

Review

Prelithiation of electrodes in lithium-ion capacitors: A review

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Abstract: Lithium-ion capacitors (LICs) are one of the modern state-of-the-art hybrid capacitors, comprising a high potential window and imparting a higher energy density than supercapacitors (SCs). These LICs encompass elevated power density and a longer life span than lithium-ion batteries (LIBs). Preparation of high-performance electrode materials with electrochemically active microstructure and prelithiation are two efficient approaches to fabricate highly efficient LICs. But it comes across as a real dilemma of low initial Columbic efficiency if only microstructure is considered as an efficient way to enhance the performance. Nevertheless, prelithiation plays a crucial role in the manufacturing of LICs, improving the initial Coulombic efficiency and enlarging the voltage window. This paper reviews the recent lithiation approaches for lithium-ion capacitors by providing their methods and discussing their results concerning their energy and power density.

Keywords: electrodes; columbic efficiency; prelithiation; LIC; energy density

1. Introduction

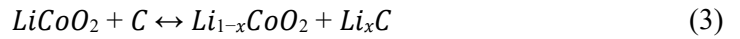
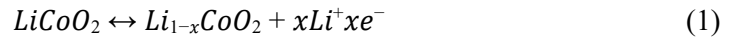
An increase in the market size of EVs and consumer electronic goods like computer systems and mobiles has swayed the research attention towards high-density energy storage systems [1]. These energy storage systems should be capable of generating fair output power and safe operational characteristics to be used in both vehicles and consumer electronic applications [2]. Among all types of energy storage devices, lithium-based materials are the most commonly used commercial systems to store energy from renewable energy sources. Among most lithium-based energy storage systems, LIBs have become the most widely used electrochemical energy storage device due to their superior energy density values, much needed for modern-day EVs and consumer electronics [3,4]. Nonetheless, both LIB's and SCs exhibit a notable discrepancy in performance due to their inability to meet the demands of fast charging and slow discharging. Thus, to overcome such discrepancy, an electrochemical energy storage device needs to be able to have both a high energy density and a high power density in order to achieve these requirements [5].

LICs are hybrid energy storage systems where SC's type cathode and LIB's type anode are used to derive high power and energy densities simultaneously. Conventionally activated carbon is used as a cathode material, which imparts greater power density, and compositions like $\text{LiTi}_5\text{O}_{12}$ are used as anode materials, which result in higher energy density. This asymmetric device construction makes these devices appropriate for both consumer electronics as well as automotive mobility. It is well known that the

electrochemical properties of LICs can be easily tuned using prelithiation, where anions move from or towards the cathode, where intercalation occurs [6]. Thus, prelithiation is one of the preferred strategies adopted to compensate for the intercalation. However, the commercialization of LIC increased traction some time ago when graphite was started to be probed as a potential negative electrode material, anticipating its capability to improve energy density and potential window [7].

2. Need for prelithiation

In LIC's, prelithiation is expected to compensate for the loss of Li-ions that occurred due to the formation of solid electrolyte interphase towards the anode terminal since a higher proportion of activated carbon is expected to facilitate higher Li-ion loss [8,9]. Hence, selection and preparation of appropriate cathode and anode materials is one of the useful approaches to make possible LICs to attain high energy density at high power density. However, despite the recent developments in electrode materials, we come across the problem of low initial coulombic efficiency in LICs. For example, let us consider $\text{LiCoO}_2//\text{graphite}$ cell where Li-ions de-intercalated from LiCoO_2 and intercalated towards graphite at the time of charging. The reaction corresponding to the movement of Li-ions is as follows:



The above equations represent the reaction of entire cells, i.e., on the anode's and cathode's, respectively. When the anode potential falls below 1V versus Li^+/Li , parasitic processes on the anode surface cause the organic electrolyte to be simultaneously reduced to create the solid electrolyte interphase (SEI) layer during the intercalation of Li-ion. While it is widely known that this SEI layer is essential in preventing electrolyte ions from co-intercalating into the electrode bulk, some lithium ions may be permanently caught in the electrode during lithiation, which causes a rapid shift in the concentration of the electrolyte and the consumption of lithium ions, resulting in low initial coulombic efficiency and high initial capacity loss [10,11]. **Figure 1** shows the formation of SEI in $\text{LiCoO}_2//\text{graphite}$ lithium ion batteries. A significant challenge for LICs is the absence of a lithium source in the cathode, which limits energy density. Here, prelithiation plays a crucial role in the manufacturing of LICs to improve the initial coulombic efficiency and boost energy density [12]. It also provides the extra Li-ions to make up the initial loss in the capacitor [13–16].

