

Review

Electrochemical Direct Lithium Extraction: A Review of Electrodialysis and Capacitive Deionization Technologies

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Abstract: The rapid expansion of lithium-ion battery (LIB) markets for electric vehicles and renewable energy storage has exponentially increased lithium demand, driving research into sustainable extraction methods. Traditional lithium recovery from brine using evaporation ponds is resource intensive, consuming vast amounts of water and causing severe environmental issues. In response, Direct Lithium Extraction (DLE) technologies have emerged as more efficient, eco-friendly alternatives. This review explores two promising electrochemical DLE methods: Electrodialysis (ED) and Capacitive Deionization (CDI). ED employs ion-exchange membranes (IEMs), such as cation exchange membranes, to selectively transport lithium ions from sources like brine and seawater and achieves high recovery rates. IEMs utilize chemical and structural properties to enhance the selectivity of Li⁺ over competing ions like Mg²⁺ and Na⁺. However, ED faces challenges such as high energy consumption, membrane fouling, and reduced efficiency in ion-rich solutions. CDI uses electrostatic forces to adsorb lithium ions onto electrodes, offering low energy consumption and adaptability to varying lithium concentrations. Advanced variants, such as Membrane Capacitive Deionization (MCDI) and Flow Capacitive Deionization (FCDI), enhance ion selectivity and enable continuous operation. MCDI incorporates IEMs to reduce co-ion interference effects, while FCDI utilizes liquid electrodes to enhance scalability and operational flexibility. Advancements in electrode materials remain crucial to enhance selectivity and efficiency. Validating these methods at the pilot scale is crucial for assessing performance, scalability, and economic feasibility under real-world conditions. Future research should focus on reducing operational costs, developing more durable and selective electrodes, and creating integrated systems to enhance overall efficiency. By addressing these challenges, DLE technologies can provide sustainable solutions for lithium resource management, minimize environmental impact, and support a low-carbon future.

Keywords: direct lithium extraction (DLE); electrodialysis (ED); capacitive deionization (CDI); electrochemical technology; sustainable resource management



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1. Introduction

Lithium (Li) is the lightest metallic and the least dense solid element found on Earth [1]. Lithium's small ionic radius (0.6 nm) gives it high electrochemical activity, along with a high

specific heat capacity and a low coefficient of thermal expansion [2–4]. These properties, combined with its high energy density and electrochemical potential (3.045 V), make lithium a crucial material for lithium-ion batteries (LIBs) [5,6]. Recent advancements in lithium-ion battery (LIB) production have significantly increased lithium demand, with 87% of the global end-use production being allocated to batteries [7,8]. LIBs are widely used in devices such as smartphones, laptops, and especially in electric vehicles (EVs) as well as larger-scale battery storage. By 2030, EV demand is expected to reach 142 million units, with lithium carbonate demand projected to rise to 3–4 million metric tons (Mt) [9,10]. Estimates suggest that demand could rise to 40 times by 2050 as a result of a move toward more environmentally friendly extraction methods [9,11].

The world’s major lithium resources, estimated to exceed 105 million tons as of 2024, are distributed across natural water resources (Salt Lake brines and geothermal brines) and solid-phase sources (hard rock and clay deposits) [8]. The greatest quantity of lithium resources is found in seawater, which contains over 230 billion tons of lithium, thousands of times more than the entire amount found in lithium ore and brine [12]. In addition, the method of recovering lithium from lithium ore is costly, challenging, and environmentally dangerous [13,14]. Thus, the lithium resources found in brine and seawater can effectively resolve this conflict, making the extraction of lithium from these sources extremely important.

World lithium reserves can be classified as illustrated in Figure 1. The chart illustrates the distribution of global lithium reserves by country. A significant portion of these reserves is concentrated in the “Lithium Triangle” of South America, comprising Chile, Argentina, and Bolivia, which collectively hold about 50% of the world’s reserves. Among them, Chile stands out with 34%, making it the leading country in lithium reserves and a key player in lithium production [7,8]. Lithium extraction from Salt Lake brines is cost effective and relatively simple to operate, compared to traditional ore extraction [15]. Notably, brines contain lithium at concentrations of up to 100–1000 ppm, making them highly attractive resources for lithium recovery [16]. However, conventional lithium extraction methods result in 85–95% water loss from brine, raising significant concerns about their impact on the water balance and biodiversity of Salt Lake ecosystems [17]. Consequently, there has been a growing interest in developing alternative technologies to replace conventional evaporation processes [11].

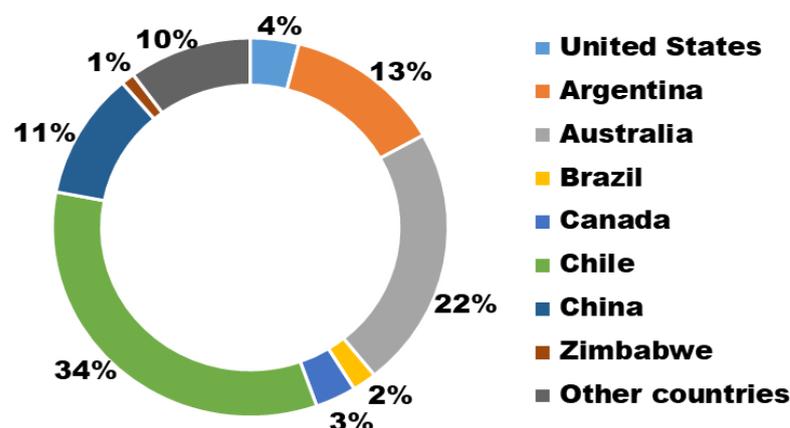


Figure 1. Graphical representation of world lithium reserves created by the authors based on data from [8].

Direct Lithium Extraction (DLE) has emerged as a promising and efficient technology for selectively extracting lithium from brine. DLE methods, serving as an alternative to the slow solar evaporation process, include ion exchange [18], adsorption [19,20], solvent

extraction [21], membrane separation [22], and electrochemical techniques [23,24]. Among these, electrochemical methods, primarily Electrodialysis (ED) and Capacitive Deionization (CDI), offer high lithium recovery efficiency, reduce water usage, and optimize energy consumption. Specifically, CDI achieves an energy consumption of less than 0.5–2.5 kWh/m³ when treating water with TDS concentrations below 2000 mg/L, making it highly efficient compared to traditional desalination methods such as reverse osmosis [23,25,26]. Table 1 provides a comparative analysis of various DLE methods, explaining the operational benefits and environmental considerations of each process.

