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Future Energy

All-climate battery energy storage

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We examine the latest developments in all-climate batteries (ACBs) that enable efficient and resilient energy storage across extreme temperature ranges, e.g., from −50°C to +60°C. A figure of merit is presented to quantify where the current state of art, the latest advances and the future targets stand in this rapidly evolving field. We review two distinctive approaches driving power and stability improvements in both low- and high-temperature environments: materials innovation (particularly electrolyte formulations) and thermal actuation. It is found that there are still two-orders-of-magnitude gaps from the ACB target of high-temperature stability by materials innovation alone and that the material-thermal synergetic approach promises to attain the dual goals of ACBs for uncompromised power and stability at both low and high temperatures. Future research should be focused on developing heat-tolerant electrolytes and electrodes that can survive in 70°C–85°C environments.

INTRODUCTION

Electrochemical energy-storage cells that function with invariable performance and reliability over a wide temperature range, e.g., from −50°C to 60°C, are called all-climate batteries (ACBs). The acute need for ACBs in transport decarbonization and grid-scale energy storage epitomizes a major deficiency of the lithium-ion battery (LIB) technology originally commercialized for man-portable electronics. Since the term's inception in 2016,[1] ACB has rapidly evolved, evidenced by hundreds of studies and reviews.[2,3] ACB is also termed "wide-temperature batteries" in literature.

Successful development of ACBs requires overcoming LIB drawbacks at both low and high temperatures simultaneously. At low temperatures, challenges include low power and little usable energy from poor electrolyte conductivity, sluggish de-solvation/charge transfer, and slow lithium diffusion in electrode materials. At high temperatures, issues include volatile and flammable solvents such as linear carbonates causing hazards, accelerated growth of the solid-electrolyte interphase (SEI) consuming cyclable lithium and shortening lifespan, and severe oxide cathode degradation via cation mixing, phase transitions, and microcracking. Balancing these conflicting requirements at both temperature extremes appears exceedingly difficult, if possible at all, via material solutions.

Here, we construct quantitative metrics of ACBs depicted in Figure 1A. The figure uses the dischargeable energy density at C/10–C/3 rate and −50°C as the y axis to measure low-temperature performance and storage/cycling time at +60°C as the x axis to measure high-temperature stability. Taking currently available room temperature metrics and applying a 10% discount to both limits, the targets of ACBs can be numerically defined by energy densities of 260 and 180 Wh/kg and storage/cycling time of 12,000 and 18,000 h for NMC811 and LFP systems. These are shown in Figure 1B, marking the successful attainment of nearly invariable power and stability over the wide temperature range from −50°C to +60°C, as implied by the term ACB. Also, Figure 1B displays state-of-the-art (SOA) single-cell values. Gigantic gaps between SOA and ACB targets signal a strong need for focused research. Below, we discuss two principal approaches: materials innovation and thermal actuation (TA).

Materials innovation

Materials innovation, especially electrolyte re-formulations, has been sought to broaden the operational temperature range of LIBs. Toward low temperatures, a popular method to boost electrolyte conductivity is lowering viscosity with highly volatile and hence flammable solvents. Examples are ethyl methyl carbonate (EMC), dimethyl carbonate (DMC), and dimethyl ether (DME), as well as methyl acetate (MA) and methyl formate (MF), with their boiling points at 107°C, 90°C, 85°C, 57°C, and 32°C, respectively, and their flash points at 27°C, 17°C, −2°C, −13°C, and −33°C. Unfortunately, these volatile solvents are non-starters for practical use in surging air temperatures. Weakly solvated and small-molecule electrolytes can enhance de-solvation and hence charge-transfer kinetics at low temperatures.[4,5] Finally, particle size reduction is commonly proposed to not only accelerate lithium diffusion in electrode materials but also to provide a large Brunauer-Emmett-Teller (BET) area for improved charge-transfer kinetics. However, such a strategy leads to excessive reactivity of electrolyte-electrode interfaces at high temperatures, thereby compromising battery lifespan and safety.