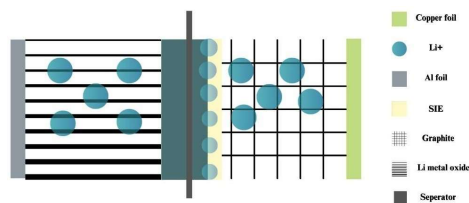


Figure 1. Formation of SEI in $\text{LiCoO}_2//\text{graphite}$ LIBs.

3. Prelithiation approaches

Prelithiation is a strategy to preemptively lithiate pre-doping of extra Li-ions into electrodes before battery/capacitor assembly for active Li⁺ loss compensation [17–22]. Currently, numerous prelithiation processes are applied for recovering initial irrevocable capacitance compensation [23–27]. The prelithiation approaches for LICs can be broadly classified into two types, namely the anode prelithiation and cathode prelithiation. Herein all the prelithiation approaches mentioned are expected to enhance the LICs performance.

3.1. Anode prelithiation

In order to enhance the initial columbic efficiency and improve the life span of LIC's, prelithiation of the anode is one of the most effective strategies. Here the anode can be prelithiated by various processes like chemical prelithiation, electrochemical prelithiation, additive prelithiation, and metal lithium foil/powder prelithiation [28,29]. But they were mainly fabricated through electrochemical prelithiation and direct contact with a Li source [30]. These prelithiation approaches are discussed in detail.

3.1.1. Chemical prelithiation

In chemical prelithiation, organic molecules are used as the mediator for lithium transfer [31,32]. The first method deals with the prelithiation of anode with the chemical prelithiation method for SiO_x/Graphite composite anode. In this, lithium ararene (LiA) was used as a prelithiation reagent for the composite anode. Anaryl lithium impregnation method with 2 min of prelithiation resulted in the increased ICE from 88% to 98%. They found that the ICE can be easily tuned using impregnation time and LiA reagent concentration. The study was limited to half-cell testing and did not assess the performance of the prelithiated anode in a complete battery system [33]. Another chemical prelithiation method deals with the use of 2-methyltetrahydrofuran (MTHF) as prelithiation solvent. MTHF solvent was used to prelithiate soft carbon (SC), where the time for prelithiation was set to be ~5 min, which resulted in the display of a small potential of 0.21 V vs. Li/Li⁺. As prelithiated SC was found to be chemically stable and deployed as cathode, excellent capacitance retention was observed even after 5000 charging and discharging cycles. Also, the energy density was computed at ~69.4 Wh/kg, and the power density was 5.7 kW/kg in the fabricated LIC made up of prelithiated SCs [34].

3.1.2. Sacrificial prelithiation material

In order to prevent the use of a metallic lithium electrode during the initial charging phase, provide lithium ions into the anode. It is to be noted that Li₃N is one of the best salts that could be used as a sacrificial source and can be easily produced by reacting lithium with nitrogen gas. Li₃N is investigated as a novel sacrificial material that offers a number of special benefits over existing prelithiation materials. **Figure 2** shows the configuration of LIC with an

AC/Li₃N electrode.

