Insights into adsorbent materials for lithium extraction by capacitive deionization: reconceptualizing the role of materials informatics

Journal:	*Journal of Materials Chemistry A*
Manuscript ID	TA-PER-11-2023-007069.R1
Article Type:	Perspective
Date Submitted by the Author:	20-Feb-2024
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Insights into adsorbent materials for lithium extraction by capacitive deionization: reconceptualizing the role of materials informatics

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Abstract

The exponential expansion of the lithium battery market has led to challenges in acquiring lithium resources. Extracting lithium from marine resources has been explored as a potential solution to address the shortage of lithium supply. However, extracting lithium ions from seawater presents significant challenges due to its low concentration and complex ion environment. Capacitive deionization (CDI) technology has demonstrated a remarkable ability to selectively separate metal ions from seawater, in which electrode materials play a crucial role. Traditional methods of discovering electrode materials have relied on the trial-and-error principle, and the experimental process of material design and synthesis requires substantial manpower and material resources. The emergence of materials informatics has revolutionized the design and development of electrode materials, substantially reducing the application cost of CDI. Combining our own research experience, we prospected the application prospects of theoretical calculations in CDI lithium extraction and introduced the development progress of electrode materials that are not limited to CDI lithium extraction in recent years. The key points of using density functional theory (DFT), molecular dynamics (MD), and finite element simulation (FES) to study the mechanism of lithium adsorption, using high-throughput computing and machine learning (ML) to screen new electrode materials are emphasized. The research emphasizes the synergistic utilization of DFT, MD, ML, and FES methodologies to provide instructive insights, aimed at advancing the design of effective CDI electrode materials tailored for lithium extraction from seawater.

1. Introduction

The lithium element plays a pivotal role in the storage of electrical energy, making it an indispensable component in the realm of energy resources. Lithium minerals, including spodumene, lepidolite, petalite, zinnwaldite, and others, are predominantly distributed across global locations such as Australia, the United States, Zimbabwe, Canada, Mexico, and China, exhibiting wide and uneven distribution characteristics. Common extraction methods from these ores involve acid leaching and roasting, offering advantages such as a straightforward operational process, high recovery rates, elevated product purity, and minimal environmental impact.[1, 2] However, the burgeoning electric vehicles and electronic intelligent equipment have led to a surge in demand for lithium. Based on a cautious estimation of future lithium consumption, focusing exclusively on the proliferation of electric vehicles, it is anticipated that terrestrial lithium reserves will be exhausted by 2080.[3] Extracting lithium resources from salt lake brines is an effective measure to alleviate the lithium resource crisis. Lithium in the salt lake brines typically exists in the forms of LiCl and Li_2CO_3.[4] Extraction and recovery of lithium from salt lake brines prove to be less costly and lower energy consumption compared to hard rocks.[5] However, challenges arise due to the presence of competing elements, particularly alkali earth metals or alkali metals with higher proportions, such as sodium, potassium, and magnesium. These elements complicate the extraction and separation of lithium from salt lake brines.[6] In this context, capacitive deionization (CDI) technology, working on principles of electric double layers or faradic reactions, emerges as a strategically valuable approach for extracting lithium ions.[7-11] By elucidating the adsorption mechanisms and significantly improving the efficiency of lithium extraction from salt lake brines, the insights gained from the comprehensive study of CDI technology can be extended to seawater scenarios, thereby contributing valuable experiences and inspiration to the field.

Extracting lithium directly from seawater appears to be more favorable primarily due to its vast availability, which amounts to approximately 230 billion tons, a stark contrast to the limited 62 million tons available on land.[3] The main obstacles to extracting lithium from seawater stem from the extremely low lithium concentration (average 0.17 ppm) and the presence of interfering ions during the extraction process.[12] Concentrated brines from seawater desalination are typically returned to the sea, where the concentrations of Na^+, Mg^{2+}, K^+, and Ca^{2+} in seawater greatly exceed that of Li^+, and these ions exhibit comparable hydration radius and chemical properties with Li^+, intensifying the complexity of lithium extraction from seawater.[12] The intricate conditions inherent to lithium extraction from seawater render conventional technologies, such as solar evaporation precipitation and solvent extraction, impractical due to their exorbitant operational expenses and protracted cycling times.[13, 14]

Relatively, the electrochemical methods enable precise control over the insertion and ejection of ions by modulating chemical potential, thereby offering the advantages of enhanced efficiency and exceptional selectivity.[15] Specially, CDI acted as a novel electrochemical method for extracting lithium from seawater. CDI is preferentially characterized by its ability to achieve selective ionic separation through ionic mobility, hydration, valence, ionic species solvation, and redox activity. Moreover, driven by an electric field, specific adsorbents have demonstrated a robust capacity for metal ions adsorption and offer a straightforward extraction process.[16] CDI process can be analogized to the charging and discharging process of a battery/supercapacitor, where a certain voltage (typically <1.23 V to prevent hydrolysis reactions) is applied between the opposite electrodes. During charging process, lithium ions in the solution are adsorbed onto the porous electrode surface. Upon reaching the saturation point of the adsorption capacity, the adsorbed ions are released either by short-circuiting or applying a reverse voltage, thereby facilitating the desorption process.[17]

