

Perspective

Enabling Rechargeability in Metal–Chlorine Batteries: The Critical Roles of Cathodes and Electrolytes

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Abstract

Metal–chlorine batteries, including Li–Cl₂, Na–Cl₂ and Ca–Cl₂ systems, have emerged as promising energy storage technologies in recent years, delivering impressive performance in terms of reversible capacity (up to 5000 mAh g^{−1}), current density (up to 16,000 mA g^{−1}), cycle life (up to 500 cycles) and low-temperature operation (down to −80 °C). The critical roles of cathodes and electrolytes, enabling the birth and performance enhancement of metal–chlorine batteries, are highlighted and discussed here, providing an overview of current research progress on metal–chlorine batteries at the same time. Finally, we demonstrate a forward-looking view of where metal–chlorine batteries are headed.

Keywords: metal–chlorine batteries; Li–Cl₂ batteries; Na–Cl₂ batteries; Ca–Cl₂ batteries; cathode; electrolyte

1. Introduction

Rechargeable batteries, with higher energy density, higher power density and longer cycle life, have become increasingly critical due to the rapid growth of emerging industries, such as wearable smart devices, electric vehicles, and clean energy storage [1,2]. As lithium-ion batteries, currently the dominant rechargeable battery technology, approach their theoretical performance limits, significant research efforts have shifted toward exploring alternative systems, including lithium–metal and post-lithium batteries [3,4]. For instance, lithium–metal batteries (e.g., lithium–oxygen [5] and lithium–sulfur [6] batteries) have attracted substantial attention owing to lithium's ultrahigh theoretical specific capacity (3860 mAh g^{−1}), which far exceeds that of graphite (372 mAh g^{−1}), the anode material widely used in conventional lithium-ion batteries. Furthermore, to address the scarcity of lithium resources, researchers are actively investigating other metal-based batteries, such as sodium (Na)-based [7] and calcium (Ca)-based [8] systems.

Building on this line of thought, in 2021, Dai and colleagues [9] first demonstrated two novel promising rechargeable metal–chlorine batteries, Na–Cl₂ and Li–Cl₂ batteries, converted from the primary Na–SOCl₂ and Li–SOCl₂ batteries, respectively. The Na–Cl₂ batteries, mainly reported in their work [9], delivered a discharge voltage of 3.5 V, a reversible capacity of up to 1200 mAh g^{−1} (based on carbon mass throughout this paper unless otherwise specified) and a cycle life up to 200 cycles. The energy density of Na–Cl₂ batteries, 1827 Wh kg^{−1}, is also significantly higher than that of other battery systems [10]. The Li–Cl₂ batteries, thoroughly investigated and reported in 2022 [11], exhibited comparable performance as the Na–Cl₂ batteries. By 2023, Chen and colleagues [12] extended the cycle life of Li–Cl₂ batteries to 500 cycles, while Sun and colleagues [13] achieved a



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unprecedented maximum discharge current density of 16,000 mA g^{−1} for Na-Cl₂ batteries, which was more than two orders of magnitude higher than previous reports [9]. In 2024, Dai and colleagues [14] demonstrated an ultralow-temperature Li-Cl₂ battery delivering a 1200 to 5000 mAh g^{−1} reversible capacity between −40 °C and −80 °C. In the same year, Sun and colleagues [15] introduced the first Ca-Cl₂ battery, expanding the family of rechargeable metal–chlorine batteries. Table 1 summarizes the performance of rechargeable metal–chlorine batteries from the literature.

Table 1. The performance of rechargeable metal–chlorine batteries.