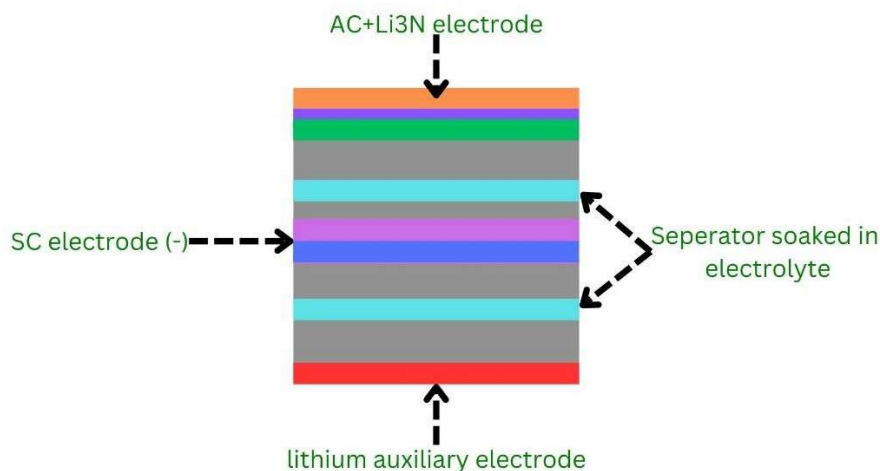


Figure 2. The configuration of LIC with AC/Li₃N electrode, SC electrode and lithium auxiliary electrode.

Here, the LIC was constructed using AC/Li₃N cathode and SC as anode, which showed a high energy density of 214.7 Wh kg⁻¹ at 0.18 kW kg⁻¹ and 65.0 Wh kg⁻¹ at a high power density of 19.5 kW kg⁻¹ with excellent cycling stability (75.1% retention after 5000 cycles at 3.2 kW kg⁻¹). Overall, the approach of using Li₃N as prelithiation material offers several advantages, including avoiding electrolyte decomposition, reducing the amount of prelithiation material needed, and simplifying the construction of pouch LICs. The results obtained in this work revealed the difficulties in obtaining high energy and power densities owing to the divergence of electrode kinetics.

Anaiz et al. reported about the use of sacrificial salts as a prelithiation tactic to get efficient LIB and SCs. The prelithiation procedure includes the usage of sacrificial salts, especially dilithium squarate (Li₂C₄O₄), in a LIC prototype fabricated in a pilot line. This process gets done during the first ten cycles, and after the needed gas exhaust, the electrochemical performance of prototypes was found to be satisfactory [35].

Veluri et al. proposed the construction of a LIC with a long life span (85% capacity retention after 10,000 charge-discharge cycles at 1A/g and a high energy density of 80 Wh/kg and a high power density of 8.4 kW/kg) utilizing carbon materials obtained entirely from petroleum coke. No additional prelithiation was done; instead, the suggested LIC was prelithiated by keeping a small amount of Li metal foil (ratio of Li to disordered carbon, 1:7) on the anode surface to supply lithium during the initial charging stage. High-surface-area activated carbon made from petroleum coke served as the cathode, and low-surface-area disorder carbon served as the anode. **Figure 3** demonstrates the capacitance retention of petroleum coke-based LIC vs. the petroleum coke-based supercapacitor [36].

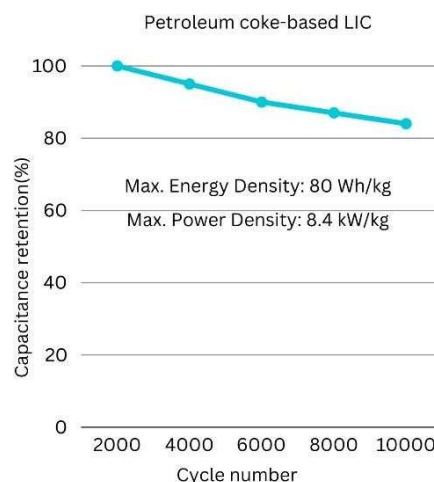


Figure 3. Petroleum coke-based LIC vs the petroleum coke-based supercapacitor.

