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Commentary

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Toward practical aqueous zinc-ion batteries for electrochemical energy storage

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Zinc-based batteries are recently attracting attention owing to their higher sustainability and affordability compared to their conventional lithium-ion analogues. Identified zinc resources are estimated at more than 1.9 billion tons (at a price of $1.45/lb) based on the United States geological survey in 2022,[1] making Zn at least 20-fold more abundant than Li (89 million tons; $7.71/lb) and 6 times more plentiful than nickel (300 million tons; $8.3/lb). A search with the keyword "zinc batteries" reveals that since 2018, more than 30,700 articles have been published on the subject. Among these, approximately 60% involve aqueous electrolyte zinc-ion batteries (ZIBs), as their inherent safety and potential low cost make them desirable candidates for small- and large-scale stationary grid storage.[2]

Alkaline ZIBs have been well studied[3] and successfully commercialized (for example, Zn-Ni$(OH)_2$ batteries). However, "close-to-neutral" ZIBs (electrolytes with a pH of ~4–7) are at a far more immature level of development despite tremendous effort and growing enthusiasm worldwide. Several major obstacles still plague commercialization of such cells, including (1) poor low-rate performance combined with unrealistically low areal capacities, (2) cathode dissolution and loss of active material during extended cycling, (3) the propensity of Zn to deposit in a range of complex, non-planar morphologies depending upon the current density and electrolyte pH value (non-planar deposits during charge of a Zn anode accelerate capacity degradation by Zn orphaning and short circuits), (4) competition between Zn^{2+} and proton intercalation and possible sluggish Zn-ion diffusion in many cathodes, (5) a narrow electrolyte stability window (especially with respect to anodic stability), (6) a lack of understanding of the pH-dependent Zn^{2+}-ion diffusion mechanism through the solid-electrolyte interface (SEI) *ex/in situ* formed on Zn anodes, and (7) impractically low Zn utilization. Unfortunately, while some of these challenges were known in the early literature, the solutions are not straightforward. This has stymied progress toward practical ZIBs that live-up to the cost and performance promises of the technology and that can be scaled up to meet contemporary stationary electrochemical energy storage needs. It has also unfortunately spawned a growing volume of literature that appears to overlook/obfuscate some of these challenges by focusing on high C-rate but low-capacity performance,

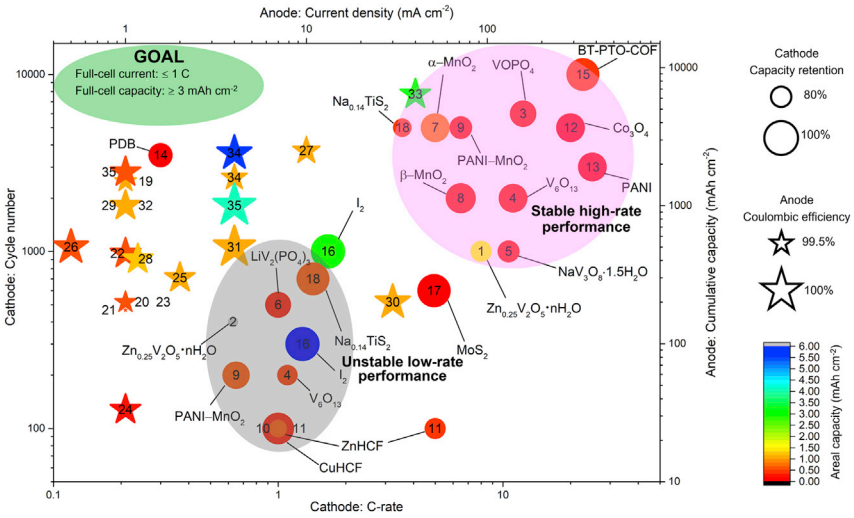


Figure 1. Current evaluation of cathodes and anodes for aqueous mild ZIBs

The published cathode data was analyzed in terms of C-rate (bottom x axis), areal capacity (heatmap), cycle number (left y axis) and capacity retention (diameter of circle). C-rate and areal capacity values are either provided in the literature or calculated based on the reported highest discharge capacity and mass loading of active materials, respectively. Many references are excluded owing to lack of information on mass loading. The published anode data were evaluated based on asymmetric cells in terms of current density (top x axis), areal capacity (heatmap), cumulative capacity (right y axis), and coulombic efficiency (size of star). Cumulative capacity is calculated based on the areal capacity and the reported cycle numbers of Zn plating/stripping. All references are listed in the supplemental information.

which does not fulfill the requirements for practical ZIBs. To bridge the gap, this commentary highlights approaches for addressing these significant challenges to encourage innovation that can accelerate progress toward practical ZIBs.