The lithium extraction efficiency from seawater by CDI mainly relies on the lithium ion selectivity of electrode materials. It is crucial to delve into the adsorption process and adsorption mechanisms of these materials under the combined influence of electric and oceanic fields.[18] Material informatics is expected to be a promising methodology for promoting CDI technology, enabling the exploration of lithium extraction materials and providing valuable references. Computational materials design discovers new materials through structure-property models and uses computer-aided design and experimental verification, becoming a new method for material discovery and optimization. In this text, a systematic understanding of CDI and materials informatics was presented, where CDI acted as an emerging technology for lithium extraction from seawater, and materials informatics aids in interpreting the adsorption mechanisms and exploring high-selectivity lithium extraction materials. Through the synergistic utilization of powerful CDI technology in experimental aspects and materials informatics in theoretical investigations, the realm of lithium extraction from seawater can be significantly expanded and refined.

2. Fundamental knowledge of CDI

CDI technology has shown high efficiency in various fields including seawater desalination,[19, 20] water purification,[21] water disinfection,[22] metal recovery,[23] and pollutant removal.[24] Innovative CDI configurations, such as membrane CDI (MCDI), flow-electrode CDI (FCDI), and hybrid CDI (HCDI) have been applied in lithium extraction (Figure 1). These systems incorporate ion exchange membranes (IEMs) to separate the electrodes from target solution, including anion exchange membrane (AEM) and cation exchange membrane (CEM), thereby controlling the efficiency of ion-desorption and ion-recovery processes. Notably, the IEMs applied on electrode surfaces exhibit selective permeability, enhancing target ion separation from specific solutions and providing prospects for selective ion recovery.[19]

In the context of MCDI, IEMs have been attached on the electrode surface to effectively separate anions and cations. This strategic implementation enables the selective capture of specific ions during adsorption process, preventing co-adsorption and allowing only cations or anions to migrate onto oppositely charged electrodes.[25] Additionally, FCDI has been optimized by incorporating a bipolar plate, known as the flowing electrode, on both sides of the electrode surface.[26] This modification greatly enhances the efficiency of seawater treatment, realizing long-term adsorption and eliminating the necessity for cleaning steps. A comprehensive analysis of existing literature on FCDI reveals various factors affecting its performance. These factors extend critical aspects beyond the parameters of feed salinity and removal efficiency, such as flow-electrodes configurations, device design, and operation modes. Furthermore, HCDI stands as a novel CDI configuration composing a faradaic electrode, IEM, and carbon electrode,[27] where anions are retained within electric double layer formed on the surface of the carbon electrode, while cations are removed from the solution by faradaic electrode, thus improving the efficiency of selective ion removal.

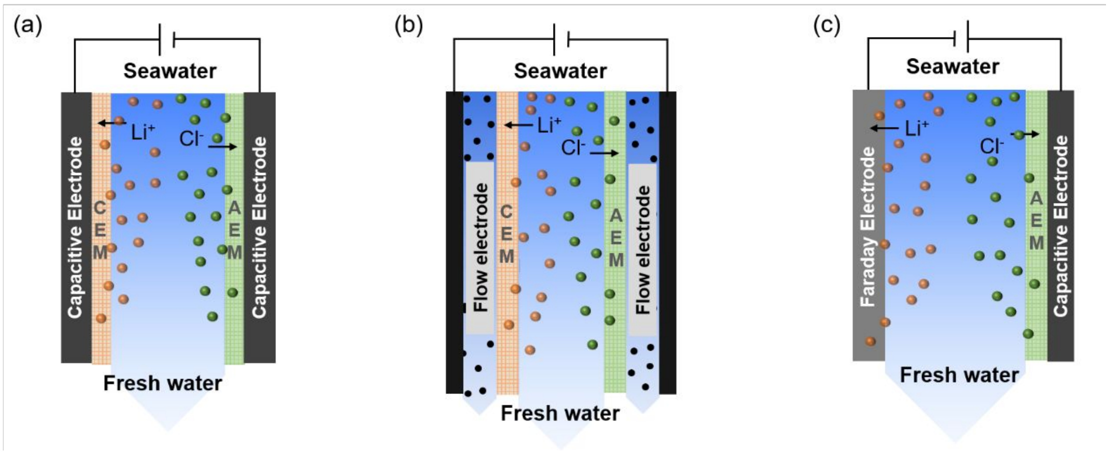

Figure 1. Schematic diagram of Li^+ extraction from seawater using (a) MCDI, (b) FCDI, and (c) HCDI.