Table 1. Comparison of different DLE methods.

Methods	Lithium Source	Advantage	Disadvantage	Ref.
Ion exchange	Seawater	High selectivity (>94.1% Li ⁺ extraction efficiency) Excellent recyclability (>90% capacity after 5 cycles) Lithium adsorption capacity (λ -MnO ₂ @IG 20.6 mg/g) Eco-friendly, high extraction efficiency (>90%)	Limited adsorption kinetics due to pore structure Potential Mn loss during operation	[18]
Adsorption	Brine	Low regeneration losses, long-term stability (>12 cycles) High lithium adsorption capacity (varies by adsorbent type) (Li _{1.6} Mn _{1.6} O ₄ 42.7 mg/g) High lithium recovery (99.8%)	Requires high temperature (>50 °C) Lower LiCl concentration than ion exchange Challenges in maintaining high Li ⁺ recovery efficiency due to Mg ²⁺ interference and washing losses	[19,20,27]
Solvent extraction	Brine	Good selectivity of Li over Mg (>9 cycles) Effective lithium stripping using water Reusable organic phase without regeneration Cost effective for high Mg/Li ratio brines	Limited commercial applications due to high organic phase consumption Mg impurities require additional steps	[21]
Membrane separation	Seawater, geothermal brine	Cost effective (USD 5–7/kg), moderate energy consumption (35–48 kWh/kg) (Nano Filtration) High selectivity for Li ⁺ recovery (>95%) Low energy consumption (supported liquid membrane) Energy efficient for low-salinity brines	Membrane fouling limits long-term use, limited separation selectivity for Mg ²⁺ /Li ⁺ (NF) High membrane and maintenance costs (SLM)	[22]
Electrochemical	Brine, seawater	(0.5–2.5 kWh/m ³) (Capacitive Ion Deionization) High energy efficiency (7–15 kWh/m ³), achieving high lithium chloride concentrations (23.15%) (Electrodialysis)	Limited performance for high Mg ²⁺ /Li ⁺ ratio, Co-ion expulsion reduces efficiency (CDI) Limited by fouling and concentration polarization (ED)	[23]

In recent years, research related to Li has accelerated, particularly in the area of recovering Li from brine sources [16]. As shown in Figure 2, interest in Li-related studies has increased significantly over the last 10 years, based on search results collected from the Google Scholar database. The search was conducted using keywords such as “lithium” and “direct lithium extraction” in the title, abstract, or keywords, covering the period from 2012 to 2023. The number of lithium-related publications increased by 128% from 2012 to 2020. However, since 2020, interest in DLE has grown, leading to a 163% increase in the number of publications from 2012 to 2023. These results emphasize the growing interest in the new DLE method, driven by the rising demand for sustainable and efficient lithium recovery.

Several recent review articles have comprehensively explored DLE technologies, including adsorption, solvent extraction, and membrane separation, highlighting their potential for sustainable lithium recovery [28–30]. However, these studies have primarily focused on general methodologies, lacking a detailed analysis of electrochemical methods, particularly Electrodialysis (ED) and Capacitive Deionization (CDI), and their potential in advancing DLE technologies.

This review explores the latest advancements and innovative approaches in electrochemical processes for DLE (Direct Lithium Extraction), providing an in-depth analysis of the fundamental mechanisms, practical applications, pilot-scale case studies, and future research directions of ED and CDI for sustainable lithium extraction. It also evaluates the feasibility of DLE as a large-scale industrial alternative to conventional lithium extraction methods, addressing key challenges such as scalability, energy consumption, and environmental impact with practical solutions. Furthermore, this review presents strategies for recovering lithium from desalination brines, transforming waste into valuable resources, and paving the way for sustainable resource management and innovative applications of DLE technologies.

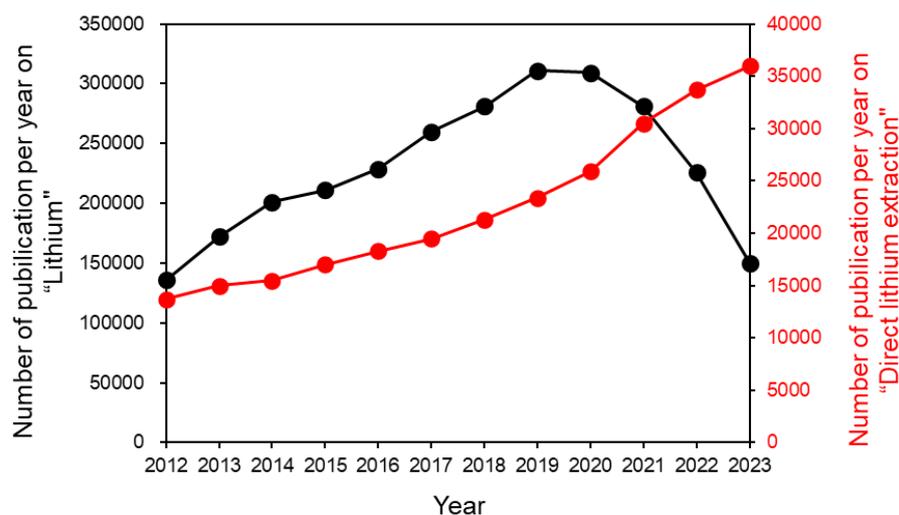


Figure 2. The number of publications related to Li based on Google Scholar searched in 2024 with the keywords 'Lithium' (black) and 'Direct lithium extraction' (red).

2. Conventional Lithium Extraction

Lithium is primarily found in brine resources, such as Salt Lakes, closed basins, and geothermal fluids [31], where it is dissolved in high-concentration brine solutions. Mostly, these brines are geologically located in Bolivia, Argentina, and Chile [8]. Conventional lithium extraction methods, including evaporation, purification, reactive crystallization, and precipitation, were initially developed and applied for industrial use.

As shown in Figure 3, conventional lithium brine processing increases lithium concentrations in solar evaporation ponds by removing water. In this extraction process, brine is transferred from Salt Lakes to large evaporation ponds, where the lithium concentration gradually rises through evaporation. Optimal conditions for this process include an arid climate, consistent wind patterns, limited brine infiltration to minimize resource loss, support effective evaporation, and minimal rainfall. The evaporation process involves circulating the brine through a series of ponds with progressively increasing salinity, a procedure that often takes several months [32]. When the lithium chloride (LiCl) concentration in the evaporation ponds reaches approximately 6000 ppm, the brine is transferred to recovery ponds [17]. At this stage, various salts precipitate as water is removed. Ion salts that do not precipitate spontaneously, such as boron, calcium, magnesium, and sulfates, must be removed through chemical treatment [32,33].