Unrealistically high C-rates and low areal capacities

In LIBs, the C-rate is a measure of the current at which a battery is discharged relative to its theoretical (or maximum) capacity; a 1C rate means that the discharge current will discharge the battery in 1 h. The same terminology is used in the ZIB field. Figure 1 summarizes the long-term cycling performances of various cathodes at both high rates (>5C) and low rates (0.5C–2C) and Zn anode studies regarding cumulative capacity at different current densities (0.5–40 mA cm^{-2}) based on "reservoir-free" galvanostatic asymmetric-cell protocols (for example, Zn||Cu or Zn||Ti).

In terms of cathode stability, one can easily identify the large gap between low-rate and high-rate cycling performance, including cells based on widely studied manganese oxides, vanadium oxides, and metal disulfides. Most of these cathode materials were reported to show very promising long-term cycling stability at high rates from 5C–30C, but poor cycling performance at low rates from 0.5C–2C. At a similar percentage of capacity retention, the cycle life achieved in these two scenarios can vary by as much as 10 fold. The source of these differences is presently unknown. It may be speculated, for example, that the electrochemical or chemical degradation of cathode active materials is much less severe at high rates, as they spend less time at the potential extremes. Proton (de)intercalation likely dominates at ultra-fast rates, giving rise to "false high Zn^{2+}" capacity. Very fast cycling rates may also contribute to better performance at the Zn anode (see below). Irrespective of the source, however, this means that ZIBs that are unable to undergo stable cycling at the moderate rates used for stationary grid storage (0.25C–2C) are of limited practical interest.[4]

A more serious concern is that often the high C-rate performance is reported in cells with areal capacities less than 0.5 mAh cm^{-2}. These values are not only quite far below typical ones used for practical LIBs (e.g., 3 mAh cm^{-2}), but the apparent performance benefits cannot be extrapolated to higher areal capacity electrodes. Specifically, higher capacities require thick electrodes. This triggers many other challenges, including sluggish mass transport, high voltage polarization, contact loss with current collectors, and crack formation during cycling. Thus, we suggest that follow-up studies of such cells with practical areal capacity and at reasonable rates are warranted.

One exceptional cathode that delivers superior low-rate and high areal capacity performance is the conversion-type iodine (I_2) cathode, which can be cycled at 1.67C (an areal capacity of 3 mAh cm^{-2}) for 1,000 cycles and 1.28C (an areal capacity of 6 mAh cm^{-2}) for 300 cycles with ~100% capacity retention.[5] However, the high corrosivity of iodine introduces new challenges for battery packaging materials design that may compromise the aforementioned cost benefits of the electrodes and electrolytes.

Additional practical considerations

We next consider some other major obstacles that must be addressed for ZIBs to achieve their potential for practical, low-cost stationary energy storage applications (Figure 2).

Dissolution of cathode active materials

Most cathode studies for ZIBs involving manganese-based and vanadium-based materials suffer from the dissolution of active materials, resulting in reduced