The future trajectory of CDI is poised towards refining CDI system designs and developing electrosorption materials. The selective separation of ions is intricately affected by factors such as ionic size, geometric structure, valence state, and electrostatic interactions.[18, 23, 28] Previous studies have demonstrated that the hydration radius and valence state of ions play a pivotal role in their sequential removal through CDI processes.[18] Specifically, ions processing larger charges exhibit a higher likelihood of adsorption, with selectivity primarily contingent upon the hydration radius for ions with identical charges.[28] Notably, compared with multivalent ions with a larger hydration radius, monovalent ions characterized by a smaller hydration radius are preferentially removed.[29] In parallel with considerations of ionic parameters, pore structure, surface groups, and charged groups within micropores of electrode materials significantly impact the selective adsorption of ions. The understanding and manipulating of these intricate factors are crucial for advancing our comprehension of CDI mechanisms, enabling the development of enhanced CDI systems with enhanced selectivity and efficiency.

3. Adsorbent materials

The optimal electrosorption materials employed in CDI electrodes must adhere to specific criteria: (i) a substantial specific surface area with an ample number of ion adsorption sites; (ii) high conductivity and ionic mobility; (iii) elevated hydrophilicity to ensure complete utilization of hole structure; and (iv) robust chemical stability to ensure the operational stability of CDI process.[30, 31] Electrode materials with features such as ion intercalation/deintercalation within internal crystal sites or atomic planes have been systematically studied to enhance overall performance. These materials shown in Figure 2 include lithium ion sieves (LISs),[32] crown ethers (CEs),[33] two-dimensional (2D) materials,[34] metal-organic frameworks (MOFs),[35] and covalent-organic frameworks (COFs),[36] which have garnered substantial attention. Their unique properties enable higher ion storage capacity, elevated energy density, and effective extraction even at low lithium concentrations. In the realm of material design, simulating the ion adsorption properties of these novel materials under complex conditions domains proves invaluable. Such simulations accelerate the iterative design and development of electrode materials suitable for CDI, improve the capabilities of the technology, and pave the way for innovative applications in different fields. In this section, an exhaustive analysis of electrode materials within the realm of lithium extraction via CDI was conducted, with a focused summary outlining the prospective materials earmarked for future applications in lithium extraction processes.

3.1 Carbon-based materials

Carbon-based materials have traditionally been integral to CDI, encompassing diverse iterations such as carbon nanofibers (CNFs), carbon aerogels (CAs), carbon nanotubes (CNTs), activated carbons (ACs), graphene, etc. However, carbon-based electrode materials are susceptible to electrode corrosion mainly due to anode oxidization, consequently limiting the cycling stability of CDI systems. In response, Cheng et al. conducted a comprehensive summary of recent advances in carbon modification methods such as element doping, metal oxide doping, and polymer formation, and put forward some prospects.[37] Generally, ACs are the most widely adopted electrode material in CDI due to its heightened adsorption capacity and costeffectiveness.[38] Nonetheless, its diminished conductivity and elevated electrical transfer resistance limit its further application in CDI. Addressing these challenges requires the exploration of novel carbon materials. In this context, the focus of researchers is increasingly on finding carbon electrode materials with specific pore structures and exploring chemical modification methods to introduce hydrophilic functional groups, essential for achieving selective adsorption or removal of specific ions. These strategies hold promise for significantly augmenting the electrosorption capacity of carbon-based materials within the CDI framework.

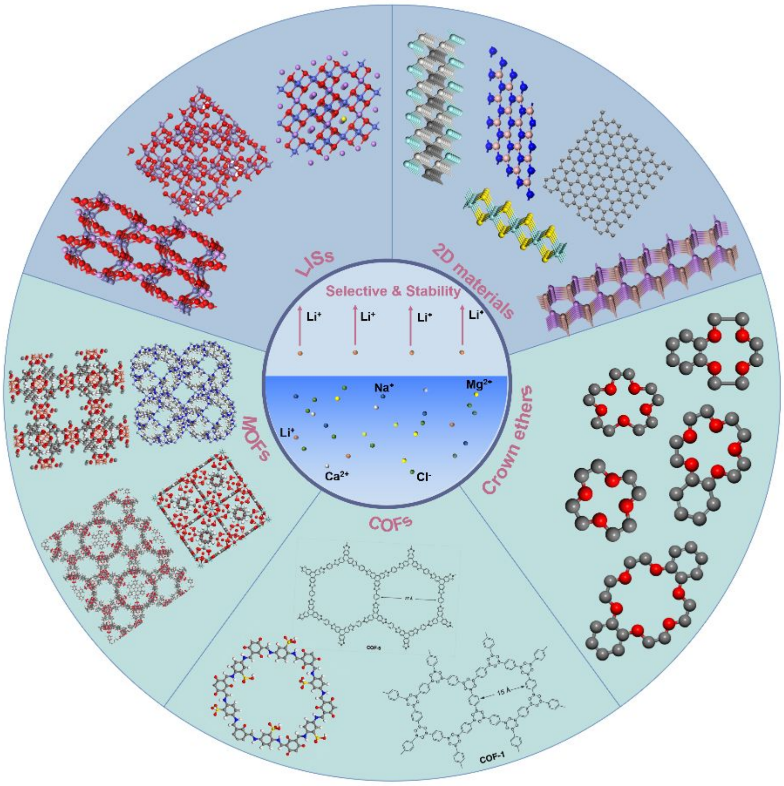

Figure 2. Schematic diagram of typical adsorbent materials for lithium extraction.