For example, Lime ($\text{Ca}(\text{OH})_2$) is added to the brine to remove magnesium ions (Mg^{2+}) as magnesium hydroxide ($\text{Mg}(\text{OH})_2$) and sulfate as calcium sulfate (CaSO_4). To eliminate any leftover Ca^{2+} as CaCO_3 through a single-replacement reaction, the remaining brine is treated with sodium carbonate (Na_2CO_3). Furthermore, the addition of Na_2CO_3 causes the brine to approach critical supersaturation, at which point Li_2CO_3 crystallization starts. As crystal ions disperse and accumulate on the surfaces of the Li_2CO_3 nuclei, the crystal nuclei start to form and enlarge. The Li_2CO_3 nuclei develop into precipitated particles [34], which then aggregate into larger particles [30]. The final Li_2CO_3 product is a chemically stable, odorless, white powder. Typically, the initial Li_2CO_3 product is dissolved and re-precipitated to achieve battery-grade purity (99.5 wt%) [17]. The final product is used as a precursor for lithium compounds utilized in the cathodes and electrodes of lithium-ion batteries [35,36].

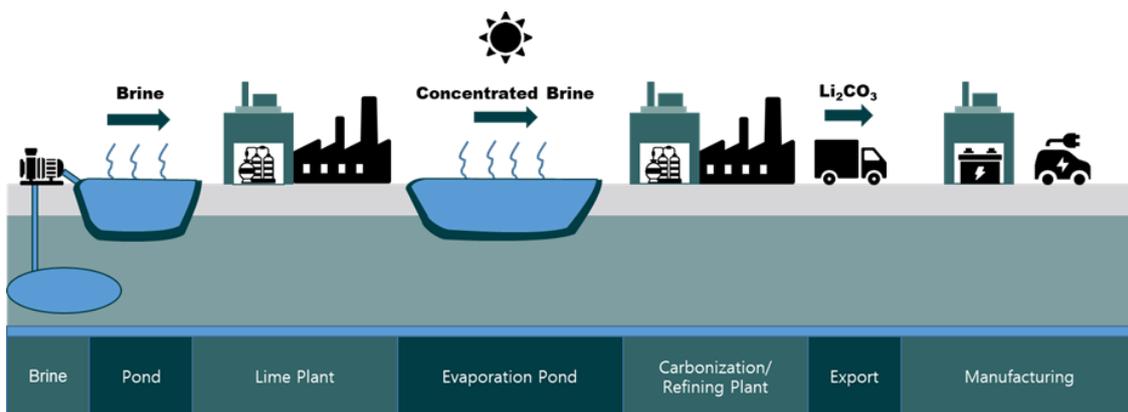


Figure 3. Conventional process to concentrate lithium brine and manufacturing lithium.

This lithium extraction method is considered the most traditional and economical approach due to its reliance on solar energy for lithium-ion concentration. However, the purity of lithium extracted from conventional brine methods is only around 50–80%, requiring further processing to achieve similar purity levels [37]. Lithium extraction via solar evaporation and chemical precipitation has drawbacks. The process is slow, energy intensive, and inefficient in water use, with a substantial portion of brine lost to the atmosphere. Additionally, it requires extensive land areas and is highly dependent on climatic conditions, leading to low predictability and sustainability [17]. Moreover, lithium extraction from brines poses higher environmental and occupational health risks compared to other lithium sources [38]. The use of large evaporation ponds in mining exposes lithium to elements like wind, raising contamination risks for nearby communities [39]. The process involves brine evaporation and mineral washing with sodium carbonate, which, if breached, could leak harmful chemicals into the environment and pollute water sources [40].

3. Direct Lithium Extraction (DLE)

DLE is a non-evaporative lithium extraction technology that eliminates the need for time- and space-consuming evaporation ponds. The DLE approach not only accelerates the extraction process but also reduces environmental impact, making lithium production more sustainable [36]. In DLE, lithium can be extracted from brine within hours or days through a single-stage chemical process. The lithium-depleted eluate is ideally reinjected, significantly reducing water consumption in the process. Additionally, DLE enables faster and more cost-effective lithium extraction not only from high-concentration brines found in Salt Lakes, salars, and geothermal resources but also from untapped resources such as oil and gas brines and groundwater brines [41]. These resources contain lithium concentrations ranging from tens to hundreds of ppm. To obtain high-purity lithium suitable for battery production, the brine must be concentrated to several thousand ppm while removing various impurities.

Therefore, traditional solar evaporation and precipitation methods have significant drawbacks, including high energy consumption, lengthy processing times, complex treatment stages, and potential secondary contamination, thereby driving interest toward more efficient extraction technologies like DLE [42]. Figure 4 illustrates various DLE technologies, including adsorption, ion exchange, solvent extraction, precipitation, and electrochemical methods. These technologies enable the selective recovery of lithium ions while offering fast processing, reduced water consumption, and improved environmental and economic efficiency.

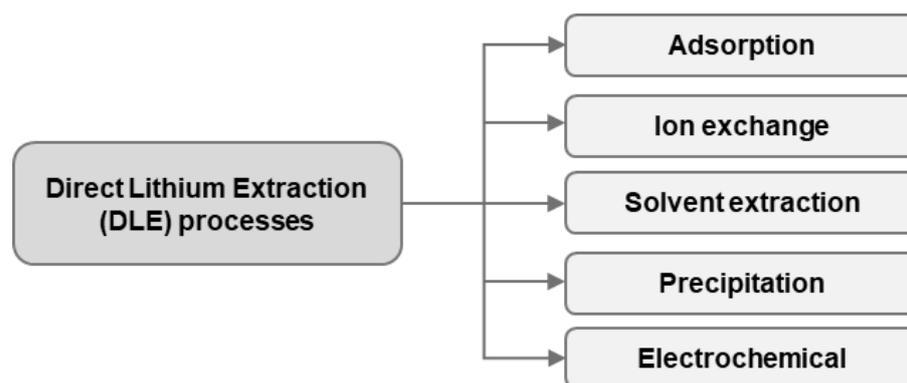


Figure 4. Classification of DLE process.