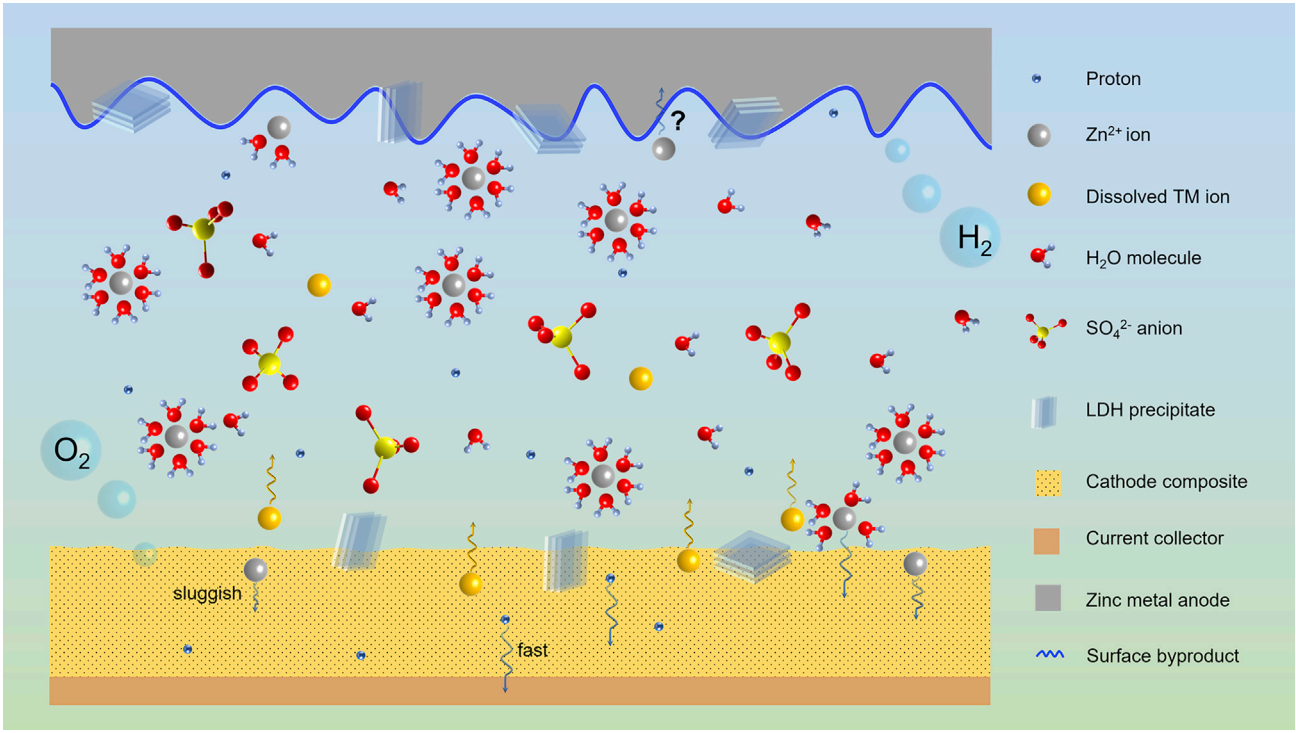


Figure 2. Schematic diagram of a prototypical aqueous ZIB

The commonly reported dilute $ZnSO_4$ electrolyte system is chosen as the platform, illustrating challenges of transition metal (TM) dissolution from the cathode, H_2 and O_2 evolution processes, layered double hydroxide (LDH) precipitation, co-proton intercalation in the cathode, inhomogeneous Zn deposition (dendrite formation), and limitations of thick SEI formation at the anode.

cycle life owing to loss of active material. A promising direction focuses on Mn^{2+} salts as electrolyte additives to suppress dissolution of manganese-based cathodes.[6] This approach has been shown to work at high rates (e.g., 92% capacity retention for 5,000 cycles at 5C) but has not yet been evaluated at the more modest rates demanded in many applications. Possible solutions to suppress cathode dissolution at practical rates involve utilizing aqueous-nonaqueous hybrid electrolytes or "water-in-salt" electrolytes to reduce the water activity or protecting cathode materials with a Zn^{2+} ion conductive coating.

The propensity of Zn to deposit in a range of complex, non-planar morphologies

It is known that in aqueous electrolytes, the Zn anode can undergo parasitic electrochemical reactions with water to form heterogeneous solid electrolyte interphases enriched in species such as zinc layered double hydroxides (LDHs) and zinc hydroxides. A consequence is that Zn deposits in low-density, non-planar mossy morphologies, which may limit battery cycle life by similar processes (e.g., metal orphaning, short circuiting, consuming electrolytes, etc.), like in Li metal batteries. Fortunately, recent progress toward understanding and ameliorating the tendency of Zn to deposit in mossy and dendritic morphologies have been steady, achieving at least 99% coulombic efficiency for Zn plating/stripping at both low and high capacities or current densities (0.5–6 mAh cm^{-2}, 0.5–40 mA cm^{-2}) to date (Figure 1).