At the high temperature end of an ACB, materials innovation has resorted primarily to thermally stable electrolytes such as LiFSI salt in dicarbonate solvents[6] and gel electrolytes,[7] solvents with low reactivity,[7] and robust interphase layers e.g., through electrolyte additives such as triallylphosphate (TAP).[8] Unfortunately, all these strategies simultaneously diminish ion transport and reactivity at low temperatures. For instance, the dicarbonate solvent proposed by the Dahn group,[6] dimethyl-2,5-dioxahexane carboxylate (DMOHC), has a boiling point of 220°C and a flash point of 82°C, yielding an outstanding 1-year lifespan at 85°C. However, DMOHC has high viscosity and only 0.3 mS/cm ionic

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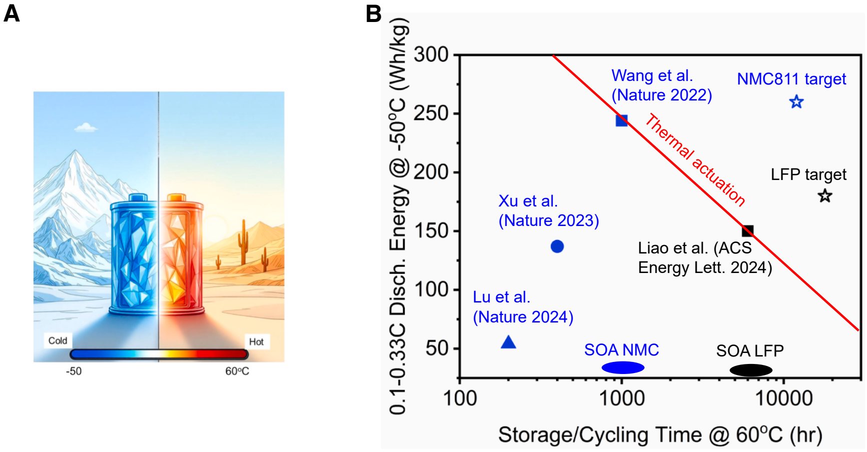

Figure 1. Figure of merit for all-climate batteries)

(A) All-climate battery (ACB) schematic.

(B) ACB figure of merit measured by dischargeable energy density at C/10–C/3 rates and −50°C and storage/cycling time at 60°C, where the targets are represented by 10% discount from the room temperature values of today's cells for NMC811/Gr (blue color) and LFP/Gr (black color) systems, respectively. State of the art (SOA) values are actual measurements from single cells over the −50 to 60°C range. Two datapoints on the left side are the best ACB examples via materials innovation. Two datapoints connected by a red line represent the best ACB results via thermal actuation for NMC811 and LFP chemistry, respectively.

conductivity at 20°C vs. ~14 mS/cm of the standard LIB electrolytes. Ge et al.[8] demonstrated 7,500 h of operation at 60°C for NMC811/Gr cells by adding a small amount of TAP. However, the cells' power capability at room temperature drops by 30%. If these high-temperature electrolytes are to be deployed in ACBs, more work is needed on (1) how to lower the electrolytes' activation energy such that low-temperature properties don't drop sharply and (2) how to strengthen their high-temperature stability by scrutinizing degradation and safety issues such as DMOHC decomposition pathways, long-term efficacy of additives (e.g., TAP) at >80°C, degradation of LiFSI salt in gel electrolytes under thermal stress, temperature thresholds for thermal runaway, etc.

Electrode-engineering approaches to enhance stability and cyclability at high temperatures include the use of active materials of low BET area[9] and single-crystal layered oxides.[10] Effectiveness of the low-BET electrodes is predicated on the square root relationship between capacity decay and storage/operational time for cells whose aging is controlled by SEI layer growth, i.e.,

$$\frac{\Delta Q}{Q_o} \sim BET * t^{0.5} \qquad \text{(Equation 1)}$$

This is the case for LFP/Gr cells where the LFP cathode is thermally stable and cell aging is dominated by the SEI growth in the graphite anode. At the same 80% capacity retention, the lifespan can be extended by a factor of four if the BET is halved, say, reducing from 2 m^2/g to 1 m^2/g of artificial graphite.[9] Other methods to enhance thermal stability at high temperatures include lowering the upper charge voltage to 4 V[6] and using no-strain insertion materials.

Two of the best examples of ACBs attainable via electrolyte innovation were reported by Xu et al.[4] and Lu et al.[5] as displayed in Figure 1B. Clearly, the low-temperature power and usable energy are markedly improved; however, their high-temperature stability and cyclability are still far inferior to the ACB target with two-orders-of-magnitude gaps in storage/cycling time. Moreover, these best examples of ACBs by materials innovation alone tend to move away from, instead of toward, the targets for high-temperature stability/cyclability. There is hope for all-solid-state battery technology to resolve the conflict between low-temperature activity and high-temperature stability; however, actual data that can directly be added to Figure 1B is yet to emerge.

