channel flow electrode capacitive deionization: theoretical analysis and experimentations, *Desalination* 548 (2023) 116288.
- [41] M. Metzger, et al., Techno-economic analysis of capacitive and intercalative water deionization, *Energ. Environ. Sci.* 13 (2020) 1544–1560.
- [42] EST, Electrosorption Technology for Desalting, EST Water and Technologies Co., Ltd, China, 2009, p. 40 (in).
- [43] Y.-W. Chen, J.-F. Chen, C.-H. Lin, C.-H. Hou, Integrating a supercapacitor with capacitive deionization for direct energy recovery from the desalination of brackish water, *Appl. Energy* 252 (2019) 113417.
- [44] R.L. Zornitta, et al., Charge and potential balancing for optimized capacitive deionization using lignin-derived, Low-Cost Activated Carbon Electrodes. *ChemSusChem* 11 (2018) 2101–2113.
- [45] M. Lee, et al., Membrane capacitive deionization for low-salinity desalination in the reclamation of domestic wastewater effluents, *Chemosphere* 235 (2019) 413–422.
- [46] Energy consumption in membrane capacitive deionization for different water recoveries and flow rates, and comparison with reverse osmosis, *Desalination* 330 (2013) 35–41.
- [47] C. Tan, C. He, J. Fletcher, T.D. Waite, Energy recovery in pilot scale membrane CDI treatment of brackish waters, *Water Res.* 168 (2020) 115146.
- [48] H.J. Chung, J. Kim, D.I. Kim, G. Gwak, S. Hong, Feasibility study of reverse osmosis–flow capacitive deionization (RO-FCDI) for energy-efficient desalination using seawater as the flow-electrode aqueous electrolyte, *Desalination* 479 (2020) 114326.
- [49] C. Zhang, et al., Evaluation of long-term performance of a continuously operated flow-electrode CDI system for salt removal from brackish waters, *Water Res.* 173 (2020) 115580.
- [50] J. Ma, et al., Flow-electrode capacitive deionization (FCDI) scale-up using a membrane stack configuration, *Water Res.* 168 (2020) 115186.
- [51] M. Favaro, et al., Unravelling the electrochemical double layer by direct probing of the solid/liquid interface, *Nat. Commun.* 7 (2016) 12695.
- [52] Q. Li, et al., Faradaic electrodes open a new era for capacitive deionization, *Advanced Science* 7 (2020) 2002213.
- [53] N. Liu, et al., Ti₃C₂-MXene partially derived hierarchical 1D/2D TiO₂/Ti₃C₂ Heterostructure electrode for high-performance capacitive deionization, *Advanced Science* 10 (2023) 2204041.
- [54] J. Zhao, et al., Efficient and durable sodium, chloride-doped Iron oxide-hydroxide Nanohybrid-promoted capacitive deionization of saline water via synergetic Pseudocapacitive process, *Advanced Science* 9 (2022) 2201678.
- [55] H. Gang, et al., Surface redox pseudocapacitance boosting Fe/Fe₃C nanoparticles-encapsulated N-doped graphene-like carbon for high-performance capacitive deionization, *J. Colloid Interface Sci.* 638 (2023) 252–262.
- [56] Z. Liu, X. Shang, H. Li, Y. Liu, A brief review on high-performance capacitive deionization enabled by intercalation electrodes, *Glob. Chall* 5 (2021) 2000054.
- [57] J. Elisadiki, King'ondo, C. K., Performance of ion intercalation materials in capacitive deionization/electrochemical deionization: a review, *J. Electroanal. Chem.* 878 (2020) 114588.
- [58] M. Liang, et al., Combining battery-type and Pseudocapacitive charge storage in ag/Ti₃C₂Tx MXene electrode for capturing chloride ions with high capacitance and fast ion transport, *Advanced Science* 7 (2020) 2000621.
- [59] U. Samukaite-Bubniene, et al., Towards supercapacitors: cyclic voltammetry and fast Fourier transform electrochemical impedance spectroscopy based evaluation of polypyrrole electrochemically deposited on the pencil graphite electrode, *Colloids Surf. A Physicochem. Eng. Asp.* 610 (2021) 125750.
- [60] Analysis of the parameters of supercapacitors containing polypyrrole and its derivatives, *Archives of materials science and engineering* 120 (2023) 5–14.
- [61] L. Yuan, C. Wan, X. Ye, F. Wu, Facial synthesis of silver-incorporated conductive Polypyrrole submicron spheres for supercapacitors, *Electrochim. Acta* 213 (2016) 115–123.
- [62] M. Raicopol, A. Pruna, L. Pilan, Supercapacitance of single-walled carbon nanotubes-Polypyrrole composites, *J. Chem.* 2013 (2013) 367473.
- [63] J. Lee, S. Kim, C. Kim, J. Yoon, Hybrid capacitive deionization to enhance the desalination performance of capacitive techniques, *Energ. Environ. Sci.* 7 (2014) 3683–3689.
- [64] G.J. Doornbusch, J.E. Dykstra, P.M. Biesheuvel, M.E. Suss, Fluidized bed electrodes with high carbon loading for water desalination by capacitive deionization, *J. Mater. Chem. A* 4 (2016) 3642–3647.

- [65] S. Yang, et al., Flow-electrode capacitive deionization using an aqueous electrolyte with a high salt concentration, *Environ. Sci. Technol.* 50 (2016) 5892–5899.
- [66] S. Yang, et al., Stack design and operation for scaling up the capacity of flow-electrode capacitive deionization technology, *ACS Sustainable Chem. Eng.* 4 (2016) 4174–4180.
- [67] Y. Gendel, A.K.E. Rommerskirchen, O. David, M. Wessling, Batch mode and continuous desalination of water using flowing carbon deionization (FCDI) technology, *Electrochem. Commun.* 46 (2014) 152–156.
- [68] D. Moreno, M.C. Hatzell, Influence of feed-electrode concentration differences in flow-electrode Systems for Capacitive Deionization, *Ind. Eng. Chem. Res.* 57 (2018) 8802–8809.
- [69] S. Porada, et al., Carbon flow electrodes for continuous operation of capacitive deionization and capacitive mixing energy generation, *J. Mater. Chem. A* 2 (2014) 9313–9321.