Anode	Cathode	Electrolyte	Initial Discharge: Capacity (mAh g ^{−1})/Current Density (mA g ^{−1})	Charge–Discharge Cycling: Cycles/Capacity (mAh g ^{−1})/Current Density (mA g ^{−1})	Reference (Year)
Na	Amorphous carbon nanospheres	4 mol/L AlCl ₃ + 2 wt% NaFSI + 2 wt% NaTFSI in SOCl ₂	2800/50	100/500/150	[9] (2021)
Li	CO ₂ -activated defective graphite	1.8 mol/L AlCl ₃ + 1.8 mol/L LiCl + 2 wt% LiFSI in SOCl ₂	1911/50	140/600/100	[11] (2022)
Li	Metal–organic frameworks	4 mol/L AlCl ₃ + 2 wt% LiFSI + 2 wt% LiTFSI in SOCl ₂	7550/150	500/1000/150	[12] (2023)
Na	Bicontinuous N-doped carbon	4 mol/L AlCl ₃ + 2 wt% LiFSI + 2 wt% LiTFSI in SOCl ₂	2963/2000	250/500/2000	[13] (2023)
Li-Si alloy	Ketjenblack carbon	4 mol/L AlCl ₃ + 4 mol/L LiCl in SOCl ₂ + 90 vol% DCE	2720/100 *	250/500/500 *	[16] (2023)
Li	CO ₂ -activated Ketjenblack carbon	1 mol/L AlCl ₃ + 0.95 mol/L LiCl + 0.05 mol/L LiFSI in SOCl ₂	29,100/50 (RT) 8521/50 (−20 °C) 5532/50 (−40 °C) 4503/50 (−80 °C)	130/1200/100 (−40 °C) 70/1200/100 (−80 °C)	[14] (2024)
Ca	Graphite	6 mol/L AlCl ₃ + 1.3 mol/L LiDFOB + 1.2 mol/L CaCl ₂ in SOCl ₂	3264/100	100/200/100	[15] (2024)
Na	Ketjenblack carbon	4 mol/L AlCl ₃ + 1 mol/L NaFSI in MDCA and CDCl ₃ (1:1)	–	700/200/150 (−40 °C)	[10] (2025)

* Specific capacity and current density are based on the mass of Si.

The basic chemical reaction of Li-Cl₂ batteries, as an example, is presented as follows to briefly illustrate the working principles of metal–chlorine batteries. This working principle is also illustrated in Figure 1. The as-prepared Li-Cl₂ battery, a Li-SOCl₂ battery, initially discharges with an overall battery reaction of:

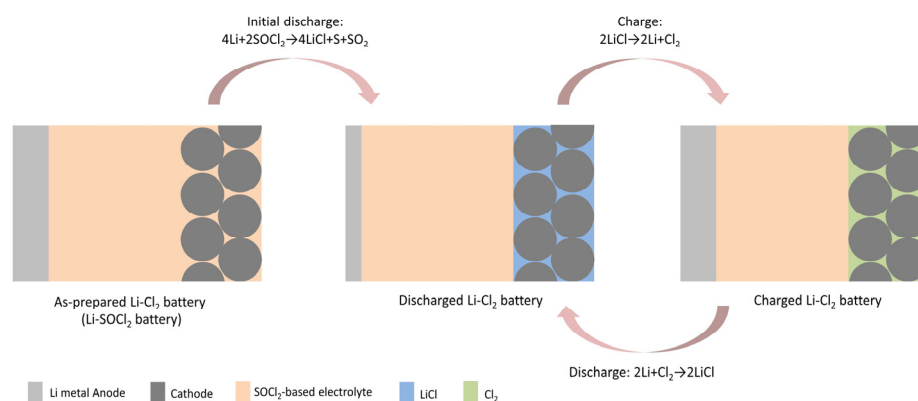
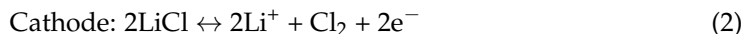


Figure 1. Schematic of Li-Cl₂ battery working principle.

During this process, LiCl was in situ formed on the cathode. The formed LiCl on the cathode was then oxidized to Cl₂ during the subsequent charging step and reduced back to LiCl during discharging as described in Equation (2). At the anode, simultaneously, the

deposition and stripping process of Li is depicted in Equation (3). The combination of these half-reactions yields the overall reaction of Li-Cl₂ batteries as shown in Equation (4). It should be noted that Equations (2)–(4) show only the main reaction of Li-Cl₂ batteries, while the actual entire reaction process is much more complex, which will be further explained later, though the full electrochemical pathway remains incompletely elucidated to date.



In brief, by charging and discharging a fully discharged Li-SOCl₂ battery, the rechargeable Li-Cl₂ battery is born. It seems very easy. However, the Li-SOCl₂ batteries had been remained strictly non-rechargeable for nearly five decades since it was truly born in the 1970s [17–19]. It was not until 2021 that this paradigm was overturned by the research of Dai and colleagues [9], whose breakthrough revealed that achieving reversibility hinges on two critical components: tailored cathode and optimized electrolyte. These findings, along with subsequent advances in metal–chlorine batteries, underscore the indispensable roles of cathode and electrolyte design. Both of these key roles, in the following, will be systematically analyzed, while the current research progress on metal–chlorine batteries will be given.