In weakly acidic or neutral electrolytes, synergistic regulation of the crystallography and electrodeposition reversibility of Zn has been reported in a variety of contexts. Approaches ranging from heteroepitiaxial deposition of Zn on low lattice-mismatched interfaces composed of textured graphene,[7] or dynamic formation by adsorption of anisotropic graphitic carbon nitride (g-C_3N_4) nanostructures dispersed in liquid electrolytes,[8] or via homoepitaxial growth from Zn anodes produced by accumulative roll bonding[9] have all been shown to be effective in enhancing Zn reversibility at practical capacities and moderate current densities. Moreover, interphase design can also extend the cycling life of Zn anodes at high areal capacity and moderate current density.[10]

Considering that the most mature cathodes for ZIBs are based on aqueous electrolytes, where SEI formation can pose significant challenges for maintaining crystallographic control at high deposition capacities, we highlight the specific need for Zn anode, SEI, and electrolyte designs that facilitate high-areal capacity ZIBs reversibility. Applying a high current usually induces

denser Zn deposition and less Zn dendrite formation in conventional dilute electrolytes, thus substantially enhancing the cycle life of Zn anodes.[11] Therefore, it is critical to screen Zn reversibility with a practical areal capacity at both low and moderate current densities for a particular system to fully investigate its viability.

Zn^{2+} versus proton intercalation and the precipitation of LDHs

Compared with organic solvents in nonaqueous electrolytes, water has the merit of high ionic conductivity (1–100 mS cm^{-1}), inflammability, low interfacial resistance, and high salt solubility. Nonetheless, water also causes many undesired side reactions, including proton co-intercalation and the simultaneous precipitation of LDHs (for example, $Zn_4SO_4(OH)_6 \cdot xH_2O$) on both inorganic[12] and organic[13] cathode interfaces on discharge. Proton intercalation is also considered as the origin of ultra-high-rate capability of many cathode materials, as Zn^{2+} diffusion in intercalation hosts may be sluggish.[14] The insulating LDHs also increase interfacial resistance and gradually detach from the cathode surface leading to short cycle life (especially at lower rates). A strategy of tuning the solvation structure of Zn^{2+} results in significantly suppressed proton intercalation;[14] nonetheless, understanding the interplay of Zn^{2+} versus proton intercalation in specific cathodes and discovering new inorganic or organic cathode materials with excellent Zn ion diffusivity and dominant Zn^{2+} (de)intercalation properties are still necessary.

Challenges ascribed to narrow electrolyte stability window

The thermodynamic electrochemical stability window of water is 1.23 V and depends on electrolyte pH. The oxygen evolution reaction (OER) and hydrogen evolution reaction (HER) occur at higher and lower voltages, respectively due to overpotentials. These reactions decompose the aqueous electrolyte. The risk of OER limits the choice of cathode materials due to the low oxidative stability of mildly acidic electrolytes (usually <2 V at galvanostatic conditions). OER also enhances the electrolyte acidity due to water decomposition, further inducing the degradation of some cathode materials. Thus, new fundamental approaches are required to enhance the anodic stability of aqueous Zn electrolytes and to identify suitable high-voltage cathode materials. The existence of HER during Zn deposition also triggers a local alkaline environment, inducing the formation of LDHs and $Zn(OH)_2$ on the Zn surface. These surface byproducts lead to low coulombic efficiency and a rough Zn surface, aggravating dendrite formation and decreasing cycle life. Recently, much effort has been devoted to suppressing HER and achieving highly reversible Zn anodes. Building *in* or *ex situ* solid electrolyte interfaces (SEIs)—such as those involving ZnF_2[15]—appears to be an effective strategy; nonetheless, the mechanism of Zn ion diffusion through such interphases remains unclear and needs follow-up scrutiny.