The selective extraction of lithium from solutions rich in lithium, such as brines or geothermal fluids, is made possible by adsorption-based Direct Lithium Extraction (DLE) techniques, which make use of specialized adsorbent materials with affinity or selectivity for lithium ions. These substances, which include polymers, clay minerals, and zeolites, have a low affinity for other ions in the solution but a considerable attraction to lithium ions [36]. However, the adsorption process has the drawback of performance degradation in ion-sieve materials over time, resulting in reduced efficiency and increased operational costs due to frequent replacements [35].

Ion exchange processes utilize specific resins or membranes designed to chemically exchange lithium ions with other ions present in the solution. These resins or membranes exhibit unique selectivity for lithium ions, enabling the efficient separation and recovery of lithium. Lithium ions can be selectively captured through these ion-exchange materials [43,44]. However, the ion exchange process requires energy-intensive regeneration using acidic solutions, which can lead to higher operating costs and potential environmental issues, including chemical waste and contamination risks [45]. Solvent extraction is a promising method for lithium recovery from brine due to its low cost and high product yield [16]. However, various organic extractants used in solvent extraction have the potential to cause environmental damage [46].

Additionally, membrane-based technology is an environmentally friendly method for lithium recovery, but it requires a relatively long operation time [22]. Therefore, there is a strong demand for a lithium recovery method that is more efficient, less time consuming, energy effective, and environmentally friendly. Numerous electrochemical techniques have recently been proposed and have garnered significant interest as an alternative solution for lithium recovery [42,47].

4. Electrochemical DLE

Electrochemical technologies play a crucial role in DLE processes, providing high efficiency and selectivity for concentrating and refining lithium ions from brine. These technologies selectively transport and concentrate lithium ions, creating a high-concentration lithium source for subsequent extraction stages. The electrochemical principles involve using electrical driving forces to move ions and employing selective membranes or electrodes to separate and concentrate lithium ions. For example, ED uses selective cation-exchange membranes to separate lithium ions, while CDI selectively adsorbs and desorbs lithium ions onto electrodes for concentration. The electrochemical adsorption of Li^+ is a promising technology for Li^+ separation from brine, providing high selectivity, high theoretical capacity, and low energy consumption [48]. Furthermore, adsorbents enhance lithium recovery efficiency by offering more active sites for ion adsorption, maximizing lithium extraction, and significantly improving overall recovery in electrochemical DLE processes.

Recent studies have shown that incorporating advanced adsorbents significantly enhances lithium recovery. For example, $H_{1.6}Mn_{1.6}O_4$ spherical adsorbents with internal hollow and porous surface structures achieved a lithium adsorption capacity of 47.54 mg/g in 360 min using the electrochemical adsorption method, highlighting the potential of electrochemical adsorption to improve lithium recovery [49]. Additionally, CeO_2 (CeLMO)-coated $LiMn_2O_4$ electrodes demonstrated a lithium adsorption capacity of $36.52 \text{ mg}\cdot\text{g}^{-1}$, achieved 96% pure Li^+ recovery, and maintained 60% capacity retention after 30 cycles at $50 \text{ mA}\cdot\text{g}^{-1}$ [48]. These developments underscore the importance of optimizing adsorbent materials and electrode configurations to improve lithium recovery efficiency, positioning electrochemical DLE processes as promising methods for large-scale lithium extraction.

Table 2 summarizes the advantages and disadvantages of electrochemical extraction methods, including ED and CDI, highlighting their applicability and limitations in various water treatment scenarios. Electrochemical methods offer not only high selectivity and environmental friendliness but also distinct advantages in ease of management, low waste production, and high lithium recovery [13,50,51].

Table 2. Summary of ED and CDI methods.

Methods	Classification	Advantage	Disadvantage	Ref.
ED	ED BMED SED ILM-ED	Suitable for aqueous solutions with high salinity High extraction efficiency Higher stability of the extraction process Mature technological development Well suited for large-scale industrial applications	Difficult to eliminate interference with monovalent ions High energy consumption Narrow selective range of electrode materials Produces concentrated brine, disposal challenges Higher operational cost due to periodic membrane replacement High operation voltage facilitates the side reaction like water splitting	[52,53]
	Combination of ED and other methods			
CDI	CDI MCDI FCDI HCDI	Simple and efficient extraction process Highly selective Li extraction Lower energy consumption for extraction More selective range of electrode materials Minimal chemical use Environmentally friendly	Potential Li loss during desorption Limited effectiveness for high salinity brine Electrode lifespan limits long-term operational efficiency Not yet mature for very large-scale desalination projects	[52,54]
	Derived from CDI			

The performance of the electrochemical lithium recovery process is assessed by key parameters such as the lithium selectivity coefficient, Li separation factor, Salt Adsorption Capacity, and specific energy consumption. These parameters are expressed by the mathematical model below, taking into account the concentration and mass ratio of coexisting ions in the feed solution [4,55,56].

The Li^+ selectivity of an electrode (α_M^{Li}) in the presence of other ions is determined by the ratio of the molar concentration of Li^+ in the recovery solution (C_{Li}^r) to the molar concentration of coexisting cations in the solution (C_M^s). Equation (1) shows this, where M represents any cation other than Li^+ (e.g., Na^+ , K^+ , Mg^{2+}) [4]:

$$\alpha_M^{Li} = \frac{C_{Li}^r}{C_M^s} \quad (1)$$

In Equation (2), the separation factor (SF) represents the higher Li concentration by the recovery process, as well as a comparison of Li purity in the recovery and in the source solution [49]. Although SF is highly dependent on the characteristics of the brine solution, it can determine the run time required to achieve the desired performance [57]:

$$SF = \left(\frac{C_{Li}}{C_M} \right)_r / \left(\frac{C_{Li}}{C_M} \right)_0 \quad (2)$$

In the CDI process, the total energy consumption (W_{total}) is calculated by integration of the current versus time plot. Thus, the total energy consumption can be given by

$$W_{total} = W_{ads} - W_{wash} + W_{des}$$

where W_{ads} is the energy consumption during electrosorption, W_{wash} is the energy recovered during cleaning, and W_{des} is the energy consumption during desorption. Equation (3) shows that the energy consumption (W) can be given by

$$W_{ads} = V \int_0^{t_{ads}} I dt, \quad W_{des} = V \int_0^{t_{des}} I dt \quad (3)$$

where t_{ads} and t_{des} represent the time of the applied potential in both the electrosorption and electrodesorption processes, respectively, and I represent the observed current [43].