2. The Critical Roles of Cathode

Extensive research has focused on cathode material innovation, aiming to facilitate the development of metal–chlorine batteries while advancing their performance optimization. The amorphous carbon nanospheres (aCNSs) were synthesized and used as the cathode of Na-Cl₂ and Li-Cl₂ batteries by Dai and colleagues [9], which was found to be critical to subsequent reversible battery cycling. It is clear from Figure 2a that the cycling performance of aCNS is better than those of acetylene black (AB) and Ketjenblack carbon black (KJ) that are widely used in Li-SOCl₂ batteries. In another paper [11], they employed CO₂-activated defective graphite (DGr_ac) as the cathode of Li-Cl₂ batteries achieving a higher reversible capacity than that using defective graphite without a CO₂-activated (DGr) cathode, as shown in Figures 2b and 2c, respectively. The CO₂-activated KJ (KJCO₂), similarly, was used as cathode to construct a Li-Cl₂ battery by Dai and colleagues [14], delivering a longer charge–discharge cycling life (Figure 2d) than that of the KJ cathode (Figure 2a) in their previous research [9].

The cathode, as described earlier, assumes the role of holding the discharge products, LiCl (or NaCl, etc.), adsorbing the charge products, Cl₂, and providing electrochemical reaction interfaces, as schematically drawn in Figure 2e. Obviously, cathode materials that provide a larger surface area or pore volume are preferred for use in metal–chlorine batteries. Therefore, aCNS, DGr_ac and KJCO₂ with larger surface area and pore volume compared to the competition, as given in Table 2, were used accordingly and contributed to the remarkable performance in above studies. It should be noted that the relationship is not strictly linear. In other words, it is implied that not all pore space of cathode is valid. The cathode material, with larger effective pore volume, thus needs to be continuously found and investigated to further improve the performance of metal–chlorine batteries.

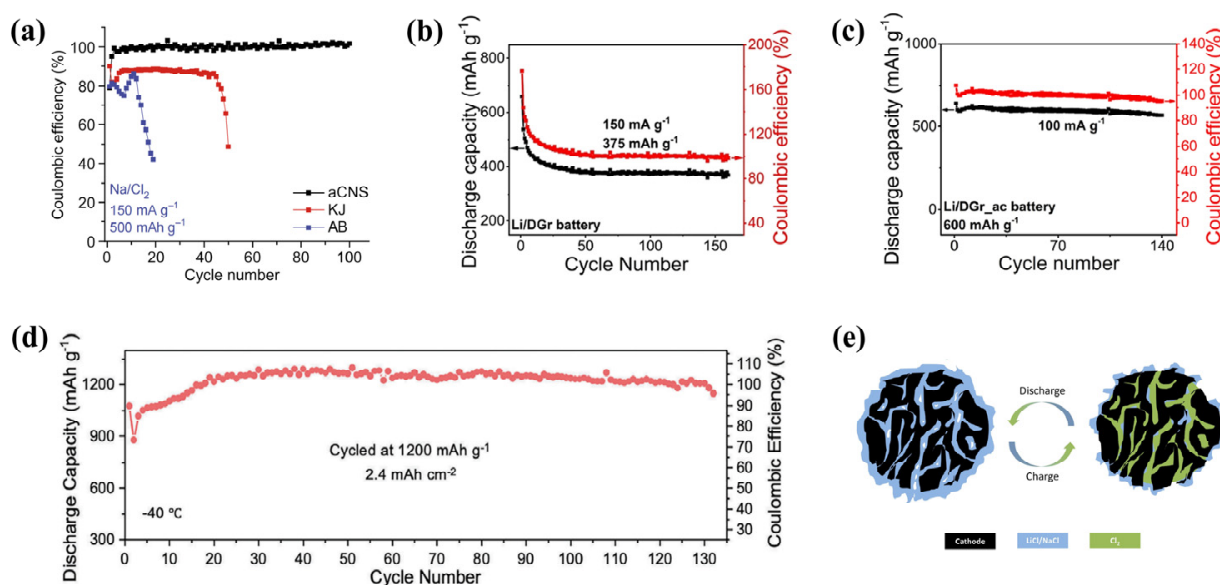


Figure 2. Critical roles of cathodes in metal–chlorine batteries. (a) Coulombic efficiencies of Na–Cl₂ batteries using aCNS, KJ and AB as cathodes [9]. (b) Cycling performance of a Li–Cl₂ battery using DGr as a cathode [11]. (c) Cycling performance of a Li–Cl₂ battery using DGr_ac as a cathode [11]. (d) Cycling performance of a Li–Cl₂ battery using KJCO₂ as a cathode [14]. (e) Schematic illustration of the working principle of the cathode of metal–chlorine batteries.