- [70] F. Yang, J. Ma, X. Zhang, X. Huang, P. Liang, Decreased charge transport distance by titanium mesh-membrane assembly for flow-electrode capacitive deionization with high desalination performance, *Water Res.* 164 (2019) 114904.
- [71] A. Rommerskirchen, C.J. Linnartz, D. Müller, L.K. Willenberg, M. Wessling, Energy Recovery and Process Design in Continuous Flow-Electrode Capacitive Deionization Processes, *ACS Sustainable Chem. Eng.* 6 (2018) 13007–13015.
- [72] A. Rommerskirchen, Y. Gendel, M. Wessling, Single module flow-electrode capacitive deionization for continuous water desalination, *Electrochem. Commun.* 60 (2015) 34–37.
- [73] Zhang, X., Yang, F., Ma, J. & Liang, P. Effective removal and selective capture of copper from salty solution in flow electrode capacitive deionization. *Environ. Sci.: Water Res. Technol.* 6, 341–350 (2020).
- [74] S. Yang, et al., Plate-shaped graphite for improved performance of flow-electrode capacitive deionization, *J. Electrochem. Soc.* 164 (2017) E480.
- [75] J. Song, J. Ma, C. Zhang, C. He, T.D. Waite, Implication of non-electrostatic contribution to deionization in flow-electrode CDI: case study of nitrate removal from contaminated source waters, *Front. Chem.* 7 (2019).
- [76] H. Park, et al., Surface-modified spherical activated carbon for high carbon loading and its desalting performance in flow-electrode capacitive deionization, *RSC Adv.* 6 (2016) 69720–69727.
- [77] Y. Cho, et al., A novel three-dimensional desalination system utilizing honeycomb-shaped lattice structures for flow-electrode capacitive deionization, *Energ. Environ. Sci.* 10 (2017) 1746–1750.
- [78] K.S. Lee, et al., Membrane-spacer assembly for flow-electrode capacitive deionization, *Appl. Surf. Sci.* 433 (2018) 437–442.
- [79] C. Zhang, et al., Flow electrode capacitive deionization (FCDI): recent developments, environmental applications, and future perspectives, *Environ. Sci. Technol.* 55 (2021) 4243–4267.
- [80] C. He, J. Ma, C. Zhang, J. Song, T.D. Waite, Short-circuited closed-cycle operation of flow-electrode CDI for brackish water softening, *Environ. Sci. Technol.* 52 (2018) 9350–9360.
- [81] T. Kim, C.A. Gorski, B.E. Logan, Low energy desalination using battery electrode deionization, *Environ. Sci. Technol. Lett.* 4 (2017) 444–449.
- [82] J. Ma, et al., Energy recovery from the flow-electrode capacitive deionization, *J. Power Sources* 421 (2019) 50–55.
- [83] S. Yang, et al., Analysis of the desalting performance of flow-electrode capacitive deionization under short-circuited closed cycle operation, *Desalination* 424 (2017) 110–121.
- [84] S. Jeon, J. Yeo, S. Yang, J. Choi, D.K. Kim, Ion storage and energy recovery of a flow-electrode capacitive deionization process, *J. Mater. Chem. A* 2 (2014) 6378–6383.
- [85] J. Ma, et al., Carbon black flow electrode enhanced electrochemical desalination using single-cycle operation, *Environ. Sci. Technol.* 54 (2020) 1177–1185.
- [86] S. Jeon, et al., Novel reuse strategy in flow-electrode capacitive deionization with switch cycle operation to enhance desalination performance, *Environ. Sci. Technol. Lett.* 6 (2019) 739–744.
- [87] S.A. Hawks, et al., Quantifying the flow efficiency in constant-current capacitive deionization, *Water Res.* 129 (2018) 327–336.
- [88] A. Rommerskirchen, C.J. Linnartz, F. Egidi, S. Kendir, M. Wessling, Flow-electrode capacitive deionization enables continuous and energy-efficient brine concentration, *Desalination* 490 (2020) 114453.
- [89] C. Zhang, J. Ma, T.D. Waite, Ammonia-rich solution production from wastewaters using chemical-free flow-electrode capacitive deionization, *ACS Sustainable Chem. Eng.* 7 (2019) 6480–6485.
- [90] C. Zhang, J. Ma, D. He, T.D. Waite, Capacitive membrane stripping for Ammonia recovery (CapAmm) from dilute wastewaters, *Environ. Sci. Technol. Lett.* 5 (2018) 43–49.
- [91] C. Zhang, J. Ma, J. Song, C. He, T.D. Waite, Continuous Ammonia recovery from wastewaters using an integrated capacitive flow electrode membrane stripping system, *Environ. Sci. Technol.* 52 (2018) 14275–14285.
- [92] A.V. Delgado, M.L. Jiménez, G.R. Iglesias, S. Ahualli, Electrical double layers as ion reservoirs: applications to the deionization of solutions, *Curr. Opin. Colloid Interface Sci.* 44 (2019) 72–84.
- [93] G. Folaranmi, et al., Investigation of fine activated carbon as a viable flow electrode in capacitive deionization, *Desalination* 525 (2022) 115500.
- [94] F. Yu, et al., Carbon aerogel electrode for excellent dephosphorization via flow capacitive deionization, *Desalination* 528 (2022) 115614.
- [95] Y. Cho, et al., Flow-electrode capacitive deionization with highly enhanced salt removal performance utilizing high-aspect ratio functionalized carbon nanotubes, *Water Res.* 151 (2019) 252–259.
- [96] Y. Cai, X. Zhao, Y. Wang, D. Ma, S. Xu, Enhanced desalination performance utilizing sulfonated carbon nanotube in the flow-electrode capacitive deionization process, *Sep. Purif. Technol.* 237 (2020) 116381.
- [97] Z. Wang, et al., Enhanced desalination performance of a flow-electrode capacitive deionization system by adding vanadium redox couples and carbon nanotubes, *J. Phys. Chem. C* 125 (2021) 1234–1239.
- [98] Y. Sun, Y. Cheng, F. Yu, J. Ma, Enhanced salt removal performance using graphene-modified sodium vanadium Fluorophosphate in flow electrode capacitive deionization, *ACS Appl. Mater. Interfaces* 13 (2021) 53850–53858.