In the ED process, specific energy consumption (E_{SEC}) was a parameter for economic evaluation. It was calculated using the following Equation (4) and can be described as electrical energy needed for extracting 1 mol Li^+ from the feed brine in the desalting compartment [58]:

$$E_{SEC} = \frac{U \int_0^t I(t) dt}{n_R} \quad (4)$$

where U is the applied voltage (V), I is the current (A), t is the operating time (h), and n_R is defined as the number of moles of Li^+ which migrated from the desalting compartment.

Salt Adsorption Capacity (SAC) is defined as the ion adsorption capacity, expressed in units of mg/g, calculated by dividing the total amount of ions removed by the mass of the electrode used in Equation (5) [59]:

$$\text{Salt Adsorption Capacity} \left(\frac{\text{mg}}{\text{g}} \right) = \frac{\int (C_{in} - C_{out}) \times \Phi \times dt}{M_{electrode}} \quad (5)$$

where C_{in} and C_{out} are the influent and effluent concentrations (mM), Φ is the flow rate (mL/min), and $M_{electrode}$ is the total weight of both electrodes (g).

4.1. ED

ED is an electrically driven separation process recognized as an emerging membrane separation technology for lithium extraction [60]. ED is classified into conventional ED, bipolar membrane (BMED), selective ED (SED), and ionic liquid membrane ED (ILM-ED). As shown in Figure 5, ED uses ion-exchange membranes to selectively transport ions under the influence of an electric field. Inside the ED stack, multiple ion exchange membranes (IEMs) are placed between anode and cathode electrodes. When an electric field is applied, cations migrate toward the CEM and anions move toward the AEM, enabling the separation and concentration of lithium ions. Spacer gaskets are used within the ED stack to separate the IEMs and create concentrate and dilute compartments. The electrolyte solution circulates through the electrode compartments, known as electrode rinse compartments [61]. Bajestani et al. studied lithium-selective cation exchange membranes (CEMs) that were developed by modifying spinel-type lithium-selective adsorbent particles. The optimized CEM achieved a molar selectivity of 32.2 for lithium over sodium, which is 62.3% higher than that of the conventional CEM [62].

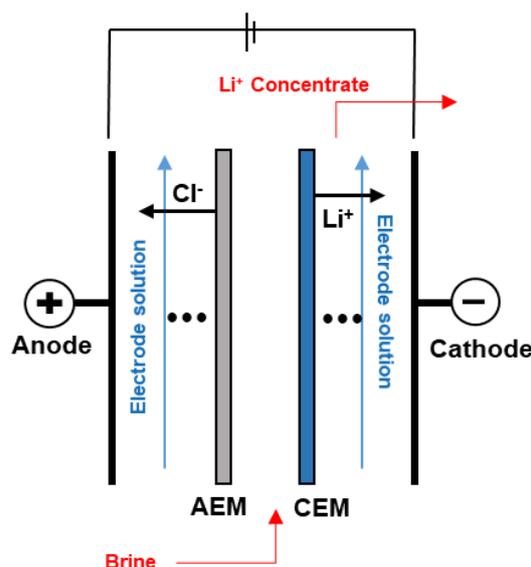


Figure 5. Scheme of ED system.

The bipolar membrane ED (BMED) process is a technology that combines traditional ED with bipolar membranes [63]. The BPM features a sandwich-like structure consisting of an AEM, a CEM, and an intermediate hydrophilic layer where water molecules dissociate into H^+ and OH^- [51]. When an electrical potential is applied, water splitting occurs at the boundary layer of the bipolar membrane, supplying H^+ and OH^- ions needed for acid and base generation and thereby enabling the simultaneous removal and recovery of ions from a salt solution [64]. Jarma et al. reported that ED stacks with different ion exchange and bipolar membranes achieved a lithium removal efficiency of 99.8% and a recovery rate of 86.4% at 20 V when 0.05 mol/L LiOH was used in the base chamber [64].

In selective dialysis (SED), monovalent-selective ion exchange membranes (IEMs) are used to replace standard IEMs in conventional ED systems. SED is a separation process based on ion-exchange membranes, offering high selectivity and making it essential for the separation of ionic species [65,66]. Using ion-selective exchange membranes, ED can effectively separate specific ions, such as monovalent and multivalent ions from a solution [67]. Li separation from multicomponent mixtures with a high Mg/Li ratio has been demonstrated to be technically and potentially economically feasible with the use of specific IEMs [51]. Guo et al. note that selective ED (SED) was used for the prefractionation of LiCl from Salt Lake brines with an optimal voltage of 10 V, which resulted in a higher Li recovery rate of 76.45% and an appropriate specific energy consumption (ESEC) of 0.66 kWh/mol Li [68].

Ionic liquid membrane ED (ILM-ED) is a system developed by integrating solvent extraction into the ED process. This process incorporates Li^+ selective organic liquid between two cation exchange membranes, forming a liquid film that exhibits high Li^+ selectivity and exceptional stability [69]. Liu et al.'s studies demonstrated an ILM-ED system where the Mg/Li decreases from 50:1 in the initial feeding brine to 0.5:1 in the receiving solution after ED. This ILM-ED system achieved higher current efficiency (65%) and lower specific energy consumption ($16 \text{ Wh}\cdot\text{g}^{-1} \text{ Li}$) compared to conventional ED methods [70]. Although ionic liquids have outstanding properties like thermal stability, high lithium selectivity, and low volatility, long-term operation may lead to solvent leakage and membrane swelling, which can reduce separation efficiency [22].

However, ED systems are known for their high initial installation costs and significant energy consumption when processing high-concentration brine. Additionally, membrane fouling and scaling can reduce efficiency, necessitating regular maintenance [71]. Comparative studies on water desalination using reverse osmosis (RO) and ED systems have

indicated that ED can concentrate influent water to higher salinity levels than reverse osmosis. However, ED also consumes more energy, requiring approximately 7–15 kWh/m³ of feed water to concentrate brine to high salinity levels [72]. A techno-economic assessment of lithium extraction using ED suggests that integrating renewable energy sources, such as solar power and wind, could reduce energy costs and improve the sustainability of the ED process [73]. Table 3 summarizes the previous ED technologies for lithium recovery along with their process parameters (the source of lithium, flow rate, and voltage), Li recovery ratio, Li separation factor, Li selectivity, and energy consumption.

Table 3. Summary of previous research on ED technology for Li recovery.