Thermal actuation

Another major approach to developing ACBs is intracell TA of Wang et al.[1] To date, two notable TA examples[11,12] have been provided for NMC811 and LFP cells, respectively, as summarized in Figure 1B. Evidently, these TA-enabled cells are much closer to ACB targets than materials innovation counterparts, although a 3×–10× gap remains in terms of storage/cycling time at high temperatures.

Basic principles of battery TA are rooted in the cell's heat capacity and energy density. The former is roughly 900 J/kg·K or 0.25 Wh/kg·K. If 250 Wh/kg is assumed as the average energy density of current LIBs and 500 Wh/kg for next-generation batteries, then it takes 0.1% state of charge (SOC) to heat 1°C of LIB cells and 0.05% SOC for next-gen batteries. For simplicity, let's take 0.075% SOC as the average heating energy needed to raise 1°C or 0.75% SOC for 10°C. To discharge 0.75% SOC, it takes 27, 9, and 4.5 s at 1C, 3C, and 6C rates, respectively. These yield 22°C/min, 66°C/min, and 133°C/min for intracell TA as opposed to ~1°C/min for external heating currently practiced. Thus, TA is essentially an ultrafast and energy-efficient transformation of battery states, e.g., transforming state 1 (100% SOC, −30°C) to state 2 (96.25% SOC, 25°C) in 30–120 s.

Tremendous benefits of TA are shown in Figure 2A, which plots electrolyte conductivity and charge-transfer exchange current density on graphite and solid-state diffusivity in graphite particles as functions of temperature.[13] These parameters characterize the three key processes—ion transport through an electrolyte, lithium insertion/de-insertion kinetics, and lithium diffusion in solid particles—that control LIB charge and discharge. From 0°C to 60°C, the electrolyte conductivity, charge-transfer kinetics, and solid-state diffusivity increase by 3.08×, 93×, and 16×, respectively. Thus, a single TA action leads to major improvements in all three material properties. This is why, without substantially changing battery materials, intracell TA already achieves breakthroughs in low-temperature power and usable energy, as illustrated in Figure 1B. Comparing the current SOA cells with thermally actuated counterparts, the latter shows a leap in low-temperature power and energy

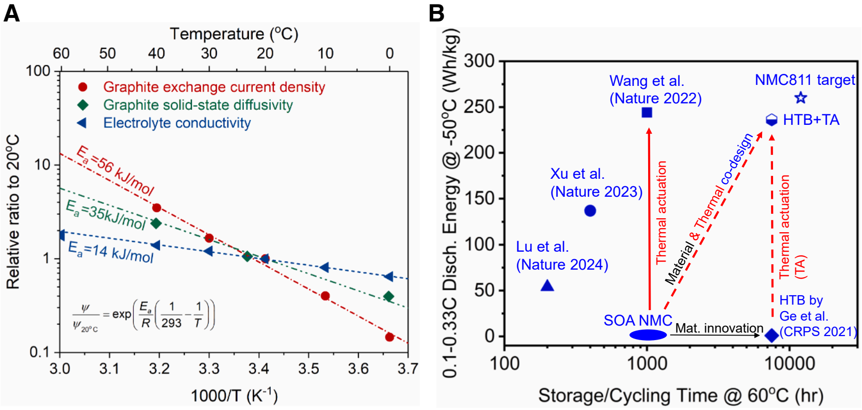

Figure 2. Material-thermal synergistic approach to ACBs

(A) Enhancement of key material properties with temperature[13]: electrolyte conductivity, exchange current density of lithium insertion kinetics on graphite, and solid-state diffusivity in graphite particles. Reproduced with permission.[13].

(B) An illustrative example for the NMC811 chemistry. Starting from the SOA baseline, Ge et al.[8] added a small amount of TAP in the electrolyte to obtain a heat-tolerant battery (HTB) version with marked improvement of thermal stability at 60°C. If thermal actuation (TA) is further applied to this HTB to recover power and usable energy at −50°C, the co-design (HTB + TA) is seen to be very close to the ACB target for NMC811 cells. Two ACB examples by materials innovation alone, Xu et al.[4] and Lu et al.,[5] are also displayed for comparison.

while keeping the high-temperature stability/cyclability unchanged. In contrast, materials innovation alone improves the low-temperature power/energy at the expense of diminishing high-temperature stability/cyclability.