Table 2. Surface area and pore volume of different cathode materials.

Cathode	Surface Area (m ² g ⁻¹)	Pore Volume (cm ³ g ⁻¹)	Reference
aCNS	3167.82	2.49	[9]
AB	51.52	0.21	[9]
KJ	1307.44	3.09	[9,14]
DGr_ac	18.73	0.07	[11]
DGr	13.11	0.05	[11]
KJCO ₂	2386.9	6.53	[14]

On the other hand, the cathode with functional groups that have strong chemisorption to the Cl atom have been employed to construct metal–chlorine batteries with higher performance. Chen and colleagues [12] reported a cathode of Li–Cl₂ battery using metal–organic frameworks (MOFs) with functional group (–NH₂) that has stronger adsorption preference to the Cl atom, UiO-66-NH₂, increasing cycle life to 500 cycles, much higher than those of MOFs without functional groups (UiO-66), as shown in Figure 3a, and the previous studies mentioned above (Figure 2). Sun and colleagues [13], inspired by sponge bionics, developed a bicontinuous N-doped carbon (Bi-NC) cathode. The constructed Na–Cl₂ batteries with the Bi-NC cathode demonstrated an incredible maximum discharge current density of 16,000 mA g⁻¹ (Figure 3b), which was more than two orders of magnitude higher than other reported metal–chlorine batteries (Figure 2). This outstanding performance is attributed to the strong adsorption to Cl₂ brought about by heteroatomic doping. Interestingly, it can also be demonstrated that the re-assembled Na–Cl₂ battery using a disassembled cathode at fully charged state retained most of the discharge capacity (Figure 3c). Also, this sheds some light on the understanding of the state of Cl₂ in the cathode in metal–chlorine batteries. In addition, the bicontinuous-structured carbon also makes an important contribution by facilitating the transport of ions and electrons (Figure 3d).

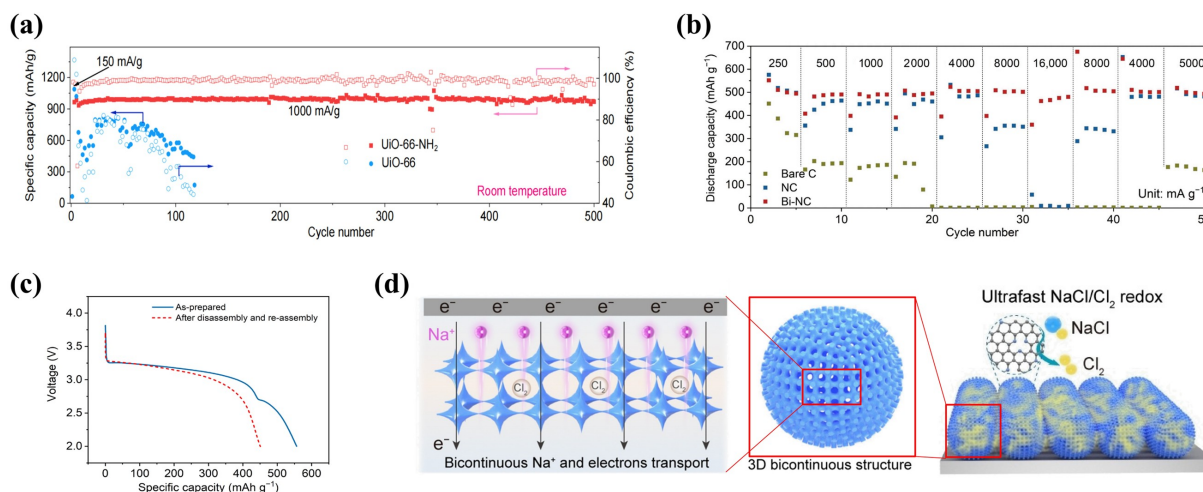


Figure 3. Cathode-induced dramatic performance enhancement of metal–chlorine batteries. (a) Cycling performance of Li-Cl₂ batteries using UiO-66-NH₂ and UiO-66 as cathode [12]. (b) Cycling performance of Na-Cl₂ batteries under different rate capability using bare C, N-doped carbon (NC) and Bi-NC as a cathode [13]. (c) Discharge curves of as-prepared and re-assembled Na-Cl₂ batteries [13]. (d) Microstructure and working mechanism of the Bi-NC cathode [13].