Methods	Membrane	Source of Li	Li ⁺ Concentration (M)	Flow Rate (LPM)	Recovery Ratio (%)	Separation Factor	Selectivity Factor	Applied Voltage(V)	Energy Consumption (Wh/mol)	Ref.
ED	CEM	Salt Lake brines	0.02	1.3	75.4	$F_{Mg/Li}$ from 60 to 7	N.A.	5	N.A.	[58]
	rGO-SDDS rGO CEM	Salt Lake brines	0.02	N.A.	N.A.	N.A.	$S_{Li/Na} = 1.19$, $S_{Li/Mg} = 5.27$, $S_{Li/Ca} = 4.72$	N.A.	N.A.	[74]
	Lithium separation membrane	Seawater	N.A.	N.A.	7.0	N.A.	N.A.	2	N.A.	[75]
	CEM	LiBr	2	N.A.	N.A.	N.A.	$S_{Li/Na} = 32.2$	5	N.A.	[62]
	CEM	LiCl + MgCl ₂	0.02	16.67	96.1	N.A.	$S_{Li/Mg} = 17.9$	6	5.4	[76]
BMED	AHA, BP-1E bipolar membrane	Li ₂ B ₄ O ₇ ·5H ₂ O	0.05	N.A.	97.8	N.A.	N.A.	30	N.A.	[77]
	Bipolar membrane	Li ₂ B ₄ O ₇ ·5H ₂ O	0.1	0.75–0.83	86.4	N.A.	N.A.	20	N.A.	[64]
	Bipolar membrane	Li ₂ B ₄ O ₇ ·5H ₂ O	0.04	N.A.	88.4	SLi = 99.6	N.A.	15	N.A.	[78]
	Bipolar membrane	Li ₂ B ₄ O ₇ ·5H ₂ O	0.05	N.A.	62.0	SLi = 94.7	N.A.	30	N.A.	[79]
	Bipolar membrane	Li ₂ B ₄ O ₇ ·5H ₂ O	0.04	0.83	73.0	SLi = 93	N.A.	25	N.A.	[80]
SED	Monovalent selective CEM	LiCl + MgCl ₂	0.02	10–20	95.3	$F_{Mg/Li}$ reduced by 21.8 times	$S_{Li/Mg} = 20.2–33.0$	N.A.	13.19	[67]
	Monovalent selective CEM	Synthetic brine	0.05	N.A.	77.5	N.A.	N.A.	5	32–850	[81]
	Monovalent selective CEM	Salt Lake brine	0.02	N.A.	76.5	$F_{Mg/Li}$ from 35.18 to 3.91	N.A.	10	660	[68]
	Monovalent selective IEM	Salt Lake brine	0.07	0.02	68.0	N.A.	N.A.	10	1770	[82]
	Monovalent selective IEM	LiCl+MgCl ₂ +NaCl	0.05	N.A.	71.9	$F_{Mg/Li}$ from 8.73 to 1.83	N.A.	7	270	[83]
	Monovalent selective CEM	Li ₂ SO ₄	0.6	0.42	75.8	N.A.	N.A.	6	N.A.	[84]
ILM-ED	PP13-TFSI	Seawater	2.45×10^{-5}	N.A.	22.2	$S_{Li} = 95$	N.A.	2	N.A.	[85]
	SELEMION CSO	Seawater	2.45×10^{-5}	N.A.	63.0	N.A.	N.A.	2–3	N.A.	[86]
	TBP+ 4mim TFSI liquid membrane	Simulated brine	0.1	N.A.	68.0	N.A.	N.A.	3	111.04	[70]
	Sandwiched TBP + ClO ₄ liquid membrane	Brine	0.1	N.A.	N.A.	N.A.	N.A.	3	130	[69]

4.2. CDI

CDI is an emerging electrochemical technology that uses charged electrodes to adsorb and remove ions from saline solutions. CDI operates with low energy consumption and can adapt to varying lithium concentrations in brine, making it particularly advantageous for lithium extraction [87]. This technology has attracted attention due to its low cost, low energy consumption, high efficiency, and no secondary chemical pollution [88]. CDI is applied in various fields, including water softening, desalination, water purification, and the recovery of high-value ions [89,90].

As an electrochemical water treatment method, CDI removes ions from saline water by applying an electric field across two porous carbon electrodes. As shown in Figure 6, when voltage is applied, cations are absorbed into the cathode and anions into the anode, effectively removing ions from the water. Reversing the voltage desorbs the ions back into the solution. By using specific electrode materials and operational conditions, CDI

can be effectively applied for the selective separation and extraction of target elements. The applied voltage in CDI is typically less than 2V, which is lower than that used in ED. Additionally, reverse voltage desorption can be used to regenerate saturated electrodes [91].

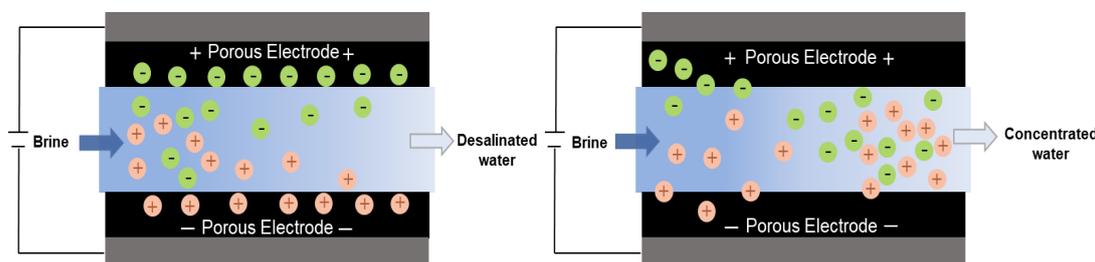


Figure 6. Schematic diagram of adsorption (left) and desorption (right) processes of CDI.

CDI technology includes several types, such as traditional CDI, membrane CDI (MCDI), flow CDI (FCDI), and hybrid CDI (HCDI). Among these, the primary component determining the performance of CDI systems is the electrode material, which offers greater accessibility. Commonly used materials include graphene, carbon nanoparticles, and lithium-ion sieves [52]. The electrochemical performance evaluation of these electrode materials is crucial for gauging their practical viability [92]. Yang et al. fabricated LMO/GO electrodes by integrating GO flakes with surface-grown LMOs. This study achieved a high lithium separation factor of 47.8, excellent stability with 80% capacity retention after 150 cycles, and a high Li^+ adsorption capacity of $720.2 \mu\text{mol g}^{-1}$, with negligible interference from other cations, demonstrating its potential for efficient Li^+ recovery [93]. These findings highlight the critical role that advanced electrode materials play in CDI-based lithium extraction. In particular, the ability to engineer electrode materials with tailored properties such as high specific surface area, selective ion affinity, and enhanced conductivity has been shown to directly impact the extraction performance and energy efficiency of the system. Moreover, the design and development of innovative CDI electrode materials enable the selective capture of specific ions, facilitating more efficient and precise lithium recovery from complex brine compositions [94].