3.2 LISs

LISs fundamentally comprise lithium ion adsorbed oxides, embodying the remarkable capacity to selectively recognize and screen lithium ions within the mixed saline solution. This attribute endows LISs with notable advantages, including superior extraction selectivity for Li^+, high adsorption efficiency, and environmental benignity.[39] Within diverse ion solutions, LISs exhibit high selectivity for Li^+, rendering them the most extensively researched and proficient inorganic lithium-ion adsorbents to date. Consequently, they find widespread application in lithium extraction processes.[40] Lithium titanium oxide,[41] a typical LIS variant, stands out due to its non-toxicity to aquatic environments and ease of removal from aqueous solutions. The aftermentioned attributes bestow the process of lithium extraction with operational simplicity, maturity, and robust reliability.[42]

To further improve the selectivity, tailored modifications can be employed by considering structural/compositional parameters such as specific surface area, morphology, surface wettability, and functional groups. Various modification methods have been explored to improve the conductivity and stability of materials, including the incorporation of coating materials. Additionally, strategies have also been developed to mitigate the manganese dissolution loss in lithium manganese oxide, encompassing optimizations in the crystal structure, surface modification, and doping with anions, cations, or transition metals,[43, 44] all of which contribute significantly to refining the efficacy and applicability of lithium extraction methodologies.[45, 46]

3.3 CEs

As a size-based ion-selective material, CEs possessing cavities of comparable sizes to lithium ions are selected for their distinct properties.[47] Utilizing electrochemical methods, these CEs effectively extract and store lithium ions. Typical examples of selective CE include 15-crown-5,[48] 9-crown-3,[49] 12-crown-4,[50] benzo-15-crown-5,[51] etc. The critical determinant of ionic selectivity lies in the relationship between the size of the CE cavity and ionic radius.[49] Proper modification strategies, including structural adjustment and morphological improvement,[51] have been instrumental in optimizing the characterization and adsorption capacity of specific CEs while ensuring their sustained selectivity for lithium ions.[52]

Specifically, structural adjustment requires techniques including element doping, functional group diversification, vacancy construction, and others.[53] Concurrently, morphology improvements have been achieved through the design of macrocyclic structures, nanoparticles, 2D layered structures, and three-dimensional polymeric structures. This strategic amalgamation has not only enhanced the selectivity and stability of the resulting adsorbents but has also broadened the horizons of ion extraction methodologies.[54, 55]

3.4 2D materials

2D materials, renowned for their exceptional mechanical strength, stand out as prime candidates for constructing ultrathin membranes with minimal transport resistance and maximal permeance. They serve as ideal building blocks, promoting the development of efficient electrode materials. The geometric size, surface charge, and chemical properties of 2D materials all have significant effects on the selective separation and capture of lithium ions.[56] On one hand, ions need to overcome energy barriers when passing through the binding sites within nanochannel. The active sites can affect the transport of ions through intermolecular forces, chemical bonds, and static electricity, leading to the different ion migration rates involved in the nanochannel.[57] On the other hand, 2D materials are frequently employed in electrochemical intercalation techniques, wherein an electric field applied on the electrodes facilitates the charge/discharge process, and the adjustable transformable distance enables the efficient lithium storage.[58] A key challenge in CDI research is the precise design of 2D materials to restrain lamellar accumulation and ensure sufficient active surface. For example, the interaction effects between layered graphene oxide and ions significantly influence the removal, including coordination, chelation, hydrogen bonding, electrostatic interaction, hydrophobic effect, π-π interaction, acid-base interaction, etc.[59] Similar to graphene-based materials, the size, density, and chemical functionalization of pores in 2D transition metal carbides, nitrides, and carbonitrides (MXenes) also play a pivotal role in membrane performance. Furthermore, manipulating point defects, doping, and adjusting boundaries can improve the adsorption behavior and specific capacity. The long-term stability, deposition, surface roughness, and stacked structure of electrodes profoundly influence their performance.[60] Notably, heterostructures combining MXenes with other 2D nanomaterials exhibit improved selectivity, representing a promising avenue for further advancements in lithium extraction by CDI.