- [99] H.H.A. Mansoor, S.P. Thomas, S. Ramanujam, N. Mohan, B. Natesan, Synthesis of zinc oxide based nitrogen doped graphene oxide with polyaniline (ZnO/N-GO/PANI) flow electrode for desalination application using flow capacitive deionization, *J. Mater. Sci. Mater. Electron.* 34 (2023) 1081.
- [100] D.J. Ahirrao, N. Jha, Comparative study on the electrosorption properties of carbon fabric, functionalized multiwall carbon nanotubes and solar-reduced graphene oxide for flow through electrode based desalination studies, *Carbon* 152 (2019) 837–850.
- [101] D. Li, X.-A. Ning, Y. Li, J. Zhang, Nanoarchitected reduced graphene oxide composite C2N materials as flow electrodes to optimize desalination performance, *Environ. Sci. Nano* 7 (2020) 1980–1989.
- [102] A. Abbas, et al., Development of reduced graphene oxide from biowaste as an electrode material for vanadium redox flow battery, *Journal of Energy Storage* 41 (2021) 102848.
- [103] Y. Li, et al., Enhancing the electronic and ionic transport of flow-electrode capacitive deionization by hollow mesoporous carbon nanospheres, *Desalination* 550 (2023) 116381.
- [104] K.B. Hatzell, et al., Effect of oxidation of carbon material on suspension electrodes for flow electrode capacitive deionization, *E and T Contents* 49 (2015) 3040–3047.
- [105] C.-C. Hsu, Y.-H. Tu, Y.-H. Yang, J.-A. Wang, C.-C. Hu, Improved performance and long-term stability of activated carbon doped with nitrogen for capacitive deionization, *Desalination* 481 (2020) 114362.
- [106] Y. Liu, et al., Nitrogen-doped porous carbon spheres for highly efficient capacitive deionization, *Electrochim. Acta* 158 (2015) 403–409.
- [107] Y. Li, et al., Design of nitrogen-doped cluster-like porous carbons with hierarchical hollow nanoarchitecture and their enhanced performance in capacitive deionization, *Desalination* 430 (2018) 45–55.
- [108] D.K. Kohli, et al., Enhanced salt-adsorption capacity of ambient pressure dried carbon aerogel activated by CO₂ for capacitive deionization application, *Desalin. Water Treat.* 54 (2015) 2825–2831.
- [109] Y. Zhang, et al., Fabrication of porous graphene electrodes via CO₂ activation for the enhancement of capacitive deionization, *J. Colloid Interface Sci.* 536 (2019) 252–260.
- [110] A.S. Yasin, et al., Theoretical insight into the structure-property relationship of mixed transition metal oxides nanofibers doped in activated carbon and 3D graphene for capacitive deionization, *Chem. Eng. J.* 371 (2019) 166–181.
- [111] H. Yin, et al., Three-dimensional graphene/metal oxide nanoparticle hybrids for high-performance capacitive deionization of saline water, *Adv. Mater.* 25 (2013) 6270–6276.
- [112] J. Yang, L. Zou, H. Song, Z. Hao, Development of novel MnO₂/nanoporous carbon composite electrodes in capacitive deionization technology, *Desalination* 276 (2011) 199–206.
- [113] B. Kastening, W. Schiel, M. Henschel, Electrochemical polarization of activated carbon and graphite powder suspensions: part I. Capacity of suspensions and polarization dynamics, *J. Electroanal. Chem. Interfacial Electrochem.* 191 (1985) 311–328.
- [114] P. Liang, et al., Optimized desalination performance of high voltage flow-electrode capacitive deionization by adding carbon black in flow-electrode, *Desalination* 420 (2017) 63–69.
- [115] Y. Cho, et al., Flow-electrode capacitive deionization with highly enhanced salt removal performance utilizing high-aspect ratio functionalized carbon nanotubes, *Water Res.* 151 (2019) 252–259.
- [116] J. Ma, et al., Development of redox-active flow electrodes for high-performance capacitive deionization, *Environ. Sci. Technol.* 50 (2016) 13495–13501.
- [117] B. Anasori, M.R. Lukatskaya, Y. Gogotsi, 2D metal carbides and nitrides (MXenes) for energy storage, *Nat. Rev. Mater.* 2 (2017) 1–17.
- [118] Y.-Y. Peng, et al., All-MXene (2D titanium carbide) solid-state microsupercapacitors for on-chip energy storage, *Energ. Environ. Sci.* 9 (2016) 2847–2854.
- [119] Z. Guo, et al., High electrical conductivity 2D MXene serves as additive of perovskite for efficient solar cells, *Small* 14 (2018) 1802738.
- [120] G. Hwang, et al., Analysis of stability behavior of carbon black nanoparticles in ecotoxicological media: hydrophobic and steric effects, *Colloids Surf. A Physicochem. Eng. Asp.* 554 (2018) 306–316.
- [121] A.N. Shocron, M.E. Suss, The effect of surface transport on water desalination by porous electrodes undergoing capacitive charging, *J. Phys. Condens. Matter* 29 (2017) 084003.
- [122] K.B. Hatzell, et al., Direct observation of active material interactions in flowable electrodes using X-ray tomography, *Faraday Discuss.* 199 (2017) 511–524.
- [123] A. Alekseev, T.H. Wu, L.G.J. van der Ven, R.A.T.M. van Benthem, G. de With, Global and local conductivity in percolating crosslinked carbon black/epoxy-amine composites, *J. Mater. Sci.* 55 (2020) 8930–8939.
- [124] R. Gloukhovski, M.E. Suss, Measurements of the electric conductivity of MWCNT suspension electrodes with varying potassium bromide electrolyte ionic strength, *J. Electrochem. Soc.* 167 (2020) 020528.

- [125] R. de Levie, On porous electrodes in electrolyte solutions: I, Capacitance effects, *Electrochimica Acta* 8 (1963) 751–780.
- [126] M. Foygel, R.D. Morris, D. Anez, S. French, V.L. Sobolev, Theoretical and computational studies of carbon nanotube composites and suspensions: electrical and thermal conductivity, *Phys. Rev. B* 71 (2005) 104201.
- [127] C.R. Dennison, et al., Effects of flow cell design on charge percolation and storage in the carbon slurry electrodes of electrochemical flow capacitors, *J. Power Sources* 247 (2014) 489–496.
- [128] F. Yang, et al., Flow-electrode capacitive deionization: a review and new perspectives, *Water Res.* 200 (2021) 117222.