MCDI is a variation of classical CDI that introduces ion-exchange membranes (IEMs) or ion-selective membranes (ISMs) between two opposing electrodes, preventing co-ion repulsion and reducing anode oxidation and energy consumption. Accordingly, it enhances the deionization efficiency and adsorption capacity, and it can operate effectively with low-concentration solutions [87,95]. MCDI significantly enhances the efficiency of CDI by improving operating conditions such as charge efficiency and ion adsorption [96]. Yu et al. explored MCDI with ZIF-8-PDA membranes, achieving a lithium selectivity of 1.50 for Li/Na solutions and 1.85 for Li/K solutions at 0.5 V in solutions containing Li and other ions (Na, K, Mg, and Ca) [95].

Flow CDI (FCDI) was developed to address the limitations of traditional CDI, such as the low electrode capacity and discontinuous operation. In FCDI, liquid carbon electrodes flow from each end, and ion exchange membranes are positioned to form ion removal channels. This design allows for the easy expansion of electrode capacity and enables the continuous removal of up to 95% of seawater ions without the need for an additional “ion release” stage [97]. FCDI mitigates the co-ion expulsion effect, enhancing charge and removal efficiency, while slurry electrodes allow for continuous ion adsorption and desorption, optimizing overall performance [98]. Saif et al. utilized an FCDI cell (Li-MFCDI) equipped with a ceramic lithium-selective membrane and achieved a lithium selectivity of 141 for Li^+/Na^+ and 46 for Li^+/K^+ . Additionally, the energy consumption of the Li-MFCDI process was 16.70 kWh/kg, demonstrating its efficiency and sustainability [99].

To further enhance the deionization capacity, various hybrid systems combining CDI with other deionization technologies have been developed. For instance, a nanofiltration NF-FCDI hybrid system consumes 16~20% less energy to produce potable water compared to RO systems [100]. Bae et al. developed an advanced bifunctional CDI-ELR system using LMO@ACC and Ag@ACC composite electrodes with a four-step constant voltage process (forward/zero/reverse/zero). The system achieved selective Li⁺ enrichment and deionized water production within a single cycle, with simultaneous competing ion adsorption and Li⁺ liberation [101]. Siekierka studied a hybrid CDI (HCDI) system with lithium–manganese–titanium oxides (LMTOs) for lithium recovery from brines. The system achieved a lithium separation factor ($\beta_{\text{Li/Mg}}$) of 2.14 and released lithium ions with over 70% efficiency. The separation process was influenced by the activity coefficient of the initial feed and the applied voltage, with lower lithium-ion concentrations enhancing separation efficiency [102]. Table 4 summarizes the previous research on CDI technologies for lithium recovery, along with their process parameters (the source of lithium, flow rate, and adsorption capacity), Li separation factor, Li selectivity, and energy consumption.

Table 4. Summary of previous research on CDI technology for Li recovery.

Methods	Cathode	Anode	Source of Li	Concentration (ppm)	Flow Rate (mL/min)	Adsorption Capacity (mg/g)	Separation Factor	Selectivity Factor	Energy Consumption (Wh/mol)	Ref.
CDI	GA/CoP/Co ₃ O ₄	C/CoP/Co ₃ O ₄	Salt Lake brine	50	N.A.	37	N.A.	N.A.	N.A.	[103]
	LVO-rGO	AC	LiCl	610	40	39.53	N.A.	N.A.	N.A.	[104]
	LMO-GO	AC	LiCl	69	16	5	$\alpha_{\text{Mg}}^{\text{Li}} = 47.8$	N.A.	N.A.	[93]
	λ -MnO ₂ /rGO	AC	LiCl	69	10	4.17	$\alpha_{\text{Na}}^{\text{Li}} = 38$, $\alpha_{\text{K}}^{\text{Li}} = 57$, $\alpha_{\text{Mg}}^{\text{Li}} = 41$, $\alpha_{\text{Ca}}^{\text{Li}} = 8$	N.A.	N.A.	[105]
MCDI	AC (monovalent selective CEM)	AC (AEM)	LiCl+MgCl ₂	40	30	N.A.	N.A.	$S_{\text{Li/Mg}} = 2.95$	1.8	[87]
	HMO	AC (AEM)	LiOH+LiCl	694	20	2.43	N.A.	N.A.	161.4	[43]
	LMO, LiMn ₂ O ₄	AC (AEM)	LiOH	50	20	1.36	N.A.	N.A.	N.A.	[106]
	AC/ZIF-8-PDA	AC (AEM)	LiCl	69	20	N.A.	N.A.	$\rho_{\text{Li/Na}} = 0.95$, $\rho_{\text{Li/K}} = 0.97$, $\rho_{\text{Li/Mg}} = 0.28$, $\rho_{\text{Li/Ca}} = 0.41$	60.4–145	[95]
FCDI	Nanoporous AC (CEM)	Nanoporous AC (AEM)	LiCl	1–100	3–9	N.A.	N.A.	N.A.	N.A.	[107]
	ZIF-8/CNT	AC (AEM)	LiCl	58	5	N.A.	N.A.	$S_{\text{Li/Na}} = 6.3$, $S_{\text{Li/Ni}} = 6.8$, $S_{\text{Li/Mg}} = 7.2$	N.A.	[108]
	AC	AC (AEM)	LiCl	16	10	N.A.	$\alpha_{\text{Na}}^{\text{Li}} = 141$, $\alpha_{\text{K}}^{\text{Li}} = 46$, $\alpha_{\text{Mg}}^{\text{Li}} = 3$	N.A.	115.9	[99]
HCDI	LMO-ACC	Ag-ACC	LiCl+NaCl	69	1	N.A.	N.A.	N.A.	N.A.	[101]
	LMTO	AC (AEM)	Geothermal brine	13	100	28.6	$\alpha_{\text{Mg}}^{\text{Li}} = 2.14$	N.A.	477–3704	[102]
	LMTO	AC (AEM)	Geothermal brine	16	67	N.A.	N.A.	N.A.	N.A.	[109]
	LMTO/graphite	AC	LiCl	70	67	36.5	N.A.	N.A.	N.A.	[110]
	P500-LiMn ₃ Ti _{0.15}	AC (AEM)	Geothermal water	16	100	34	N.A.	$S_{\text{Li/Mg}} > 3$	N.A.	[111]
	LiO-FeO-Mn ₂ O ₃	AC (AEM)	LiCl	26	67	32	N.A.	N.A.	N.A.	[112]
	LMTO	AC (AEM)	LiCl	139	67	N.A.	N.A.	N.A.	N.A.	[113]
	LNMO	AC (AEM)	LiCl	70	10	1.8	$\alpha_{\text{Ca}}^{\text{Li}} = 334$, $\alpha_{\text{Mg}}^{\text{Li}} = 167$, $\alpha_{\text{K}}^{\text{Li}} = 47$, $\alpha_{\text{Na}}^{\text{Li}} = 11$	N.A.	4.1	[114]