3.5 MOFs

In recent years, MOFs and their derivatives[61] have gained prominence as electrode materials in CDI and other separation technology. These MOFs demonstrate remarkable efficacy in the separation of Mg^{2+}/Li^{+} in aqueous solutions. However, the application of a single MOF-derived carbon structure is not sufficient to form a continuous pore architecture suitable for CDI, primarily due to the large internal pore between MOF-based particles.[62] In addition, most MOFs are unstable in water and prone to aggregation during preparation process, which limit their further use.

Recently, some researchers have explored techniques such as synthesizing MOFs on substrates[63, 64] and changing morphology to create core-shell[65] and yolk-shelll[66] derivatives. Others have combined MOFs with polymers to improve the adsorption performance, such as polyvinyl chloride (PVC), polystyrene sulfonic acid, sulfonated tetrafluoroethylene-based fluoropolymer-copolymer (Nafion), polyphenylsulfone with pyridine moiety (PPSU-Py), and others.[67-69]

3.6 COFs

As an emerging porous polymer material, COFs exhibit various advantages such as low density, large specific surface area, uniform pore size distribution, adjustable structure, and high chemical stability.[70, 71] These properties have positioned COFs as promising electrode materials, finding applications in diverse fields including seawater desalination, organic pollutant adsorption, and ion capture.[72] The Li^+ adsorption selectivity and cycling stability can be effectively improved by embedding lipophilic functional groups in the nanochannels of COFs and by improving hydrogen bonding sites on the surface of COFs during synthesis.[73]

It is challenging to accurately detect the distribution and motion of lithium ions in 2D COFs through experimental techniques.[74] In response, computer simulations have proven invaluable, enabling precise characterization of lithium-ion distribution and motion states. Previous research has employed DFT calculations to study adsorption configurations and energy barriers along the migration path. Ab-initio MD (AIMD) simulations have elucidated the motion of lithium on several picoseconds (ps) trajectories in COF-5 and explored the influence of charge distribution in COFs with different functional groups on separation performance.[75]

To improve the practical applications of both MOFs and COFs, their hybrids have been developed,[76] namely MOF@COF and MOF-in-COF composites, where MOFs serve as coating layers with COFs acting as substrates, introducing dynamic covalent bonds, covalent condensation or π-π stacking interactions of COFs structure into MOFs system.[77, 78] In the composite system, MD simulation could be employed to facilitate the direct analysis of ion diffusion in membranes, enabling the strategic design of diverse MOF/COF composites tailored to specific applications.

4. Materials informatics for potential design of lithium-ion adsorbent materials of CDI

The field of materials informatics is a subfield of materials science that has great potential in providing theoretical guidance for CDI electrode materials, performing post-processing analysis of characterization techniques, virtually screening compounds with customized properties, and describing lithium capture processes at interfaces under complex conditions. Experimental, theoretical, simulation and data-intensive science have been widely used in the exploration of new materials.[79] Within this paradigm, the integration of materials informatics has demonstrated the potential to shorten the development and commercial cycles associated with new materials.[80] The convergence of data science with advanced materials research and manufacturing practices exemplifies a trajectory of progress across disciplines.[81] The computational design and laboratory synthesis of new materials have been demonstrated through several successful examples, establishing these methods as a novel avenues for material discovery and optimization. Atomic computing technology can predict the structural properties of electrode materials with high accuracy, allowing the interaction between atoms and the environment to be accurately and effectively simulated.

4.1 DFT

First-principles quantum mechanical calculations, based on DFT, possess the capability to accurately predict the structural properties of electrode materials before their experimental synthesis. These calculations allow for precise and efficient computation of atom interactions solely from quantum mechanics, electrostatic potential distribution, charge transfer, and others, providing in-depth insights into the adsorption mechanisms of specific lithium-ion adsorption materials and enabling theoretical predictions of structural improvements.[82, 83] Additionally, computationally driven databases, typically harnessing crystal structure information from experimental databases, offer a robust approach for discerning patterns and correlations from previously inaccessible datasets. For instance, Iklima Oral et al. investigated the selective complexation of Li^+ over other metal ions by DFT calculations, for 15-, 12-, and 9- membered CEs and their derivatives, where the best selectivity of Li^+ over Mg^{2+} was achieved by thio-benzo-15-crown-5 (thio-B15C5).[49] Experimental liquid extraction and DFT calculations conducted by Rey Eliseo C. Torrejos et al. revealed that crown ethers with small cavity sizes exhibited good selectivity, whereas the larger radius of M^+ (M^+=Li^+, Na^+, K^+) coordinated with bulky structures and higher O-M^+ distances hindered effective adsorption.[84]