- [129] L. Gao, X. Zhou, Y. Ding, Effective thermal and electrical conductivity of carbon nanotube composites, *Chem. Phys. Lett.* 434 (2007) 297–300.
- [130] B.J. Sabacky, J.W. Evans, The electrical conductivity of fluidized bed electrodes—its significance and some experimental measurements, *Metall. Trans. B* 8 (1977) 5–13.
- [131] L. Daintree, S. Biggs, Particle-particle interactions: the link between aggregate properties and rheology, *Part. Sci. Technol.* 28 (2010) 404–425.
- [132] C.J.E. Farina, K.B. Oldham, The diffuse charge region in thin layer cells, *J. Electroanal. Chem. Interfacial Electrochem.* 81 (1977) 21–36.
- [133] E.B. Halfon, M.E. Suss, Measurements of the electric conductivity of an electrode as it transitions between static and flowable modes, *Electrochem. Commun.* 99 (2019) 61–64.
- [134] H. Kim, et al., Electrochemical analysis of high-performance flow-electrode capacitive mixing (F-CapMix) under different operating conditions, *ACS Sustain. Chem. Eng.* 9 (2021) 9199–9208.
- [135] A. Rommerskirchen, et al., Unraveling charge transport in carbon flow-electrodes: performance prediction for desalination applications, *Carbon* 145 (2019) 507–520.
- [136] T.J. Petek, N.C. Hoyt, R.F. Savinell, J.S. Wainright, Characterizing slurry electrodes using electrochemical impedance spectroscopy, *J. Electrochem. Soc.* 163 (2015) A5001.
- [137] C. Portet, G. Yushin, Y. Gogotsi, Effect of carbon particle size on electrochemical performance of EDLC, *J. Electrochem. Soc.* 155 (2008) A531.
- [138] X. Gao, et al., Complementary surface charge for enhanced capacitive deionization, *Water Res.* 92 (2016) 275–282.
- [139] M. Boota, et al., Activated carbon spheres as a Flowable electrode in electrochemical flow capacitors, *J. Electrochem. Soc.* 161 (2014) A1078.
- [140] Y. Tang, et al., Design of uniform hollow carbon Nanoarchitectures: different capacitive deionization between the hollow Shell thickness and cavity size, *Advanced Science* 10 (2023) 2206960.
- [141] K. Tang, S. Yiacoumi, Y. Li, C. Tsouris, Enhanced water desalination by increasing the Electroconductivity of carbon powders for high-performance flow-electrode capacitive deionization, *ACS Sustainable Chem. Eng.* 7 (2019) 1085–1094.
- [142] W. Tang, et al., Various cell architectures of capacitive deionization: recent advances and future trends, *Water Res.* 150 (2019) 225–251.
- [143] V. Presser, et al., The electrochemical flow capacitor: a new concept for rapid energy storage and recovery, *Adv. Energy Mater.* 2 (2012) 895–902.
- [144] X. Xu, M. Wang, Y. Liu, T. Lu, L. Pan, Ultrahigh desalination performance of asymmetric flow-electrode capacitive deionization device with an improved operation voltage of 1.8 V, *ACS sustainable, Chem. Eng.* 5 (2017) 189–195.
- [145] K.Y. Choo, C.Y. Yoo, M.H. Han, D.K. Kim, Electrochemical analysis of slurry electrodes for flow-electrode capacitive deionization, *J. Electroanal. Chem.* 806 (2017) 50–60.
- [146] N.H.J. Freire, C.J. Linnartz, L.A. Montoro, V.S.T. Ciminelli, M. Wessling, Flow electrode capacitive deionization with iron-based redox electrolyte, *Desalination* 578 (2024) 117313.
- [147] S. Mani, et al., New insights into the performance analysis of flow-electrode capacitive deionization using ferri/ferrocyanide redox couples for continuous water desalination, *Chem. Eng. J.* 480 (2024) 147887.
- [148] C. Shi, et al., Process model for flow-electrode capacitive deionization for energy consumption estimation and system optimization, *Water Res.* 230 (2023) 119517.
- [149] L. Wang, C. Zhang, C. He, T.D. Waite, S. Lin, Equivalent film-electrode model for flow-electrode capacitive deionization: experimental validation and performance analysis, *Water Res.* 181 (2020) 115917.
- [150] G. Shen, et al., Mechanism of ball milled activated carbon in improving the desalination performance of flow- and fixed-electrode in capacitive deionization desalination, *Front. Environ. Sci. Eng.* 17 (2022) 64.
- [151] D.J. Ahirrao, S. Tambat, A.B. Pandit, N. Jha, Sweet-lime-peels-derived activated-carbon-based electrode for highly efficient supercapacitor and flow-through water desalination, *ChemistrySelect* 4 (2019) 2610–2625.
- [152] M. Tauk, et al., Influence of particle size distribution on carbon-based flowable electrode viscosity and desalination efficiency in flow electrode capacitive deionization, *Sep. Purif. Technol.* 306 (2023) 122666.
- [153] A. Alsultan, et al., Surface-treated carbon black for durable, efficient, continuous flow electrode capacitive deionization, *Sep. Purif. Technol.* 313 (2023) 123444.
- [154] T. Chen, L. Xu, S. Wei, X. Tang, H. Chen, Enhanced ammonia-rich solution production and electrode separation using magnetic nickel-loaded carbon black in flow-electrode electrochemical deionization (FEED), *Desalination* 544 (2022) 116152.
- [155] L. Hadebe, Z. Cele, B. Gumbi, Properties of porous carbon electrode material derived from biomass of coffee waste grounds for capacitive deionization, *Materials Today: Proceedings* 56 (2022) 2178–2183.
- [156] C. Tsouris, et al., Mesoporous carbon for capacitive deionization of saline water, *Environ. Sci. Technol.* 45 (2011) 10243–10249.
- [157] Y. Liu, et al., Metal–organic framework-derived porous carbon polyhedra for highly efficient capacitive deionization, *Chem. Commun.* 51 (2015) 12020–12023.
- [158] Y. Li, et al., Facile dual doping strategy via carbonization of covalent organic frameworks to prepare hierarchically porous carbon spheres for membrane capacitive deionization, *Chem. Commun.* 54 (2018) 14009–14012.