3. The Critical Roles of Electrolytes

The electrolyte is the other key role in making the metal–chlorine batteries come into being. It needs to be noted beforehand that, unlike most batteries, SOCl₂ is both the cathode's active material, participating in the battery reaction as shown in Equation (1), and the electrolyte's solvent in Li-SOCl₂ batteries. The addition of NaFSI and NaTFSI in the solution of AlCl₃ dissolved in SOCl₂, the electrolyte commonly used in Li-SOCl₂ batteries, gave the Na-Cl₂ batteries a decent cycle life owing to the formation of uniform and robust solid electrolyte interface (SEI), as shown in Figure 4a [9]. Similarly, the importance of LiFSI was also reflected in the Li-Cl₂ batteries (Figure 4b) [11]. LiDFOB, additive in SOCl₂ electrolyte, played a key role in enabling the rechargeability of the Ca-Cl₂ batteries by improving the electrochemical stability of the Ca metal anode and facilitating the dissociation and distribution of Cl-based species and Ca²⁺ (Figure 4c) [15].

Besides the additives, the importance of SOCl₂ for metal–chlorine batteries cannot be ignored. Sun and colleagues [13] replaced the SOCl₂ with a commonly used organic solvent (mixture of ethylene carbonate/diethyl carbonate/dimethyl carbonate, 1:1:1 in volume), finding that the Na-Cl₂ battery could not be normally charged, as seen in Figure 4d. It implied that SOCl₂ played an important role in enabling reversible NaCl/Cl₂ redox reaction. On the other hand, thanks to the very low melting point of SOCl₂ (−104.5 °C), the Li-Cl₂ batteries with SOCl₂-based electrolyte have excellent low-temperature performance down to −40 °C and −80 °C, as shown in Figures 2d and 4e, respectively [14]. This offers significant natural advantages over existing lithium-ion batteries [20]. In addition, it is worth mentioning that charging to higher capacity also has to rely on the oxidation of SOCl₂ to form SCl₂/S₂Cl₂, while the SCl₂/S₂Cl₂ contributes a higher voltage plateau at the beginning of discharging (Figure 4f), as shown in Equations (5) and (6), respectively [14]. The entire specific reaction process of the Li-Cl₂ batteries has also been described in detail by Dai and colleagues [14], although it is still proposed that it could provide meaningful help in understanding the intrinsic reaction mechanism of metal–chlorine batteries.