Kong et al. reported the LICs with anode compositions of high-content P and N co-doped porous carbon nanomaterials (X-PNC) and cathode made using pyrrolic-N-doped porous carbon nanomaterials (X-NC). The working electrodes in the half-cells were X-PNC and X-NC electrodes, whereas the counter electrode was lithium metal foil. With great cycling stability (75.1% retention after 5000 cycles at 3.2 kW kg^{-1}), the built-in LIC demonstrated a high energy density of 214.7 Wh kg^{-1} at 0.18 kW kg^{-1} and 65.0 Wh kg^{-1} at a high power density of 19.5 kW kg^{-1} [37].

Han et al. reported the fabrication of LIC built utilizing pre-lithiated nitrogen-doped porous carbon (NPC) anodes and hierarchical porous carbon (CHPC) cathode electrodes generated from coal. It has a gravimetric capacitance of 139 mAh g^{-1} cathode at 0.5 A g^{-1} . In this work, the NPC electrode was in contact with Li metal foils for 45 minutes for prelithiation without any use of a polypropylene membrane. The CR2032 button cells fabricated in this work demonstrated a specific capacity of 139 mAh g^{-1} with 99% retention after 1000 cycles of charging and discharging at 1 A g^{-1} . The energy density was estimated to be 220 Wh kg^{-1} at a specific power of 805 W kg^{-1} . **Figure 4** shows the schematic representation of pre-lithiated cells prepared using NPC and CHPC [38].

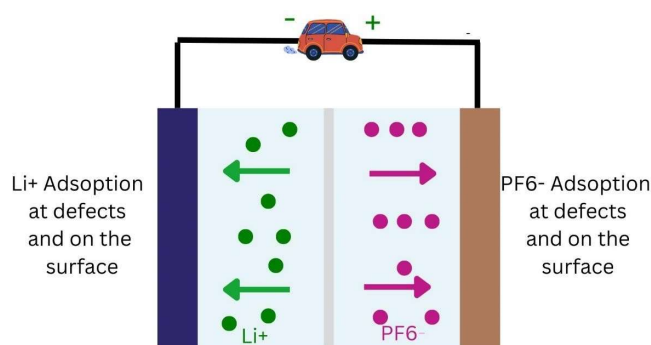


Figure 4. The diagram illustrating charge-storage mechanisms (gray ball: carbon atoms; blue ball: oxygen atoms; red ball: nitrogen atoms; yellow ball: sulfur atoms; purple ball: PF_6^- anion; green ball: Li^+ cation).

3.1.3. Prelithiation by charging/discharging a half cell

Tan et al. reported the fabrication of LIC by means of hollow porous $\alpha\text{-Fe}_2\text{O}_3$ nanoparticles and carbon nanospheres derived from glucose as anodes and cathodes, where excellent electrochemical characteristics were observed. Half cell prepared using hollow porous $\alpha\text{-Fe}_2\text{O}_3$ nanoparticles was prelithiated using charging/discharging at 0.2 A g^{-1} , resulting in 107 Wh kg^{-1} energy density at 0.24 W kg^{-1} and an energy density of 86 Wh kg^{-1} at a power density of 9.68 kW kg^{-1} . **Figure 5** shows typical LIC comprising important parts like cathode, anode, and electrolyte layers. Besides, it displayed towering-capacity retention $\sim 84\%$ after 2000 CV cycles $\sim 1 \text{ Ag}^{-1}$ [39].

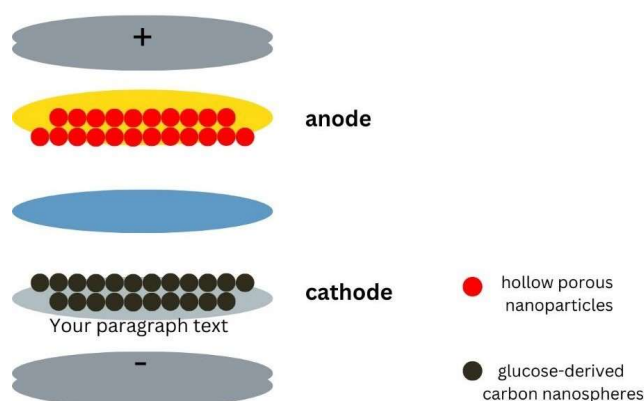


Figure 5. Graphical representation of approach for the Lithium-ion capacitor in coin cell.