- [159] S. Porada, et al., Capacitive deionization using biomass-based microporous salt-templated heteroatom-doped carbons, *ChemSusChem* 8 (2015) 1867–1874.
- [160] S. Kim, et al., Lithium recovery from brine using a λ -MnO₂/activated carbon hybrid supercapacitor system, *Chemosphere* 125 (2015) 50–56.
- [161] B.W. Byles, D.A. Cullen, K.L. More, E. Pomerantseva, Tunnel structured manganese oxide nanowires as redox active electrodes for hybrid capacitive deionization, *Nano Energy* 44 (2017).
- [162] K.B. Hatzell, M. Boota, Y. Gogotsi, Materials for suspension (semi-solid) electrodes for energy and water technologies, *Chem. Soc. Rev.* 44 (2015) 8664–8687.
- [163] H. Park, J. Choi, S. Yang, M.H. Han, D.K. Kim, Electrochemical characterization of electrolyte-filled porous carbon materials for electrosorption process, *J. Electroanal. Chem.* 801 (2017) 179–184.
- [164] H. Huang, et al., Anion-/cationic compounds enhance the dispersion of flow electrodes to obtain high capacitive deionization performance, *Desalination* 515 (2021) 115182.
- [165] J. Lim, Y.-U. Shin, S. Hong, Enhanced capacitive deionization using a biochar-integrated novel flow-electrode, *Desalination* 528 (2022) 115636.
- [166] J. Ma, et al., Enhanced charge efficiency and electrode separation utilizing magnetic carbon in flow electrode capacitive deionization, *ACS EST Eng.* 1 (2021) 340–347.
- [167] T. Wang, Z. Zhang, Z. Gu, C. Hu, J. Qu, Electron transfer of activated carbon to anode excites and regulates desalination in flow electrode capacitive deionization, *Environ. Sci. Technol.* 57 (2023) 2566–2574.
- [168] Sebok, E. B. & Taylor, R. L. Carbon Blacks. In *Encyclopedia of Materials: Science and Technology* (eds. Buschow, K. H. J. et al.) 902–906 (Elsevier, Oxford, 2001). doi: <https://doi.org/10.1016/B0-08-043152-6/00173-X>.
- [169] S. Nadakatti, M. Tendulkar, M. Kadam, Use of mesoporous conductive carbon black to enhance performance of activated carbon electrodes in capacitive deionization technology, *Desalination* 268 (2011) 182–188.
- [170] J. Kang, O.L. Li, N. Saito, Hierarchical meso–macro structure porous carbon black as electrode materials in Li–air battery, *J. Power Sources* 261 (2014) 156–161.
- [171] X. Sun, et al., Effects of carbon black on the electrochemical performances of SiO_x anode for lithium-ion capacitors, *J. Power Sources* 499 (2021) 229936.
- [172] S. Iijima, Helical microtubules of graphitic carbon, *Nature* 354 (1991) 56–58.
- [173] Y. Cheng, et al., A review of modification of carbon electrode material in capacitive deionization, *RSC Adv.* 9 (2019) 24401–24419.
- [174] P. Clemens, X. Wei, B.L. Wilson, R.L. Thomas, Anatase titanium dioxide coated Single Wall carbon nanotubes manufactured by Sonochemical-hydrothermal technique, *Open Journal of Composite Materials* 3 (2013) 21–32.
- [175] W. Shi, et al., High-performance capacitive deionization via manganese oxide-coated, vertically aligned carbon nanotubes, *Environ. Sci. Technol. Lett.* 5 (2018) 692–700.
- [176] Y. Liu, et al., Review on carbon-based composite materials for capacitive deionization, *RSC Adv.* 5 (2015) 15205–15225.
- [177] P. Liu, et al., Graphene-based materials for capacitive deionization, *J. Mater. Chem. A* 5 (2017) 13907–13943.
- [178] Z. Peng, D. Zhang, L. Shi, T. Yan, High performance ordered mesoporous carbon/carbon nanotube composite electrodes for capacitive deionization, *J. Mater. Chem.* 22 (2012) 6603–6612.
- [179] K.-Y. Chen, Y.-Y. Shen, D.-M. Wang, C.-H. Hou, Carbon nanotubes/activated carbon hybrid as a high-performance suspension electrode for the electrochemical desalination of wastewater, *Desalination* 522 (2022) 115440.
- [180] S.B. Rauer, et al., PEDOT:PSS-CNT composite particles overcome contact resistances in slurry electrodes for flow-electrode capacitive deionization, *Adv. Funct. Mater.* 33 (2023) 2303606.
- [181] F. Li, et al., Boosting the specific surface area of hierarchical porous carbon aerogel through the multiple roles of the catalyst for high-performance supercapacitors, *ChemElectroChem* 4 (2017) 3119–3125.
- [182] M. Huang, F. Li, F. Dong, Y.X. Zhang, L.L. Zhang, MnO₂-based nanostructures for high-performance supercapacitors, *J. Mater. Chem. A* 3 (2015) 21380–21423.
- [183] L.M. Chang, X.Y. Duan, W. Liu, Preparation and electrosorption desalination performance of activated carbon electrode with titania, *Desalination* 270 (2011) 285–290.
- [184] K. Tang, T.Z.X. Hong, L. You, K. Zhou, Carbon–metal compound composite electrodes for capacitive deionization: synthesis, development and applications, *J. Mater. Chem. A* 7 (2019) 26693–26743.
- [185] H.A. Maddah, M.A. Shihon, Activated carbon cloth for desalination of brackish water using capacitive deionization, *Desalin. Water Treat.* 17–36 (2018).
- [186] S. Ramanavicius, A. Ramanavicius, Insights in the application of stoichiometric and non-stoichiometric titanium oxides for the Design of Sensors for the determination of gases and VOCs (TiO_{2-x} and TinO_{2n-1} vs. TiO₂), *Sensors (Basel)* 20 (2020) 6833.
- [187] G. Folaranni, et al., Synthesis and characterization of activated carbon co-mixed electrosup titanium oxide nanofibers as flow electrode in capacitive deionization, *Materials* 14 (2021) 6891.
- [188] C. Kim, J. Lee, S. Kim, J. Yoon, TiO₂ sol–gel spray method for carbon electrode fabrication to enhance desalination efficiency of capacitive deionization, *Desalination* 342 (2014) 70–74.

