Between Promise and Practice: A Comparative Look at the Energy Density of Li Metal-Free Batteries and Li Metal Batteries

Due to the increasing dependence on renewable energy and electric transportation worldwide, lithium metal batteries (LMBs) have become one of the hottest topics in the battery field.¹ Li metalfree batteries (LMFBs) are characterized by the avoidance of preloaded Li metal or host materials such as graphite on the anode.^{2, 3} The terminology "anode-free" is commonly used to signify that a metal battery lacks any metal source on the negative electrode side in the initial state. However, strictly speaking, "anode-free" is somewhat misleading, because anode is essential for electrochemical systems. Therefore, recently the use of terminologies such as "Li metal-free", "Lifree", "excess Li metal-free" or "anode-less" has been suggested more accurately to convey the absence of Li metal in the anode.⁴ In the LMFBs, the Li sources from the cathode material are plated onto the current collector during charging and then stripped during discharging.⁵ As a result, LMFBs can achieve high energy densities, raising the energy density of the batteries to their theoretical limit while simultaneously reducing manufacturing costs.^{2, 6-8} Nevertheless, the progression of LMFB faces significant challenges. The actual energy density and cycling performance of LMFB are highly restricted by the loss of Li inventory during the initial charge and the limited reversibility of Li plating/stripping during battery cycling.⁹ On the other hand, lean lithium metal batteries (LLMBs) that employ a minimal amount of Li metal on the anode are one of the strong countermeasures to this problem. This viewpoint describes the benefits of LLMBs in contrast to LMFBs by comparing the differences in their theoretical and practical energy density and cycleability.

The schematic drawing of LMFB is shown in **Figure 1**. This LMFB exhibits theoretical gravimetric and volumetric energy densities (GED and VED) of 472 Wh kg⁻¹ and 1410 Wh L⁻¹, respectively, when using the LiNi_{0.8}Co_{0.1}Mn_{0.1}O₂ (NCM811) cathode material (capacity loading of 5.3 mAh cm⁻², see **Table S1** for the cell properties and calculations). Given that the anode

contains no active material (N/P ratio = 0, where the N/P ratio is defined as the ratio of the capacity of the negative active material to that of the positive active material at the fully discharge state), it does not contribute to the gravimetric or volumetric energy density, allowing for the theoretically highest energy density for LMBs.

For many studies, LMBs embodied with the N/P ratio > 1 (**Figure S1**) are generally used in mathematical calculations or visual schematics to highlight the advantages of LMFB. In these instances, the GED of the LMB reduces to < 457 Wh kg^{-1,} and VED substantially declines to < 1,190 Wh L⁻¹. However, this comparison certainly underestimates the prospects of LMBs. The N/P ratio can practically be further decreased when configured as LLMB (**Figure 1b**). For instance, with a N/P ratio of 0.2, the GED and VED minorly reduce to 469 Wh kg⁻¹ and 1360 Wh L⁻¹, respectively. Considering other indispensable cell components, a minor increment in Li metal accounts for a mere 0.66% and 3.58% in mass and volume, respectively (note that the theoretical capacity of Li metal is 3,860 mAh g⁻¹). Moreover, when auxiliary components of a battery pack, like interconnectors, are considered, the proportion attributed to Li metal further decreases. Although the preparation of thin Li metal is quite challenging, recently, thin Li metal foil with a thickness of 20 µm has been commercially available,¹⁰ and the preparation of ultra-thin Li metal has also been vigorously studied.^{11, 12}

Undoubtedly, merely having similar energy density is insufficient to decisively justify the acceptance of LLMB. The main objective of this viewpoint is to evaluate how the addition of a minimal Li metal anode affects batteries' performance such as energy density and lifetime. During the first charge of LMFB, Li plates onto the anode current collector. Nevertheless, the use of prevalent current collectors such as Cu foil can result in an irregular Li deposition morphology, which stems from overcoming significant nucleation barriers due to the lithiophobicity of Cu foil.¹³