4.2 MD

Based on classical mechanics, quantum mechanics, and statistical mechanics, MD simulation studies the structural properties of molecular systems by numerically solving their motion equations with computers. This approach offers valuable insights into the energy and structure of a system at a specific temperature and time, elucidating microscopic structural changes occurring within picoseconds. The MD method is widely used in exploring liquid electrolytes, deriving the macroscopic properties of electrolytes such as ionic conductivity and dielectric constant, and revealing the reaction mechanism at the electrode-electrolyte interface.[74, 85]

Through MD simulation, researchers can study the selectivity of ion extraction materials for different ions, simulate the diffusion path and diffusion barrier of lithium ions inside the crystal, and analyze the dynamic evolution of the adsorption model.[86, 87] Additionally, the influence of external environment on lithium ion adsorption can be studied, such as electric field intensity, temperature, stress, and concentration. MD simulation also enables the analysis of the capacity and cycling stability of Li^+ adsorption, as well as the mechanism of Li^+ diffusion in the mixed saline solution.[88-90]

4.3 ML

ML, a fundamental component of artificial intelligence, possesses the potential to rapidly acquire knowledge from historical data, thereby catalyzing revolutionary advancements across diverse domains.[91] With the deepening combination of big data and artificial intelligence, data-driven ML has made great progress in structural design, material screening, and performance prediction. The combination of ML and materials science has significantly accelerated the development of novel functional materials, including 2D materials, single atomic catalysts, and organic lithium-ion battery materials.[92-95] The concept of designing catalytic substances using high-throughput screening and ML methodologies has also found applicability in the design of electrode materials with high selectivity and stability.

Employing ML facilitates swift screening of potential lithium extraction materials, providing theoretical knowledge for diverse extraction mechanisms. Details of ML analyses leverage descriptors such as metal-oxygen vacancy size, electronegativity, electron transfer ability, and structural stability. These descriptors help identify inherent patterns, guiding the structural design of lithium-ion adsorption materials.

4.4 FES

FES has proven instrumental in resolving numerous challenges encountered in CDI experiments, encompassing tasks such as predicting the concentration distribution of ions in the electrode materials, determining ion diffusion paths and transport rates, addressing the ion transfer/diffusion kinetics, and some complex puzzles.[96-98] Meanwhile, multi-physics field models composed of electrochemistry, fluid dynamics, mass transport, heat transfer, and solid mechanics have been developed to investigate the effects of macroscopic performance.[99, 100]

Although FES may provide visual and macroscopic insights, electrochemical reactions are complex and multi-scale, necessitating the consideration of thermodynamic and kinetic factors across various temporal and spatial scales at both micro and macroscopic levels. Particularly at microscopic scales, FES encounters limitations. Therefore, integrating methods such as DFT, MD, ML, and FES is essential for tackling the challenges encountered in electrochemical research. This integrated approach enhances the ability to address complex problems, generating valuable theoretical data and insights that are challenging to verify experimentally.

The intricate mechanism of ion selectivity in CDI necessitates diligent efforts in designing optimal electrode materials. The electrochemical performance of optimal electrode materials in CDI is influenced by external factors such as electric fields, stress, and temperature. The field of materials informatics provides a perfect solution by simulating ion-material interactions and offering valuable insights. The procedure employed in the explanation of the mechanism for lithium selective materials is illustrated in Figure 3. The design of electrode materials is mainly reflected in the selection of adsorption, where systematic high-throughput screening or ML methods can guide the theoretical designs, DFT, MD, and FES explore adsorption mechanisms from physical and chemical aspects, leading to the discovery of superior lithium extraction materials.