TA is also bi-directional. Imagine a future battery cell with sluggish kinetics at room temperature that operates optimally at elevated temperatures, e.g., 60°C, for good power delivery and driving. Once such a battery, e.g., the dicarbonate electrolyte battery developed by Li et al.,[10] rests at open circuit upon vehicle parking, it cools down naturally to the ambient temperature. Then the battery enters dormancy, resulting in virtually no cell degradation and absolute safety. This is the new concept of batteries with high power on demand by TA while keeping low reactivity and high stability at rest for minimal self-degradation and maximum safety.

One example of TA practiced in commercial vehicles is Tesla's on-route battery warmup, which preheats the battery while navigating to a supercharge station. However, this practice has limited benefits because it takes ∼45 min of driving to warm up a battery from the freezing point to an optimal charging temperature of 45°C at a heating rate of ∼1°C/min for external heating. Additionally, consuming battery energy for heating while driving on an already low SOC battery poses great range anxiety. Indeed, the on-route warmup strategy appears to be ineffective, as many mainstream media reported in the 2023 winter that "electric vehicle charging stations [Tesla supercharge stations] became scenes of desperation: depleted batteries, confrontational drivers, and lines stretching out onto the street." The concept of TA discussed here, however, is broad and profound, being ultrafast, energy-efficient, and flexible in the choice of supplying heating energy (whether from a 100% SOC battery or from a roadside charger for a 0% SOC battery). Moreover, the TA opens viable doors to heat-tolerant electrolytes/electrodes and their batteries, promising great survivability and safety in surging air temperatures.

Material-thermal synergistic approach

As discussed above, materials and thermal synergistic co-design is arguably the most promising approach to realize ACB targets. Consider a case in point. Starting from the SOA baseline of today's NMC811/Gr pouch cells, Ge et al.[8] demonstrated that by adding 1.5wt % TAP in the standard electrolyte, the interphase layers can be significantly passivated to extend the cycling time at 60°C from 1,000 to 7,500 h, albeit room temperature power is reduced, and the dischargeable energy at −50°C becomes minimal. Such a heat-tolerant battery (HTB) via electrolyte innovation is illustrated in Figure 2B. Now, if we apply TA to this HTB version in the same way as the base case, it is projected that the dischargeable energy from −50°C ambient will reach 236 vs. 244 Wh/kg for the baseline without TAP addition,[11,14] because the more sluggish HTB version with TAP needs to heat up to ∼40°C instead of 25°C to match the same level of power as the baseline system without TAP and hence expends slightly more heating energy. Such a projection of combining materials innovation with TA is sketched in Figure 2B as a dashed line. Evidently, the synergistic approach is the fastest way to reach the ACB target. The same synergistic approach should also be applicable to the dicarbonate electrolyte battery of Taskovic et al.,[6] as well as to future batteries with condensed-matter or solid-state electrolytes. That is, one uses materials innovation to establish thermal tolerance and operational stability at high temperatures and an intracell TA structure[1,15] to activate and provide power/energy in low-temperature environments.

Paradigm shift in battery thermal management

Current thermal management systems are sophisticated, bulky, energy intensive, and costly because conventional LIB cells are designed and operated around *comfortable* 25°C due to its original design for man-portable electronics. This has become *awkward* 25°C for electric vehicles and grid-scale energy storage systems that necessarily experience extreme temperatures. As shown in Figure 3A, only between 10°C and 30°C are the LIBs immune from thermal management. Below 10°C, external heating is applied. Below −30°C, external heating becomes unbearably slow and energy inefficient so that current LIBs are deemed inoperable. Between 30°C and 45°C ambient, liquid or air-conditioning cooling is requisite, as LIB cells must be kept between 25°C and 35°C, and heat dissipation driven by rather small temperature differences becomes challenging. Above 45°C ambient, current LIBs are not recommended due to intense energy consumption to