Such behavior leads to the growth of Li dendrites, which, in turn, reduces the reversibility of Li plating/stripping cycle.^{14, 15} Moreover, highly reductive Li metal causes the electrolyte decomposition on its surface, creating solid electrolyte interphase (SEI), which irreversibly consumes Li inventory. In particular, when Li is first deposited on the Cu surface, normally extra SEIs are formed.¹⁶ Therefore, the initial Coulombic efficiency (CE₁) of Li plating/stripping is less than CE_n ($n \ge 2$, where *n* is the cycle number). Concurrently, throughout the cell's life cycle, the electrolyte persistently undergoes side reactions and the formation of "dendrite and dead Li", leading to lower CE_n ($n \ge 2$).¹⁷

The capacity of LMB (Q_n) after the *n*th cycle (*n*) can be estimated by **Equation** (1) with an assumption that degradation of the cathode is minuscule in comparison to the Li loss at the anode during the subsequent cycles (where $n \ge 2$) that incorporates two factors, the initial discharge capacity ($Q_{initial}$) and the CE of Li plating/stripping:²

$$Q_n = Q_{initial} \prod_{i=2}^n CE_i \tag{1}$$

It should be noted that certain studies have assessed Q_n for both LMFB and LMB using the same value of $Q_{initial}$. However, due to the absence of a supplementary Li source in LMFB, irreversible Li depletion remains uncompensated.^{18, 19} During the 1st discharge, a limited fraction of Li can revert to the cathode, resulting in an observable initial capacity loss ($Q_{A,initial}$), substantially deviating from the ideal capacity of the cathode material.² On the other hand, LLMB has a compensatory release of Li from the excessive Li reservoir at the anode which minimizes $Q_{A,initial}$.¹⁷ Thus, the same $Q_{initial}$ value for these two different cases should not be adopted for comparison of LMBs. According to one example, $Q_{initial}$ in an LMFB using NCM811 is approximately ~170 mAh g⁻¹ due to the high $Q_{A,initial}$, lagging behind that combined with Li metal anode(a typical value of

~200 mAh g⁻¹),^{20, 21} which is attributed to the initial Li loss. The greater $Q_{\Delta,initial}$ of the LMFB than that of the LMB with excess Li anode under similar conditions is also seen regardless of cathode materials.²² Therefore, when factoring in $Q_{\Delta,initial}$, the GED and VED for practical LMFBs are likely to show a lower value than their potentials.

An in-depth assessment was carried out to discern both the theoretical and actual energy densities of LMFBs and LLMBs along with their lifespan. **Figure 2a,b** illustrates embodiments of LMFBs that can theoretically achieve the utmost GED, but a mere 5% ($Q_{A,initial}/Q_{ideal} = 0.05$) consideration of the initial $Q_{A,initial}$, resulting in a decrease of the GED and VED for LMFB to 449 Wh kg⁻¹ and 1339 Wh L⁻¹, respectively. These values are surprisingly lower than those of the LLMB with a N/P ratio of 0.2 (469 Wh kg⁻¹ and 1360 Wh L⁻¹). Consequently, the LMFB's competitive edge over the LLMB diminishes in this scenario.