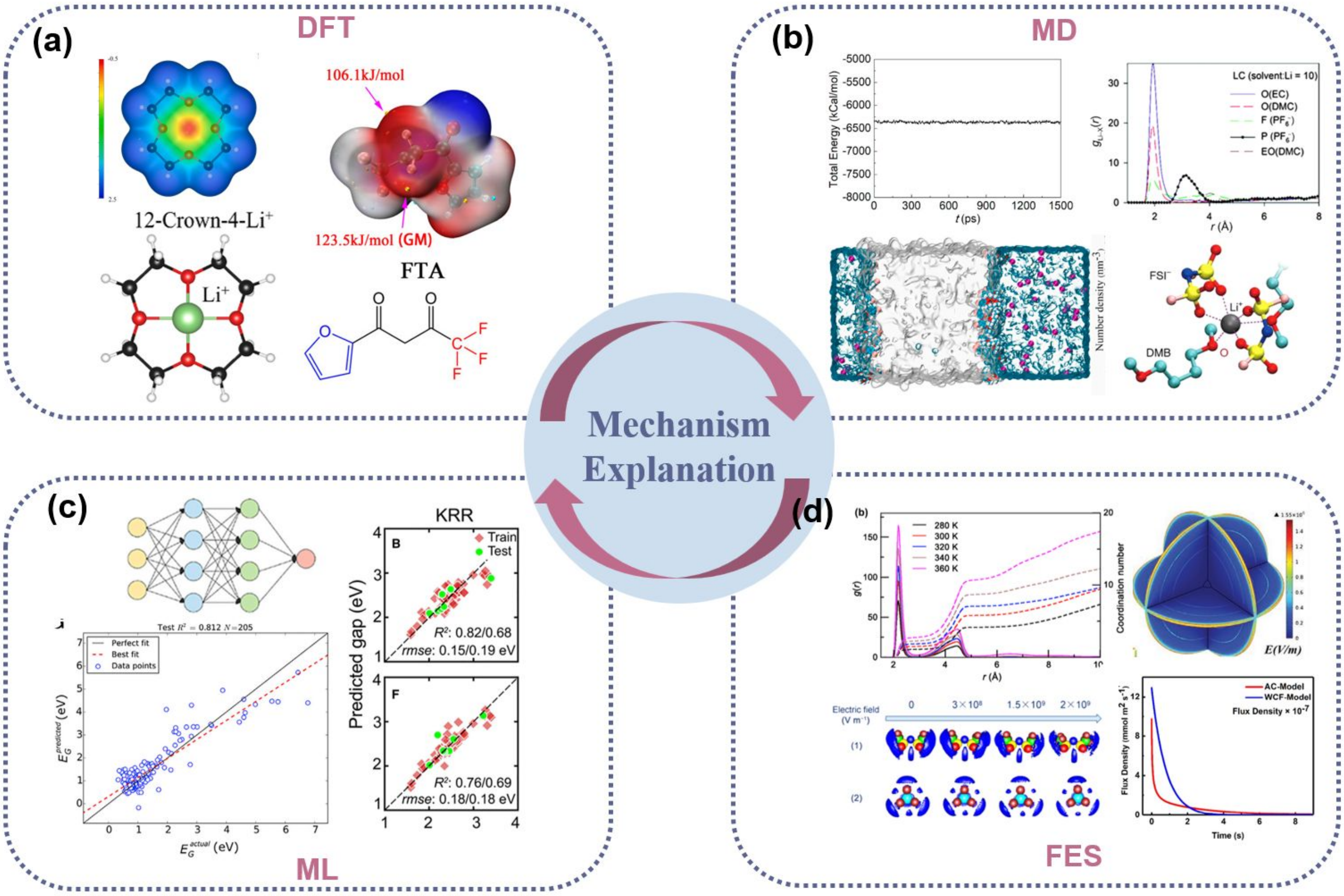

Figure 3. Schematic diagram of DFT, MD, FES, and ML methods, used in the mechanism explanation for the lithium selective materials, developed in Computational materials science and engineering. (a) The DFT mechanism diagram. Reproduced from ref. 82, with permission from Elsevier, Copyright 2021; Reproduced from ref. 83 with permission from The Global Home of Chemical Engineers, Copyright 2023. (b) The radial distribution function and dynamic pattern derived from MD. Reproduced from ref. 88 with permission from Elsevier, Copyright 2020; Reproduced from ref. 89 with permission from Elsevier, Copyright 2022; Reproduced from ref. 90 with permission from The Royal Society of Chemistry, Copyright 2016. (c) The data fitted by ML. Reproduced. from ref. 93 with permission from Cell Reports Physical Science, Copyright 2021; Reproduced from ref. 94 with permission from ACS Publications, Copyright 2016; Reproduced from ref. 95 with permission from ACS Publications, Copyright 2018. (d) The temperature and external field influence carried by FES. Reproduced from ref. 96 with permission from ACS Publications, Copyright 2018; Reproduced from ref. 97 with permission from Elsevier,

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Generally, selective structures characterized through experimental methods have found application in electrochemical lithium extraction. ML techniques, combined with existing lithium extraction materials and material databases such as COFs, MOFs, 2D materials, and CEs, facilitate the investigation into the underlying mechanisms governing lithium adsorption selectivity. The ion-selective electrode materials, after undergoing high-throughput and ML simulation, are found to exhibit superior lithium-ion selectivity, adsorption capacity, and stability characteristics in the solvent. DFT simulation, incorporating the implicit solvent effect, provides deeper insights into the adsorption mechanisms. Combined with MD simulation, the transport and movement of water molecules on the electrode surface can be simulated. FES models the internal movement of lithium ions under specific pressure and temperature in mixed solutions, thereby facilitating the lithium extraction from seawater. In a word, lithium extraction materials screened out by high-throughput and ML simulation, employing DFT and MD simulations in computational chemistry elucidate the underlying mechanisms and dynamic aspects of adsorption reactions, and FES simulation enhanced chemical and physical parameters in the CDI process. The details are found in Figure 4.