An additional step in the prelithiation process is used to activate a graphite half-cell by charging and discharging it five times before assembling it into a full-cell configuration. New peaks were observed in the XPS spectra, and atomic percentages change as the graphite electrode is exposed to particular electrolytes. These changes in elemental composition and chemical bonding occur during this process. These alterations show that during the prelithiation phase, an artificial solid electrolyte interphase (SEI) is formed on the graphite surface [40].

Among the other techniques is the use of lignin, the second most common natural polymer, which is also the primary waste byproduct from the pulp and paper industry and is used as an electrode material for lithium-ion batteries. Here, lignin is transformed into two types of functional carbons that act as the electrode materials in LICs by controlling the porous framework and graphitic degree with the goal of building high-performance LICs. In a manufactured capacitor, lignin-derived hierarchical porous carbon (LPC-3) served as the anode and lignin-derived graphitic carbon (LGC-1500) as the anode. The assembled LGC-1500//LPC-3 LIC delivered a high-energy density of 97 Wh kg^{-1} , a high-power density of 11.4 kW kg^{-1} , and a superior cyclic stability of 92.3% after 5000 cycles at 1 Ag^{-1} [41].

The “in-situ electrolyte” is a sustainable alternative for the realization of high-power devices [42,43]. These devices, including electric double-layer and metal-ion capacitors prepared using waste products for carbon synthesis,

enable the realization of Li- and Na-based EDLCs containing aqueous, water in salt, and organic electrolytes, as well as an alternative for Li-ion capacitors. The prelithiation process involves generating a lithium inventory to ensure the formation of a SEI on the anode and to avoid concentration gradients within the electrolyte solution. This is achieved by utilizing activated carbon (AC) loaded with a selected lithium salt to create a LIC with a high and tunable lithium inventory directly during the assembly process. A mixture of carbonaceous materials, including hard carbon and graphite, serves as the anode material. The study does highlight some of the difficulties in determining the ideal concentration of activating agents for carbon porosity. The fabricated LIC displayed a power density of 10 kW kg^{-1} and a specific capacitance of $\sim 21 \text{ Fg}^{-1}$ [44].

The developed hybrid LICs with a prelithiated graphite anode and two cathodes exhibit superior electrochemical performance, including high energy density, power density, and capacity retention. The prelithiation process was conducted by lithiating the graphite anode using a perforated LiCO_2 electrode. The anode material was composed of graphite, carbon black, carboxymethyl cellulose (CMC), and styrene butadiene rubber (SBR), and the cathode material was prepared using active carbon, carbon black, CMC, SBR, and LiCO_2 . Within a voltage range of 2.2 V to 3.8 V, the hybrid LICs showed excellent discharge and charge performance, together with high energy and power densities. After 300 cycles, the hybrid LICs capacitance dropped by less than 1%, demonstrating exceptional cycling performance and capacity retention of up to 99% [45].

The potential for easy and low-cost fabrication of LICs and the feasibility of adopting new technologies such as sodium ion and potassium ion capacitors are extensively reported. Arnaiz et al. reported in-house synthesized olive pit-derived hard carbon (HC) as anode and AC as cathode material. In this report, 1 M LiPF_6 in ethylene carbonate (EC) and dimethyl carbonate (DMC) with 3 wt% of vinylene carbonate (VC) was used as an electrolyte. The energy density of LICs, with a 2- to 3-fold enhanced performance, was observed compared to EDLCs in terms of power density and specific capacitance [46].

Anothumakkool et al. reported the development of prelithiated electrodes in LIC, which could help overcome a significant scientific obstacle in the field. The prelithiation process in this paper involves a cascade-type mechanism with two consecutive irreversible reactions within the positive electrode, where oxidative electro-polymerization of pyrene moieties releases electrons and protons. These protons are captured by Li_3PO_4 and exchanged for a stoichiometric amount of Li^+ into the electrolyte, aiming to compensate for the loss of active lithium and electrons during cycling and reach full utilization of the active materials. Carbon black and activated carbon are used as anodes in the construction of LIC, whereas pyrene and Li_3PO_4 additives are used as

cathodes. The prelithiation process used in the construction of the LIC revealed an energy density of 63 WhKg^{-1} . However, challenges in commercialization and inability to achieve long-term stability, instability in ambient air, and low electron conductivity limit its applications [47].