As CE_n (where $n \ge 2$) represents in **Equation (1)**, LLMBs display an advantage on lifetime over LMFBs, benefiting from its auxiliary Li source, retaining an elevated initial CE.¹⁶ Its rate of degradation parallels that of the half cell, remaining relatively steady, and is much lower than that of LMFB under similar conditions (**Figure 2c**).¹⁷ Only upon Li depletion, LLMBs undergo a genuine shift, mimicking the LMFB,⁹ and subsequently enter a swift degradation phase stemming from non-reversible Li losses. As shown in **Figure 2c**, when an excess of Li metal remains in the anode, the CE of LLMB derives exclusively from the cathode. Considering *Q*_{A,initial}, the GED of LMFB faces an initial 5% capacity reduction and ongoing Li loss. With an average CE reaching 99.9%, its energy density dwindles to 58% after 500 cycles. Conversely, in LLMB systems, minimal Li presence leads to a slight decrease in GED (0.66 wt% contribution, **Figure 1b**) initially but significantly bolsters battery performance. The extra Li ensures that the *Q*_{initial} closely mirrors *Q*_{ideal} and LLMBs display a unique "energy density plateau" due to its high initial CE, slowing down battery degradation rates. Note that this unique energy density plateau can be extended by adjusting the N/P to compensate for energy density.

Here, the above assessment was experimentally demonstrated. Given the factors previously discussed, Cu/Li and lean Li/Li cells are constructed to examine the performance of the Li metalfree negative electrode and a lean Li metal negative electrode with 20% capacity excess (Li predeposition of 0.3 mAh cm⁻²), employing the widely recognized 4 M Li[FSA] in DME (4 M lithium bis(fluorosulfonyl)amide in 1,2-dimethoxyethane) electrolyte.²³ **Figure 3a** and **b** show the galvanostatic test curves of the two half-cells during Li metal plating/stripping cycles. The Li stripping capacity is significantly lower than the plating capacity in Cu/Li, while lean Li/Li behaves more closely to conventional excess Li/Li cells. **Figure 3b and S2** vividly illustrate that the CE of the Cu/Li cell for the first cycle stands merely at 93.5% due to the initial SEI formation coupled with irreversible Li depletions.²³ From the cycles onwards, CE witnesses a steady ascent, exceeding 98.73% at the 10th cycle, aligning with reported values for this specific electrolyte system.^{23, 24} Conversely, the lean Li/Li cell exhibits the initial CE of 100.0% owing to the predeposition of excess Li which acts as an inventory against Li loss and CE consistently remains at 100.0% over the span of 10 cycles (Figure 3b and S2).

Furthermore, both LMFB and LLMB (N/P ratio of 0.2) full cells were tested to discern their actual energy densities, along with their lifespan. These tests utilized the consistent electrolyte and NCM811 cathode, justified by the fact that solvent molecules in concentrated Li[FSA]-ether electrolytes render themselves compatible with the NCM cathode.²⁵ **Figure 3c,d** presents the charge-discharge profiles of the LMFB and LLMB. While both of them display similar behavior as reported by literature,²⁴ LLMB distinctively exhibits higher capacity retention (**Figure 3d**). As further delineated in **Figure 3c**, LMFB started with a $Q_{initial}$ of 190.2 mAh g⁻¹, indicating a $Q_{A,initial}$

of 6.8 mAh g^{-1} . The 2nd cycle hinted at a rise of discharge capacity, possibly stemming from incomplete lithiation of the NCM due to the restricted depth of discharge as well as the slow kinetics of NCM at high Li content and the formation of an overlithiated Li₂MO₂ surface phase during the initial cycle,^{26, 27} leaving behind the residual Li to offset the losses. Nevertheless, the following cycles for LMFB portrayed the capacity dropping rapidly to 79.1% retention in the 20th cycle. In stark contrast, LLMB exhibited a $Q_{initial}$ of 193.4 mAh g-(cathode + anode)⁻¹ closely mirroring that of Li-rich LMB. In the cycles that followed, there was a marginal elevation in Q_n to > 195.0 mAh g-(cathode + anode)⁻¹, and this level persisted before waning noticeably after the 20th cycle, considerably greater than that of LMFB (Figure 3e). Excluding the initial inherent irreversible CE attributed to the NCM811 electrode, LLMB sustains an impressive CE above 99.4% while LMFB registers an average CE of 98.3% over 20 cycles. The CE of the full cells also follows the trends noted in the prior scenarios of Cu/Li and lean Li/Li cells. Recognizing that the 20% capacity excess of Li affects the overall specific energy density based on the mass of cathode and anode materials. Figure 3f further shows a comparative cycle performance of energy densities. The irrecoverable $Q_{\Delta, initial}$ for the LMFB diminishes its energy density advancement against the LLMB. Such superior performance of LLMBs compared to LMFBs has also been reported in previous studies (**Table S2** provides a comparative summary across various studies. In each certain study, LLMBs and LMFBs were investigated under analogous conditions).^{10, 20, 22, 28-30}