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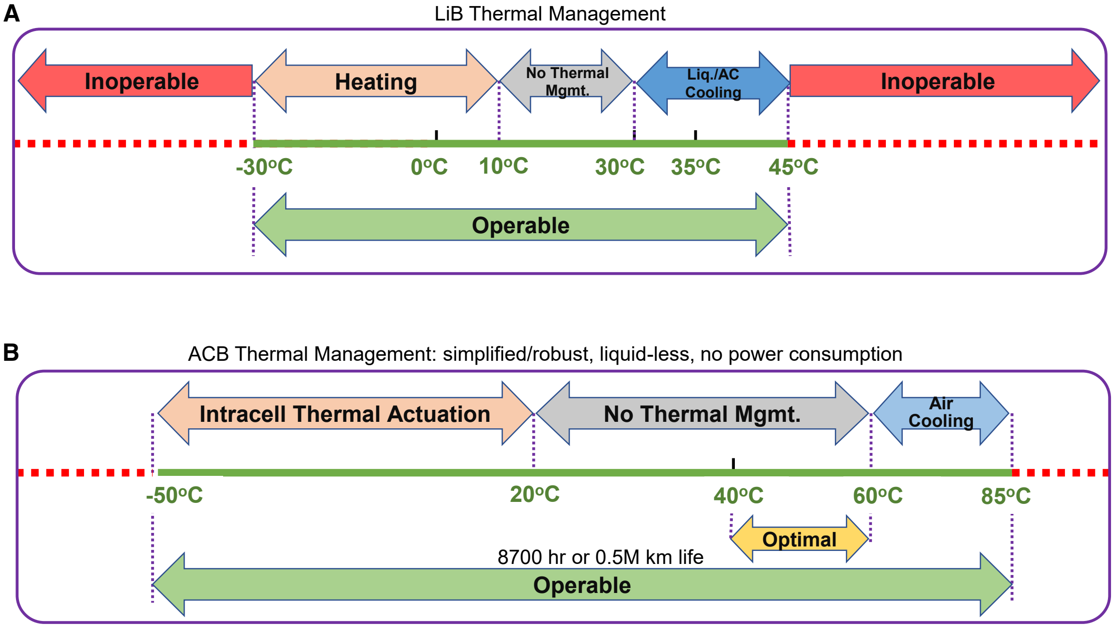

Figure 3. Paradigm shift in battery thermal management
(A) For conventional LIBs.
(B) For ACBs.

continuously run air conditioning, even during open-circuit storage. To the contrary, if ACBs via the material-thermal synergy described above are successfully deployed to stably operate up to, e.g., 60°C, no external thermal management would be needed between −50°C and 60°C, as schematically displayed in Figure 3B. When the cell temperature exceeds 60°C, only air cooling suffices, either passive or active, because the temperature difference between the battery and the coolant is enlarged by 5×–10×, thus greatly facilitating heat removal. The end result is a much simplified battery thermal management (BTM) system for ACBs, shown in Figure 3B, where there is no liquid, no pump, and no parasitic power consumption.

The added volume of internal heating elements to enable intracell TA are estimated to be ~0.1%, e.g., embedding a 10 μm heating foil in a 10 mm-thick pouch cell. Potential overheating risks due to the embedded heaters are mitigated by use of temperature-regulated switching (active) and positive-temperature-coefficient (PTC) heaters (passive). Critically, when TA is applied to HTBs, the temperature margins for appreciable degradation and thermal runaway become much higher, further relaxing the overheating risk of intracell TA.

Conclusions and outlook

ACB technology that requires power at low temperatures and operational stability at high temperatures presents daunting challenges for materials innovation alone. To date, materials advances toward ACBs largely lag in thermal stability/cyclability at high temperatures in the 70°C–85°C range. Highly volatile solvents with a boiling point around or less than 100°C are unsuitable for ACBs that must survive in surging air temperatures. The material-thermal synergistic approach appears more promising and fruitful. In the co-design approach, materials innovation focuses on improving thermal stability/cyclability at high temperatures, preferably up to 70°C–85°C, while power and resilience at low temperatures are enabled by intracell TA with low heating energy and rapid response. Meaningful advances for ACBs can now be quantified via Figure 1B. Finally, ACB technology will lend itself to simple BTM systems owing to a wide temperature range requiring no management, a low load of heat removal (thanks to the high round-trip efficiency of LIBs), and a large temperature difference driving heat dissipation and making air cooling feasible.

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AUTHOR CONTRIBUTIONS

Conceptualization and methodology, C.-Y.W. and K.Q.; writing, C.-Y.W., K.Q., and N.G.

DECLARATION OF INTERESTS

The authors declare no competing interests.

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