Park et al. [48] reported the feasibility and advantages of using prelithiated Si/SiO_x nanospheres as an advanced negative electrode. This LIC shows improved cycle performance and excellent energy density compared to conventional graphite electrodes. When compared to traditional graphite LICs, prelithiated C-coated Si/SiO_x nanospheres have better reversible capacity and rate capability. Traditional LICs employing prelithiated C-coated Si/SiO_x nanospheres exhibit significantly better cycle performance and rate capability. The as reported LIC achieves an energy density that is more than twice as high as conventional LICs.

Sugiawati et al. [49] reported another prelithiation technique that is suggested with Li⁺ to reduce its potential, which would bring the insertion voltage of anodes closer to 0.1 V. LICs can function within a broader voltage window, which increases the voltage gap between the anode and cathode. When repeating Li⁺ insertion/desorption procedures, the extra Li⁺ can also restore the consumption of lithium-ion sources. A half-cell containing anode and Li-foil without separators can be assembled, and the anode can be prelithiated for several hours by allowing the anode to have a lower Li⁺ insertion plateau. This chemical reaction between metal oxides and lithium sources results in an energy density of around 200 Wh kg^{-1} .

3.2. Cathode prelithiation

The energy density of lithium-ion batteries is greatly reduced by the irreversible loss of lithium during the first cycle. Although anode prelithiation is a popular solution to this issue, it has drawbacks of its own, including strong chemical reactivity and instability in battery production and environmental conditions. Therefore, cathode prelithiation is far less complicated in contrast [50]. Many cathode prelithiation additions have been created to date. Sacrificial Li salt additives, for instance (Li₂C₂, Li₃N, Li₂O, etc.), can effectively offset the ICL. However, undesirable gaseous N₂, CO, or CO₂ will inevitably evolve. On the other hand, ternary compounds (such as Li₅FeO₄, Li₆CoO₄, and Li₂NiO₂) don't raise issues with gaseous evolution when used as cathode prelithiation additives. We will talk about various cathode prelithiation techniques in the following section [51–55].

3.2.1. Cathode prelithiation approaches

Sacrificial Li-rich additives have recently been considered a promising approach for cathode prelithiation. Due to their irreversible specific capacities during the initial charging process to provide an additional Li⁺ source for anode prelithiation, a number of Li-rich additives, such as inorganic binary Li compounds (e.g., Li₃N and Li₂S), inorganic ternary Li compounds (e.g., Li₅Re

and Li_6CoO), and organic Li salts (e.g., Li_2DHBN and $\text{Li}_2\text{C}_2\text{O}_4$), have been investigated as cathode prelithiation additives. However, gas escape is typically associated with the delithiation of cathode prelithiation additives and must be eliminated using a secondary sealing method. A workable and economical prelithiation strategy has been proposed, along with a compatible modification that involves the introduction of Li_2S as a cathode prelithiation additive and lithium difluoro (oxalato) borate (LiDFOB) as an electrolyte additive. The built LICs demonstrated an exceptional capacity retention of 92% after 10,000 cycles at a current density of 1 A g^{-1} , along with a high energy density of 155 Wh kg^{-1} at a power density of 210 Wkg^{-1} and retaining 32 Wh kg^{-1} at an ultrahigh power density of 105 kW kg^{-1} [56].

3.2.2. Capacitive adsorption approach

Using capacitive adsorption, lithium ions are pre-stored in the cathode in this method [57]. Additionally, in the initial cycles, they serve as the supply of lithium to accomplish prelithiation. In the mean time, the capacitive adsorption's charge voltage can be altered to control the prelithiation level. The complete LIC device prelithiated using this technique has a specific capacity of 31.4 mAh g^{-1} .