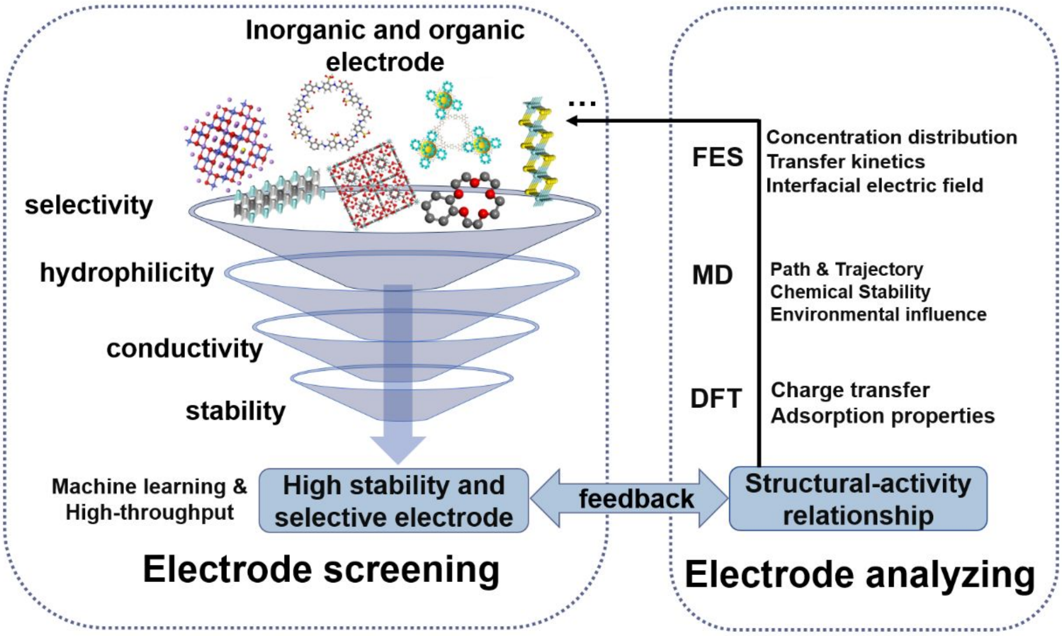

Figure 4. Schematic diagram of DFT, MD, FES, and ML methods used for lithium extraction materials, where ML and high-throughput for the electrode screening, DFT, MD, and FES for the electrode analyzing.

5. Outlook

In summary, CDI stands as a promising technology for seawater treatment, specifically for lithium extraction through electrochemical processes. The key challenge lies in achieving selective and stable lithium extraction, necessitating the selection of novel ion-exchange membranes and electrode materials to mitigate the influence of interfering ions. In this review, the promising materials used for future applications are specially summarized, including carbon-based materials, LIS, CEs, 2D materials, MOFs, and COFs. The pursuit of ideal materials with large specific surface areas, sufficient ion adsorption sites, high conductivity and ionic mobility, excellent hydrophilicity, and exceptional chemical stability poses a compelling scientific challenge in the field of lithium extraction. The screening of novel materials with exceptional performance and the quantitative exploration of structure-activity relationships are key focuses in the researches.

Materials informatics is expected to be an efficient methodology in CDI technology, facilitating the exploration of lithium extraction materials and providing valuable references for future development. Specifically, DFT enables the interpretation of the adsorption mechanisms of individual electrode materials, ML methodologies uncover common regularities in enhancing electrode materials, and high-throughput screening applies these regularities to explore potential novel electrode forms within databases. Furthermore, MD and FES simulations enable the modeling of the internal movement of lithium ions under specific pressure and temperature conditions in mixed solutions, enhancing our understanding of the intricate processes involved.

This study offers valuable and constructive guidance, providing essential support for the advancement of research methodologies and developmental strategies in the lithium extraction from seawater using CDI technology.

Acknowledgements

This research was funded by the National Natural Science Foundation of China (51909066), the Zhejiang Ocean University Talent Introduction Research Fund (No. JX6311103723), the Zhejiang Provincial Natural Science Foundation of China (No. LR23C160001), and the JST-ERATO Yamauchi Materials Space-Tectonics Project (Grant number: JPMJER2003). This work was performed in part at the Queensland node of the Australian National Fabrication Facility, a company established under the National Collaborative Research Infrastructure Strategy to provide nano and microfabrication facilities for Australia's researchers. We demonstrate our appreciation for English editing software such as ChatGPT, Grammarly, etc. as well as the English Editing Service, which helped us check grammatical errors in our manuscript.

Author contributions

L. Bai: Conceptualization, Writing – original draft; R. Xu: Writing – original draft; Wenjie Wu: Project administration, Writing – review & editing; C. Ma: Writing – original draft; S. Li: Writing – original draft; H. Gao: Writing – original draft; D. Luo: Writing – review & editing; B. Liu: Writing – review & editing; S. Arabia: Writing – review & editing; Y. Zhao: Writing – review & editing; Z. Liu: Supervision, Writing – review & editing; Y. Yamauchi: Supervision, Writing – review & editing; X. Xu: Conceptualization, Project administration, Writing – review & editing

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