- [189] A.S. Yasin, D. Kim, Hyun, & Lee, K., One-pot synthesis of activated carbon decorated with ZnO nanoparticles for capacitive deionization application, *J. Alloys Compd.* 870 (2021) 159422.
- [190] B. Gao, Acidified activated carbon with enhanced electrochemical performance for supercapacitors, *Int. J. Electrochem. Sci.* 12 (2017) 116–127.
- [191] C. Liu, et al., Improved capacitive energy storage via surface functionalization of activated carbon as cathodes for lithium ion capacitors, *Carbon* 109 (2016) 163–172.
- [192] J. Zhou, et al., Ultrahigh volumetric capacitance and cyclic stability of fluorine and nitrogen co-doped carbon microspheres, *Nat. Commun.* 6 (2015) 8503.
- [193] H. Chen, et al., Nitrogen doping effects on the physical and chemical properties of mesoporous carbons, *J. Phys. Chem. C* 117 (2013) 8318–8328.
- [194] H. Zhang, et al., Boosting pseudocapacitive charge storage in in situ functionalized carbons with a high surface area for high-energy asymmetric supercapacitors, *Sustainable Energy Fuels* 2 (2018) 2314–2324.
- [195] Z. Ding, V. Trouillet, S. Dsoke, Are functional groups beneficial or harmful on the electrochemical performance of activated carbon electrodes? *J. Electrochem. Soc.* 166 (2019) A1004.
- [196] Y.-H. Liu, H.-C. Hsi, K.-C. Li, C.-H. Hou, Electrodeposited manganese dioxide/activated carbon composite as a high-performance electrode material for capacitive deionization, *ACS Sustainable Chem. Eng.* 4 (2016) 4762–4770.
- [197] P. Singh, K. Pal, Activated carbon-polyaniline composite active material slurry electrode for high capacitance, improved rheological performance electrochemical flow capacitor, *Electrochim. Acta* 354 (2020) 136719.
- [198] J. Xiong, et al., Regulating surface wettability and charge density of porous carbon particles by in situ growth of polyaniline for constructing an efficient electrical percolation network in flow-electrode capacitive deionization, *Langmuir* 38 (2022) 12263–12272.
- [199] R. Chen, et al., Enhanced desalination performances by using porous polyaniline-activated carbon composite flow-electrodes in capacitive deionization system, *Desalination* 557 (2023) 116568.
- [200] R. Chen, et al., A newly designed graphite-polyaniline composite current collector to enhance the performance of flow electrode capacitive deionization, *Chem. Eng. J.* 435 (2022) 134845.
- [201] R. Chen, et al., Enhanced desalination performances by using porous polyaniline-activated carbon composite flow-electrodes in capacitive deionization system, *Desalination* 557 (2023) 116568.
- [202] W. Zhang, et al., Selection and optimization of carbon-based electrode materials for flow-electrode capacitive deionization, *Sep. Purif. Technol.* 315 (2023) 123649.
- [203] R. Chen, et al., A novel two-stage continuous capacitive deionization system with connected flow electrode and freestanding electrode, *Chem. Eng. J.* 491 (2024) 152133.
- [204] F. Yu, et al., Carbon aerogel electrode for excellent dephosphorization via flow capacitive deionization, *Desalination* 528 (2022) 115614.
- [205] Y. Li, et al., Enhancing the electronic and ionic transport of flow-electrode capacitive deionization by hollow mesoporous carbon nanospheres, *Desalination* 550 (2023) 116381.
- [206] L. Yan, et al., A hexagonal honeycomb-shaped Flow Channel for high-efficient desalination and Flowability in flow-electrode capacitive deionization, *ACS EST Water* 3 (2023) 2753–2764.
- [207] V. Bakola, et al., Development of composite nanostructured electrodes for water desalination via membrane capacitive deionization, *Macromol. Rapid Commun.* 45 (2024) 2300640.
- [208] J. Lim, H. Lee, S. Lee, S. Hong, Capacitive deionization incorporating a fluidic MOF-CNT electrode for the high selective extraction of lithium, *Desalination* 578 (2024) 117403.
- [209] L. Xu, L. Tang, S. Peng, Y. Mao, D. Wu, Magnetic array for efficient and stable flow-electrode capacitive deionization, *Chem. Eng. J.* 446 (2022) 137415.
- [210] J.C. Ehring, A.V. Mizrak, L. Agartan, B. Akizim, E.C. Kumbur, A new static mixer concept for enhanced desalination performance in flow-electrode capacitive deionization (FCDI) systems, *Desalination* 566 (2023) 116887.
- [211] R. Chen, et al., Enhanced desalination performances by using porous polyaniline-activated carbon composite flow-electrodes in capacitive deionization system, *Desalination* 557 (2023) 116568.
- [212] A. Alsultan, et al., Surface-treated carbon black for durable, efficient, continuous flow electrode capacitive deionization, *Sep. Purif. Technol.* 313 (2023) 123444.
- [213] T.-Y. Ying, K.-L. Yang, S. Yiacoimi, C. Tsouris, Electrosorption of ions from aqueous solutions by nanostructured carbon aerogel, *J. Colloid Interface Sci.* 250 (2002) 18–27.
- [214] C.-H. Hou, C. Liang, S. Yiacoimi, S. Dai, C. Tsouris, Electrosorption capacitance of nanostructured carbon-based materials, *J. Colloid Interface Sci.* 302 (2006) 54–61.
- [215] S. Choi, B. Chang, J.H. Kang, M.S. Diallo, J.W. Choi, Energy-efficient hybrid FCDI-NF desalination process with tunable salt rejection and high water recovery, *J. Membr. Sci.* 541 (2017) 580–586.
- [216] R.K. Nagarale, G.S. Gohil, V.K. Shahi, Recent developments on ion-exchange membranes and electro-membrane processes, *Adv. Colloid Interface Sci.* 119 (2006) 97–130.
- [217] A. Rommerskirchen, B. Ohs, K.A. Hepp, R. Femmer, M. Wessling, Modeling continuous flow-electrode capacitive deionization processes with ion-exchange membranes, *J. Membr. Sci.* 546 (2018) 188–196.
- [218] Zhao, R., Biesheuvel, P. M. & Wal, A. van der. Energy consumption and constant current operation in membrane capacitive deionization. *Energy Environ. Sci.* 5, 9520–9527 (2012).