Figure 6 shows the graphical representation of prelithiation by capacitive absorption. LICs experience capacity loss during their initial charge and discharge cycles due to irreversible lithium intercalation behaviors. Even though prelithiation frequently uses lithium metal, there are concerns about safety and operational complexity. As an alternative, cathode sacrificial lithium salts (CSLSs) present a feasible option because of their affordability, ease of use, and safety. This review compares several prelithiation techniques and thoroughly examines the function of prelithiation in LICs. In particular, it explores the use of CSLSs for prelithiation methods, going over the mechanisms of lithium extraction from them as well as the effects of doping levels and intrinsic properties on LIC performance.

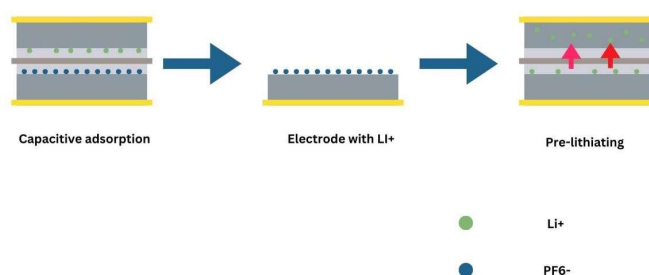


Figure 6. The prelithiation method by capacitive absorption.

The prelithiation procedure begins with the identification of sacrificial lithium salts that have a high capacity, a high degree of irreversibility in the first cycle, a low voltage for lithium extraction, and no “dead” materials left over after the initial charge. Additionally, it makes use of a number of prelithiation technologies, including the in-

situ chemical (ISC) and in-situ electrochemical (ISEC) methods. CSLs, which are used for prelithiation in LICs, are essential to the process, which attempts to make up for the irreversible capacity loss in LICs and is critical to their electrochemical performance [58]. **Table 1** shows the example of LICs using various prelithiation methods and their energy and power densities.

Table 1. Examples of LICs prepared using various prelithiation methods.

Prelithiation materials	Cathode	Anode	Energy density (Wh/kg)	Power density (kW/kg)	Cyclic stability	Ref.
Li		Soft Carbon (SC)	69.4	5.7	96.7 % after 5000 cycles	[34]
Li ₃ N	AC/Li ₃ N	SC	214.7	19.5	75.1% retention after 5000 cycles at 3.2 kWkg ⁻¹	[8]
Li	activated carbon	low surface area disordered carbon	80	8.4	85% capacity retention after 10,000 cycles	[36]
Li	X-NC	X-PNC	214.7	19.5	75.1% retention after 5000 cycles	[37]
Li	CHPC	NPC	220	0.805	90% capacity retained	[38]
Li	GCNS	HPNP	86	9.68	84% after 2500 cycle	[39]
Li	LGC-1500	LPC-3	197	11.4	92.3% retention after 5000 cycles	[41]
Li ₂ S	NPCNF	MNCNF	155	0.210	92% after 10,000 cycles	[12]

Basically, Li metal, Li₃N, and Li₂S are extensively used as lithium sources for prelithiation, and it is observed from **Table 1** that, among all prelithiation sources, Li metal is observed to be the best source, notably improving the energy and power densities of devices. Also, it is understood that AC/Li₃N is the best cathode material when used with SC as an anode material, where retention ration was observed to be around 75% after 5000 cycles.

4. Conclusion

A review of prelithiation processes on LICs reveals its significant potential to enhance the performance and efficiency of these devices. The various methods have been discussed with respect to prelithiation, such as chemical prelithiation, lithiation by direct contact of lithium foil, and prelithiation by half cell assembly. Further advancements in prelithiation techniques and materials expected to drive the continued improvement of LICs are discussed. In conclusion, electrochemical prelithiation holds great promise for enhancing the performance and efficiency of LICs. Continued research and development in this area is essential to unlocking the full potential of prelithiation and advancing the field of energy storage technologies.

Conflict of interest: The authors declare no conflict of interest.

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