Impractical low utilization of Zn anodes

The often low areal capacity (<0.5 mAh cm^{-2}) of cathode materials reported in ZIBs highlights the low utilization of Zn anodes in these studies. As the theoretical areal capacity of commonly used Zn foil with a thickness of 100 μm is ~58.5 mAh cm^{-2}, the Zn utilization is less than 1%. Thus, nearly 100% coulombic efficiency and very promising specific energy density based only on cathode loading was achieved in most such studies. However, these conditions are not applicable to practical stationary batteries. In order to evaluate the cycling performance of ZIBs in a more realistic way, it is important to report both specific and volumetric energy density with increased Zn utilization or controlled capacity ratios between negative and positive electrodes (N/P ratios).[8] The following equation gives a plausible estimation of energy densities of ZIBs based on both cathode and anode volumes ($E_{cathode+anode}$) when considering a reasonable N/P ratio:

$$E_{cathode+anode} = \frac{Q_{Zn} \times Q_{cathode}}{Q_{Zn} + Q_{cathode} \times \left(\frac{N}{P}\right)} \times \overline{V} \quad \text{(Equation 1)}$$

where the Q_{Zn} and $Q_{cathode}$ are the theoretical specific or volumetric capacity of Zn and cathode materials, respectively. $\overline{V}$ is the average voltage output.

According to Equation 1, the critical impact of N/P ratios on both energy densities can be easily seen in Figure 3. Considering the same cathode (Figure 3A), its specific energy density at an N/P ratio of one (corresponding to 100% Zn utilization) is nearly twice than that of an N/P ratio of four (corresponding to 25% Zn utilization). Using a more rigorous protocol with an N/P ratio of zero—a called anode-less cell configuration—the energy density is around 450 Wh kg^{-1} based on a typical α-MnO_2 cathode[6] (specific capacity of ~300 mAh g^{-1}, average voltage of ~1.5 V). In terms of volumetric energy densities, taking $Zn_{0.25}V_2O_5 \cdot nH_2O$ (n = 1)[16] as an example (Figure 3B), its energy density can be as high as 760 Wh L^{-1} in an anode-free cell. Therefore, screening the intermediate-rate cycle life of a cathode material at reasonable N/P ratios (lower than 3) to fully evaluate its strengths and limitations is important. However, we note that the difficulty of fabricating thick cathodes also needs to be addressed, as the mass loading of typical oxide cathodes (α-MnO_2 or $Zn_{0.25}V_2O_5 \cdot nH_2O$) can be as high as 10 mg cm^{-2} for a target of 3 mAh cm^{-2}. (Figure 3A). The current lab-level mass loading of these materials is usually lower than 2 mg cm^{-2}, far less than practical values. Therefore, more future work should be focused on developing thick electrodes with excellent Zn^{2+} ion diffusion properties or