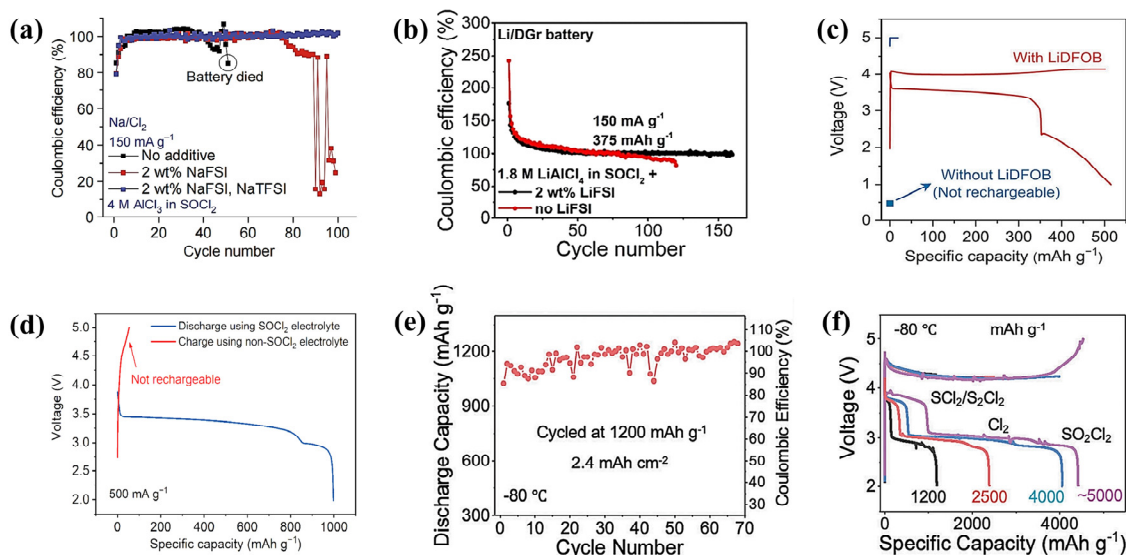


Figure 4. Critical roles of electrolyte on metal–chlorine batteries. (a) Coulombic efficiencies of Na-Cl₂ batteries using the electrolyte of 4 mol L⁻¹ AlCl₃ in SOCl₂ with different additives [9]. (b) Cycling performance of Li-Cl₂ batteries using 1.8 mol L⁻¹ AlCl₃ in SOCl₂ with and without 2 wt.% LiFSI as the electrolyte [11]. (c) Charge–discharge curves of Ca-Cl₂ batteries using 6 mol L⁻¹ AlCl₃ and 1.2 mol L⁻¹ CaCl₂ in SOCl₂ with and without 1.3 mol L⁻¹ LiDFOB as the electrolyte [15]. (d) Discharge and charge curves of Na-Cl₂ batteries using SOCl₂ and non-SOCl₂ electrolyte, respectively [13]. (e) Cycling performance of a Li-Cl₂ battery at -80 °C [14]. (f) Charge–discharge curves of Li-Cl₂ batteries with 1200 to 5000 mAh g⁻¹ cycling capacities [14].

4. Conclusions and Prospects

This perspective has summarized the state-of-the-art progress in metal–chlorine batteries, with a systematic analysis of the critical roles played by cathode and electrolyte in enabling the batteries birth and performance enhancement. The key insights emerging from recent studies are as follows:

- (1) The microstructure of the cathode is highly important for metal–chlorine batteries. It is preferred that the cathode have a larger surface area or pore volume to accommodate the discharge/charge products and provide redox sites. Additionally, the microstructure of the cathode should be favorable for the transport of ions and electrons.
- (2) The functional groups or heteroatomic doping attached to the cathode is also an important approach to improve performance of metal–chlorine batteries due to their strong adsorption to Cl₂.
- (3) The SOCl₂ electrolyte and specific additives in it are necessary to make metal–chlorine batteries possible.
- (4) The SOCl₂ electrolyte can also enable metal–chlorine batteries with ultralow-temperature performance and higher reversible capacity.

Despite recent advancements, metal–chlorine batteries research remains in its nascent stage, with numerous fundamental questions unresolved. Here, we propose the following priority research directions:

- (1) The mechanisms involved in metal–chlorine batteries need to be further clarified. Firstly, except for the main reactions of metal–chlorine batteries, as shown in Equations (2)–(4), other reactions, such as the reactions of the first discharge and the reactions of charging and discharging to a higher capacity, are still proposed and need further clarification. Secondly, the mechanism of the effects of the microstructure of different cathodes on the accommodation of charge/discharge products needs to be further clarified. Thirdly, the working mechanism of the electrolyte, including

SOCl_2 and additives, on the construction of SEI and electrochemical reaction process of metal–chlorine batteries needs to be further clarified.

- (2) Based on new knowledge of the above mechanisms, accordingly, new cathode materials and electrolyte compositions, that have greater ability to accommodate more charge/discharge products and facilitate faster redox reactions, need to be explored further to improve the performance of the metal–chlorine batteries even more.
- (3) Currently, the assembled metal–chlorine batteries are all coin cells except for the pouch cells assembled by Chen and colleagues [12], which is still some way from the requirements of practical applications. In addition, other requirements for the practical use of metal–chlorine batteries, such as safety and economy, need to be investigated.
- (4) The severe challenges faced by metal–chlorine batteries cannot be ignored. Firstly, batteries utilizing lithium metal anodes still encounter the same safety concerns as other lithium metal batteries. Secondly, SOCl_2 , the core component of the electrolyte in metal–chlorine batteries, is highly corrosive, creating safety hazards during both battery manufacturing and operation. Finally, the highly reactive anode and electrolyte employed in metal–chlorine batteries also present significant challenges for battery recycling. To address these challenges, targeted research has been undertaken. For example, Sun and colleagues [16] used Li–Si alloy rather than the lithium metal that conventionally used as the anode for Li– Cl_2 batteries, achieving advances in low cost and high safety. Additionally, non-corrosive organic electrolyte has been introduced into metal–chlorine batteries [10]. However, these measures have resulted in a decline in the performance of metal–chlorine batteries. Consequently, the challenges facing metal–chlorine batteries remain to be addressed through further research.
- (5) More innovative research on metal–chlorine batteries should be carried out. For example, an anode-free metal–chlorine batteries could be developed, where the chlorine-based salt is pre-added in the cathode and then the anode metal is generated in situ during the first charge, to improve safety and reduce costs. Also, solid metal–chlorine batteries could be developed by using a suitable solid electrolyte and possibly adding a very small amount of SOCl_2 in the cathode to enable redox reactions on the cathode.

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