These experimental findings bolster the theoretical discussions in the former part of this viewpoint; the actual LMFB, when integrated with $Q_{A,initial}$, falls behind the LLMB in terms of energy density. Furthermore, the presence of an auxiliary Li source significantly prolongs the lifetime of the LLMB. An advanced electrolyte with higher CE might further decelerate pre-

deposited Li consumption and degrade more tenderly,⁹ which might accentuate LLMB's practical superiority over LMFB.

To summarize, LMFBs are often touted for their superior energy density, but their practical applications imply certain limitations due to irreversible losses of Li during the initial and cyclic stages. Thin Li layer and deposited Li are expected to exhibit different morphologies and interfacial properties, influencing the formation of the SEI and subsequently impacting the performance of LMBs. This aspect needs further investigation to optimize LMBs. Yet, integrating lean Li at the anode can notably bolster the capacity and life cycle of LMBs, even if it slightly compromises its energy density, making it a pragmatic choice under lenient technical prerequisites. It is important to note that this discussion does not endorse one over the other; LMFB and LLMB inherently share numerous similarities and overlapping technologies, such as the benefits gleaned from advanced electrolytes and protocol management. In addition, the manufacturing advantage of LMFB—bypassing the need for stringent measures to handle the fragile Li metal—affords it distinct benefits. Moreover, the performance metrics derived from LLMBs can be invaluable in guiding future LMFB evaluations. Some LMFB enhancements, like crafting artificial SEI layers or employing 3D current collectors, might inadvertently augment battery weight and size, negating the foundational purpose of LMFB. In such scenarios, the energy density and battery performance of LLMB, under comparable conditions, can serve as a benchmark to assess the tangible impact of such interventions on LMFBs, aiming to genuinely elevate battery efficacy.



Figure 1. Schematic illustrations of (a) LMFB (N/P ratio of 0) and (b) LLMB (N/P ratio of 0.2) using an NCM811 cathode of 100 μ m thickness and 5.33 mAh cm⁻² capacity loading, with scaled volume (thickness). The pie graph shows the corresponding mass distribution. The parameters for the cells are listed in **Table S1**.



Figure 2. Calculated relative (a) GED and (b) VED based on the embodiments of ideal LMFB, practical LMFB (here the GED and VED were calculated based on the assumption of $Q_{\Delta,initial}/Q_{ideal}$ = 0.05) and practical LLMB (N/P = 0.2). (c) The relative GED retention with cycling under the same conditions as (a).



Figure 3. The galvanostatic Li metal plating/stripping on (a) Cu/Li cell and (b) 20% capacity excess lean Li/Cu cell (0.3 mAh cm⁻² capacity of pre-deposited Li). (c) LMFB (N/P ratio of 0) and (d) LLMB (N/P ratio of 0.2) using NCM811 as the cathode electrode. Capacity is based on the mass of NCM811 for LMFB and the total mass of NCM811 and pre-loaded Li. (e,f) Cycleability plot of discharge capacity, Coulombic efficiency, and energy density corresponding to (c) and (d). The energy density is based on the mass of anode and cathode active materials.

ASSOCIATED CONTENT

Supporting Information.

The Supporting Information is available free of charge at DOI: xx.

Experimental section, detailed calculations, electrochemical results (PDF)

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Notes

Views expressed in this Viewpoint are those of the authors and not necessarily the views of the

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