5. Pilot Scale of Lithium Recovery

ED and CDI are promising technologies for desalination and ion separation, but scaling them to pilot-scale applications presents several challenges. Both systems face issues such as membrane fouling caused by organic matter, microorganisms, and inorganic pre-

precipitates, as well as scaling resulting from calcium and magnesium salts, which necessitate frequent cleaning and maintenance [61]. Additionally, their energy efficiency decreases with increasing salinity; CDI, in particular, requires significant energy for electrode regeneration, making it less competitive than reverse osmosis in high-salinity contexts [115]. Furthermore, the cost and durability of ion-exchange membranes and high-surface-area electrodes remain critical barriers [116]. Addressing these challenges through optimized designs, advanced materials, and effective fouling prevention strategies is essential to enable the pilot scale for industrial applications of these technologies.

Recent studies on ED and CDI for lithium recovery have yielded promising results at the pilot scale. Melnikov et al. demonstrated a pilot-scale ED system for lithium hydroxide production from lithium chloride solutions containing organic solvents. The system achieved an average flux of $5.73 \text{ mol} \cdot \text{m}^{-2} \cdot \text{h}^{-1}$ and a specific energy consumption of 0.15 kWh/mol. It processed solutions with organic solvent contents ranging from 1.8% to 59%, and the final lithium hydroxide product was free of ionic impurities, highlighting the feasibility of using ED for lithium recovery in complex chemical environments [117]. Joo et al. developed a pilot-scale ED system utilizing $\lambda\text{-MnO}_2$ and Ag electrodes for lithium recovery from desalination concentrate. The system achieved lithium-ion enrichment from 0.035 mM in the feed to 62 mM in the final product, with purity increasing from 0.0048% to 88% and an enrichment factor of 1800. Operating at a rate of $0.25 \text{ m}^3/\text{h}$, it demonstrated selective lithium-ion recovery compared to competing ions such as Na^+ , Mg^{2+} , and Ca^{2+} [118]. Together, these studies underscore the potential of ED technology for efficient and selective lithium recovery across diverse chemical matrices and operational conditions.

In parallel, the pilot-scale CDI system has also shown promise in lithium recovery through a two-step desalination process using six CDI cells. This system reduced feed water salinity from 1 g/L to 0.5 g/L and produced approximately 200 L/h of demineralized water. Optimized operational voltages between 0.85 and 0.9 V facilitated energy recovery of approximately 30%, underscoring its energy efficiency [119]. Yoon et al. further emphasized the system's preferential removal of lithium ions over sodium ions, with ion selectivity enhanced under conditions such as low feed concentrations, moderate flow rates, and extended adsorption/desorption times. These findings demonstrate the feasibility of CDI technology for lithium recovery when operational parameters are optimized [120]. Table 5 summarizes the pilot-scale experimental results for ED and CDI in lithium recovery.

Table 5. Summary of pilot-scale experimental results for ED and CDI in lithium recovery.

Technology	Feed Water	Flow Rate	Energy Consumption	Lithium Recovery Rate	Purity of Recovered Product	Operating Voltage	Specific Advantages/Challenges	Ref.
ED	Lithium chloride with 1.8–59% organics	$0.25 \text{ m}^3/\text{h}$	0.15 kWh/mol	N.A.	100%	N.A.	Handles organic solvents; moderate energy consumption	[117]
	Brine concentrate, Li^+ : 0.035 mM	$0.25 \text{ m}^3/\text{h}$	N.A.	88%	88%	N.A.	High selectivity over Na^+ , Mg^{2+} , Ca^{2+}	[118]
CDI	Brackish water, Li^+ : 1 g/L	200 L/h	N.A.	N.A.	-	0.85–0.9 V	High water recovery and energy efficiency	[119]
	Brackish water, low salinity	N.A.	N.A.	N.A.	Enhanced Li^+ selectivity	N.A.	Effective at low concentrations, moderate scalability	[120]

6. Conclusions

This review emphasizes the critical role of electrochemical technologies, particularly ED and CDI, in addressing the growing demand for sustainable lithium extraction. As global lithium consumption continues to rise due to the expansion of electric vehicles and renewable energy storage systems, traditional evaporation-based recovery methods are increasingly showing their limitations. Electrochemical approaches present a promising alternative, offering advantages such as high lithium selectivity, reduced water consumption, and compatibility with renewable energy sources.

ED has demonstrated a strong performance in selectively separating lithium ions from brines, while CDI provides flexibility to adapt to varying lithium concentrations with low energy consumption. Pilot-scale studies have validated the efficiency and selectivity of both technologies, highlighting their potential for commercial applications in lithium enrichment and ion separation. However, these technologies face significant challenges, including energy-intensive operations, limited lithium selectivity at low concentrations, and the degradation of electrodes and membranes. Addressing these challenges will require advancements in electrode and membrane materials, as well as the optimization of operational parameters to enhance both efficiency and cost effectiveness.

Future research should prioritize the development of lithium-selective materials with enhanced stability and performance under real-world conditions. Additionally, hybrid systems that integrate electrochemical methods with complementary technologies should be explored to further improve efficiency and expand their applications. For instance, combining ED with adsorption techniques or integrating CDI with membrane filtration systems could optimize performance and reduce operational costs. Successful demonstrations at the pilot scale indicate the feasibility of transitioning these technologies from laboratory-scale studies to industrial-scale applications.

Such advancements will enable electrochemical lithium extraction technologies to meet the growing demand for lithium while minimizing environmental impacts. These technologies have the potential to become key solutions for sustainable resource management in the evolving global energy landscape.

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