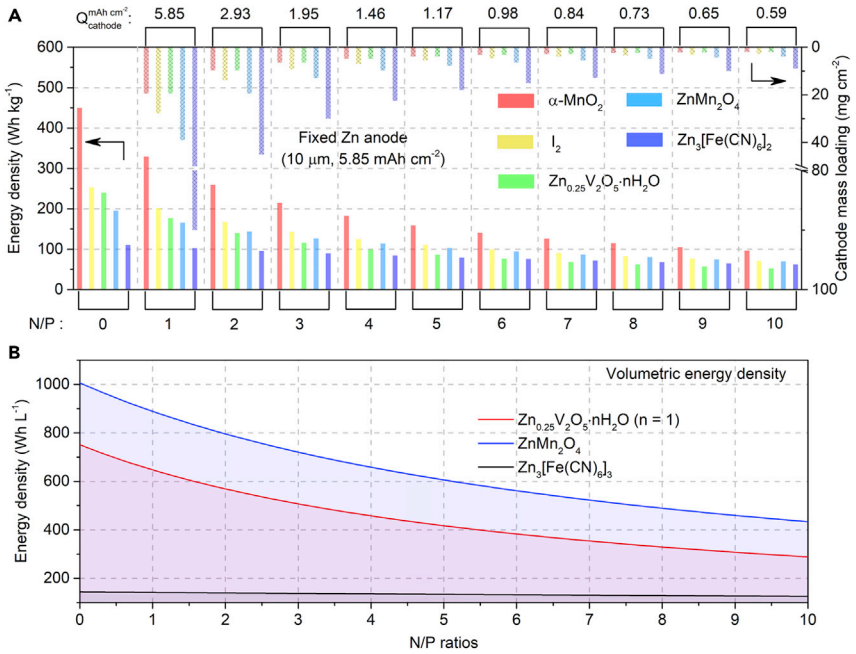


Figure 3. Energy densities of ZIBs

(A) Specific energy density (left y axis) as a function of N/P ratios (bottom x axis). These values are calculated based on Equation 1 with different N/P ratios (0–10), using five widely studied cathode materials: α-MnO_2 ($Q_{cathode}$ = 300 mAh g^{-1}, $\overline{V}$ = 1.5 V), I_2 (211 mAh g^{-1}, 1.2 V), $Zn_{0.25}V_2O_5$•nH_2O (300 mAh g^{-1}, 0.8 V), $ZnMn_2O_4$ (150 mAh g^{-1}, 1.3 V), and $Zn_3[Fe(CN)_6]_2$ (65 mAh g^{-1}, 1.7 V). For better comparison, the Zn anode was fixed as a practical metal foil with a thickness of 10 μm and an areal capacity of 5.85 mAh cm^{-2}. The required cathode areal capacity and mass loading at different N/P ratios was further calculated for reference (top x and right y axis).

(B) Volumetric energy density as a function of N/P ratios. Calculations are based on Equation 1, using three intercalative-type cathodes: $Zn_{0.25}V_2O_5$•nH_2O (n = 1, the theoretical volumetric capacity is 939 Ah L^{-1} based on the unit cell density of 3.13 g cm^{-3}), $ZnMn_2O_4$ (774 Ah L^{-1}, 5.16 g cm^{-3}), and $Zn_3[Fe(CN)_6]_2$ (84.5 Ah L^{-1}, 1.3 g cm^{-3}). It is difficult to estimate the volumetric energy density of conversion-type α-MnO_2 and I_2 because their volume change during cycling is highly dependent on the depth of discharge and many other electrochemical parameters.

developing new cathode materials with higher specific capacities to satisfy practical requirements.

Conclusions

Regarding ZIBs as a promising candidate for large-scale stationary grid storage, battery scientists have dedicated enormous efforts and performed countless studies in this field, resulting in many publications. Through a detailed analysis of achievements and improvements in ZIBs, however, many challenges underlying these claims of outstanding electrochemical performance are overlooked or underestimated. While the current work on ZIBs is still at an early stage of development, it would benefit from more realistic and reasonable evaluation. This includes screening the cycling stability of cathode materials at moderate rates (≤1C) and practical areal capacities (≥3 mAh cm^{-2}), assessing the reversibility and stability of Zn plating/stripping for a particular system at both low and moderate current densities with an areal capacity of at least 3 mAh cm^{-2}, and examining full cells at practical N/P ratios (<3) to enhance Zn utilization. More studies are also required to focus on other issues, including cathode dissolution (especially at practical rates), the interplay of Zn^{2+} versus proton intercalation, sluggish Zn^{2+}-diffusion in the cathode, widening the electrolyte stability window using low-cost current collectors, and understanding the Zn ion diffusion mechanism through SEIs on the Zn anode. We hope our perspective provided in this commentary can encourage steps toward narrowing the gap between the current status of ZIBs and future demands of practical stationary grid storage.

SUPPLEMENTAL INFORMATION

Supplemental information can be found online at https://doi.org/10.1016/j.joule.2022.06.002.

ACKNOWLEDGMENTS

This work was financially supported by the Joint Center for Energy Storage Research, an Energy Innovation Hub funded by the US Department of Energy, Office of Science, Basic Energy Sciences. This work was also supported as part of the Center for Mesoscale Transport Properties, an Energy Frontier Research Center supported by the US Department of Energy, Office of Science, Basic Energy Sciences, under award #DE-SC0012673. L.F.N also acknowledges NSERC for platform support through the Discovery Grant and Canada Research Chair programs.

DECLARATION OF INTERESTS

The authors declare no competing interests.

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https://doi.org/10.1016/j.joule.2022.06.002

Commentary

Surging lithium price will not impede the electric vehicle boom

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Lithium-ion batteries (LIBs) play a key role in the energy transition as the primary energy storage device in mobility and renewable energy systems.[1] Of the diverse materials that comprise a LIB, many—such as lithium, cobalt, and nickel—are considered "critical" due to their high supply risk and importance to product performance. A low-carbon future, therefore, depends considerably on their supply stability.

Contrary to anticipation, the global LIB supply chain is currently haunted by market fluctuations. From December 2020 to April 2022, the Chinese spot market has seen a price increase by 830% for lithium