- [219] H. Li, L. Zou, Ion-exchange membrane capacitive deionization: a new strategy for brackish water desalination, *Desalination* 275 (2011) 62–66.
- [220] E. Avraham, M. Noked, A. Soffer, D. Aurbach, The feasibility of boron removal from water by capacitive deionization, *Electrochim. Acta* 56 (2011) 6312–6317.
- [221] M. Wang, et al., Capacitive neutralization deionization with flow electrodes, *Electrochim. Acta* 216 (2016) 211–218.
- [222] I. Cohen, E. Avraham, Y. Bouhadana, A. Soffer, D. Aurbach, Long term stability of capacitive de-ionization processes for water desalination: the challenge of positive electrodes corrosion, *Electrochim. Acta* 106 (2013) 91–100.
- [223] H. Strathmann, Electrodialysis, a mature technology with a multitude of new applications, *Desalination* 264 (2010) 268–288.
- [224] S.-J. Seo, et al., Investigation on removal of hardness ions by capacitive deionization (CDI) for water softening applications, *Water Res.* 44 (2010) 2267–2275.
- [225] J.-S. Park, J.-H. Song, K.-H. Yeon, S.-H. Moon, Removal of hardness ions from tap water using electromembrane processes, *Desalination* 202 (2007) 1–8.
- [226] Bridges, S. & Robinson, L. Chapter 1 - Rheology, in *A Practical Handbook for Drilling Fluids Processing* (eds. Bridges, S. & Robinson, L.) 3–26 (Gulf Professional Publishing, 2020). doi:https://doi.org/10.1016/B978-0-12-821341-4.00001-4.
- [227] Meister, F. et al. 6 - Additional clinical applications, in *Artificial Intelligence for Computational Modeling of the Heart* (eds. Mansi, T., Passerini, T. & Comaniciu, D.) 183–210 (Academic Press, 2020). doi:https://doi.org/10.1016/B978-0-12-817594-1.00017-6.
- [228] T.M. Khoi, et al., Selective and continuous ion recovery using flow electrode capacitive deionization with polymer multilayers functionalized ion exchange membrane, *Desalination* 558 (2023) 116626.
- [229] L. Xu, et al., Selective recovery of phosphorus from synthetic urine using flow-electrode capacitive deionization (FCDI)-based technology, *ACS EST Water* 1 (2021) 175–184.
- [230] C. Zhang, J. Ma, T.D. Waite, Ammonia-rich solution production from wastewaters using chemical-free flow-electrode capacitive deionization, *ACS Sustainable Chem. Eng.* 7 (2019) 6480–6485.
- [231] H. Jiang, et al., Effective fluoride removal from brackish groundwaters by flow-electrode capacitive deionization (FCDI) under a continuous-flow mode, *Sci. Total Environ.* 804 (2022) 150166.
- [232] Y. Dong, et al., Effective and continuous removal of Cr(VI) from brackish wastewater by flow-electrode capacitive deionization (FCDI), *J. Clean. Prod.* 326 (2021) 129417.
- [233] L. Lin, et al., Selective ammonium removal from synthetic wastewater by flow-electrode capacitive deionization using a novel K₂Ti₂O₅-activated carbon mixture electrode, *Environ. Sci. Technol.* 54 (2020) 12723–12731.
- [234] Y. He, et al., Highly efficient and selective extraction of phosphorus from wastewater as vivianite in a strategically operated four-chamber flow electrode capacitive deionization, *Desalination* 544 (2022) 116089.
- [235] W. Xing, et al., Versatile applications of capacitive deionization (CDI)-based technologies, *Desalination* 482 (2020) 114390.
- [236] S. Hand, X. Shang, J.S. Guest, K.C. Smith, R.D. Cusick, Global sensitivity analysis to characterize operational limits and prioritize performance goals of capacitive deionization technologies, *Environ. Sci. Technol.* 53 (2019) 3748–3756.
- [237] J.R. Mihelcic, et al., Accelerating innovation that enhances resource recovery in the wastewater sector: advancing a National Testbed Network, *Environ. Sci. Technol.* 51 (2017) 7749–7758.
- [238] E. Zhang, B. Wang, Q. Wang, S. Zhang, B. Zhao, Ammonia–nitrogen and orthophosphate removal by immobilized *Senedesmus* sp. isolated from municipal wastewater for potential use in tertiary treatment, *Bioresour. Technol.* 99 (2008) 3787–3793.
- [239] F. Zhou, et al., Electro-synthesis of ammonia from nitrogen at ambient temperature and pressure in ionic liquids, *Energy Environ. Sci.* 10 (2017) 2516–2520.
- [240] Y. Zhang, I. Angelidaki, Recovery of ammonia and sulfate from waste streams and bioenergy production via bipolar bioelectrodialysis, *Water Res.* 85 (2015) 177–184.
- [241] Yoon, H. *Et al.* Lithium-selective hybrid capacitive deionization system with a g-coated carbon electrode and stop-flow operation. *Environ. Sci.: Water Res. Technol.* 9, 500–507 (2023).
- [242] C. Yu, L. Xu, Y. Mao, Y. Zong, D. Wu, Efficient recovery of carboxylates from the effluents treated by advanced oxidation processes using flow-electrode capacitive deionization in short-circuited closed-cycle operation, *Sep. Purif. Technol.* 275 (2021) 119151.
- [243] S. Han, et al., Efficient bicarbonate removal and recovery of ammonium bicarbonate as CO₂ utilization using flow-electrode capacitive deionization, *Chem. Eng. J.* 431 (2022) 134233.
- [244] K. T. S., S. J. B. & P. G. K., Preparation of potassium carbonate by electrodialysis technique, *Indian J. Technol.* 23 (1985) 454–457.
- [245] C.J. Linnartz, A. Rommerskirchen, M. Wessling, Y. Gendel, Flow-electrode capacitive deionization for double displacement reactions, *ACS Sustainable Chem. Eng.* 5 (2017) 3906–3912.
- [246] F. Fu, Q. Wang, Removal of heavy metal ions from wastewaters: a review, *J. Environ. Manage.* 92 (2011) 407–418.
- [247] L. Zhang, Y. Wu, X. Qu, Z. Li, J. Ni, Mechanism of combination membrane and electro-winning process on treatment and remediation of Cu²⁺ polluted water body, *J. Environ. Sci.* 21 (2009) 764–769.
- [248] J. Ma, et al., Flow-electrode CDI removes the uncharged Ca–UO₂–CO₃ ternary complex from brackish potable groundwater: complex dissociation, transport, and sorption, *Environ. Sci. Technol.* 53 (2019) 2739–2747.

- [249] Zhang, X., Yang, F., Ma, J. & Liang, P. Effective removal and selective capture of copper from salty solution in flow electrode capacitive deionization. *Environ. Sci.: Water Res. Technol.* 6, 341–350 (2020).
- [250] P. Zuo, et al., Ion exchange membranes: constructing and tuning ion transport channels, *Adv. Funct. Mater.* 32 (2022) 2207366.
- [251] A. Rommelskirchen, C.J. Linnartz, F. Egidi, S. Kendir, M. Wessling, Flow-electrode capacitive deionization enables continuous and energy-efficient brine concentration, *Desalination* 490 (2020) 114453.
- [252] K. Alsaikhan, et al., Carbon material-based flow-electrode capacitive deionization for continuous water desalination, *Processes* 11 (2023) 195.
- [253] M. Tauk, et al., Recent advances in capacitive deionization: a comprehensive review on electrode materials, *J. Environ. Chem. Eng.* 11 (2023) 111368.
- [254] A. Rommelskirchen, C.J. Linnartz, D. Müller, L.K. Willenberg, M. Wessling, Energy Recovery and Process Design in Continuous Flow-Electrode Capacitive Deionization Processes, *ACS Sustainable Chem. Eng.* 6 (2018) 13007–13015.
- [255] M. Tauk, et al., Influence of particle size distribution on carbon-based flowable electrode viscosity and desalination efficiency in flow electrode capacitive deionization, *Sep. Purif. Technol.* 306 (2023) 122666.
- [256] M.W. Kim, J.J. Kim, Energy-efficient fast-transient dynamic reconfigurable charge pump for Multi-Channel electrical stimulation, *IEEE Trans Circuits Syst II Express Briefs* 70 (2023) 2146–2150.
- [257] W. He, et al., Field demonstration of a cost-optimized solar powered electro dialysis reversal desalination system in rural India, *Desalination* 476 (2020) 114217.
- [258] C. Huyskens, J. Helsen, W.J. de Groot, A. de Haan, Cost evaluation of large-scale membrane capacitive deionization for biomass hydrolysate desalination, *Sep. Purif. Technol.* 294–300 (2015).
- [259] J. Ma, et al., Flow-electrode capacitive deionization (FCDI) scale-up using a membrane stack configuration, *Water Res.* 168 (2020) 115186.
- [260] K. Percin, O. Zoellner, D. Rall, M. Wessling, A tubular electrochemical reactor for slurry electrodes, *ChemElectroChem* 7 (2020).
- [261] G. Folaranmi, et al., Investigation of fine activated carbon as a viable flow electrode in capacitive deionization, *Desalination* 525 (2022) 115500.
- [262] L. Muttenthaler, B. Manhartgruber, Euler–Lagrange CFD simulation and experiments on accumulation and resuspension of particles in hydraulic reservoirs, *J. Braz. Soc. Mech. Sci. Eng.* 42 (2020) 209.
- [263] F. Wiesner, et al., Additive manufacturing of intertwined electrode pairs - guided mass transport with Gyroids, *Adv. Eng. Mater.* 25 (2023) 2200986.
- [264] Z. Lyu, J. Wang, Y. Chen, 4D printing: interdisciplinary integration of smart materials, structural design, and new functionality, *Int. J. Extrem. Manuf.* 5 (2023) 032011.
- [265] C. Zhang, et al., Integrated flow-electrode capacitive deionization and microfiltration system for continuous and energy-efficient brackish water desalination, *Environ. Sci. Technol.* 53 (2019) 13364–13373.
- [266] F. Liu, O. Coronell, D.F. Call, Effect of cross-chamber flow electrode recirculation on pH and faradaic reactions in capacitive deionization, *Desalination* 492 (2020) 114600.
- [267] X. Liu, S. Shanbhag, T.V. Bartholomew, J.F. Whitacre, M.S. Mauter, Cost comparison of capacitive deionization and reverse osmosis for brackish water desalination, *ACS EST Eng.* 1 (2021) 261–273.
- [268] S. Hand, J.S. Guest, R.D. Cusick, Technoeconomic analysis of brackish water capacitive deionization: navigating tradeoffs between performance, lifetime, and material costs, *Environ. Sci. Technol.* 53 (2019) 13353–13363.
- [269] F. Giacalone, et al., Application of reverse electro dialysis to site-specific types of saline solutions: a techno-economic assessment, *Energy* 181 (2019) 532–547.
- [270] W. He, et al., Field demonstration of a cost-optimized solar powered electro dialysis reversal desalination system in rural India, *Desalination* 476 (2020) 114217.
- [271] C. Huyskens, J. Helsen, W.J. Groot, A.B. de Haan, Cost evaluation of large-scale membrane capacitive deionization for biomass hydrolysate desalination, *Sep. Purif. Technol.* 146 (2015) 294–300.
- [272] J. Ma, et al., Flow-electrode capacitive deionization (FCDI) scale-up using a membrane stack configuration, *Water Res.* 168 (2020) 115186.
- [273] K. Percin, O. Zoellner, D. Rall, M. Wessling, A tubular electrochemical reactor for slurry electrodes, *ChemElectroChem* 7 (2020) 2665–2671.
- [274] Arenas, L. F., Walsh, F. C. & León, C. P. de. 3D-printing of redox flow batteries for energy storage: a rapid prototype laboratory cell. *ECS J. Solid State Sci. Technol.* 4, P3080 (2015).
- [275] F.C. Walsh, C. Ponce de León, Progress in electrochemical flow reactors for laboratory and pilot scale processing, *Electrochim. Acta* 280 (2018) 121–148.
- [276] H.J. Chung, J. Kim, D.I. Kim, G. Gwak, S. Hong, Feasibility study of reverse osmosis–flow capacitive deionization (RO-FCDI) for energy-efficient desalination using seawater as the flow-electrode aqueous electrolyte, *Desalination* 479 (2020) 114326.