Challenges and Innovations of Lithium-Ion Battery Thermal Management Under Extreme Conditions: A Review

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Thermal management is critical for safety, performance, and durability of lithium-ion batteries that are ubiquitous in consumer electronics, electric vehicles (EVs), aerospace, and grid-scale energy storage. Toward mass adoption of EVs globally, lithium-ion batteries are increasingly used under extreme conditions including low temperatures, high temperatures, and fast charging. Furthermore, EV fires caused by battery thermal runaway have become a major hurdle to the wide adoption of EVs. These extreme conditions pose great challenges for thermal management and require unconventional strategies. The interactions between thermal, electrochemical, materials, and structural characteristics of batteries further complicate the challenges, but they also enable opportunities for developing innovative strategies of thermal management. In this review, the challenges for thermal management under extreme conditions are analyzed. Then, the progress is highlighted in two directions. One direction is improving battery thermal management systems based on the principles of heat transfer, which are generally external to Li-ion cells. The other direction is designing novel battery structures, which are generally internal of Li-ion cells such as smart batteries with embedded sensors and actuators. The latter approach could greatly simplify or even eliminate the need for battery thermal management under extreme conditions. New research integrating these two approaches is recommended. [DOI: 10.1115/1.4056823]

Keywords: Li-ion battery, thermal management, extreme conditions, electric vehicle, smart battery, intracell thermoregulation

1 Introduction

Lithium-ion (Li-ion) batteries have enabled the revolutionary development of electric vehicles (EVs). As shown in Fig. 1(*a*), the sales of EVs started in 2010 with the global market share negligibly small in the first few years, but the share has increased to nearly 9% in 2021 [1] and is expected to reach 70% in 2040 [2]. Some governments have come up with plans to end sales of fossil-fuel vehicles by 2035 [3,4]. Corresponding to the rapid growth in global sales, EVs will be increasingly used under extreme conditions, such as low temperatures, high temperatures (e.g., surging air temperatures in heatwaves), and fast charging, as schematically shown in Fig. 1(*b*). According to the targets set by the United States Advanced Battery Consortium (USABC) [5], EVs need to be able to operate from $-30\,^\circ\mathrm{C}$ to $+52\,^\circ\mathrm{C}$, survive $-40\,^\circ\mathrm{C}$ to $+66\,^\circ\mathrm{C}$ for 24 h, and charge 80% of capacity within 15 min, in addition to having high specific energy, long life, and low cost. However, as dominant power sources of EVs, Li-ion batteries do not work well under these extreme conditions. Low temperature dramatically reduces power performance and driving range, while increasing charging time. High temperature causes accelerated degradation; for instance, Li-ion cells degrade rapidly under temperatures greater than $35\,^\circ\mathrm{C}$ [6], which are becoming increasingly common in hot summers. Fast charging can cause not only lithium plating, especially charging at low temperatures, but also excessive heat generation. Both of them increase degradation and safety risks. Furthermore, Li-ion batteries may experience catastrophic thermal runaway [7], a process of uncontrollable temperature rise accompanied by smoke, fire, and/or explosion when they are abused, such as during the crash of EVs. Li-ion batteries may also suddenly go to thermal runaway when the EVs are not in use [8–10], creating a challenge that is very different from traditional internal combustion engine vehicles. EV fires are also more difficult to extinguish than internal combustion engine fires [9,11] due to co-existence of reactants in the same enclosed cell and propagation of thermal runaway from one cell to other cells in an EV.

Due to the interactions between thermal, chemical, and electrochemical phenomena, thermal management plays a central role in the performance, durability, and safety of Li-ion batteries. Great progress has been made in thermal management of Li-ion batteries, as already applied in commercial EVs. However, significant gaps exist, especially under extreme conditions. In particular, many design practices compete against each other, for example, cooling capability against heating capability; performance against added mass, volume, or cost; and normal operation against storage or thermal runaway. Therefore, a holistic and fundamental understanding of the challenges are necessary in developing effective thermal management strategies and methods for EV batteries under extreme conditions. Due to the interdisciplinary nature of the challenges, information exchange and collaboration among different research communities are needed. This review aims to encourage and inspire communications between the heat transfer community, battery community, and EV community for conceiving and developing bold, novel, yet feasible solutions. While this review focuses on EV applications, thermal management of Li-ion batteries under extreme conditions is also important for various other applications, such as consumer electronics, grid-scale energy storage, aerospace, and space.

Because of the interdisciplinary nature of thermal management for Li-ion batteries, some basics about Li-ion batteries and thermal issues in EV batteries are introduced first to help understanding of the topic by a broader community, especially the heat transfer community. Then, the challenges of EV battery thermal

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Contributed by the Heat Transfer Division of ASME for publication in the Journal of Heat and Mass Transfer. Manuscript received December 6, 2022; final manuscript received January 10, 2023; published online March 24, 2023. Editor: Portonovo Ayyaswamy.

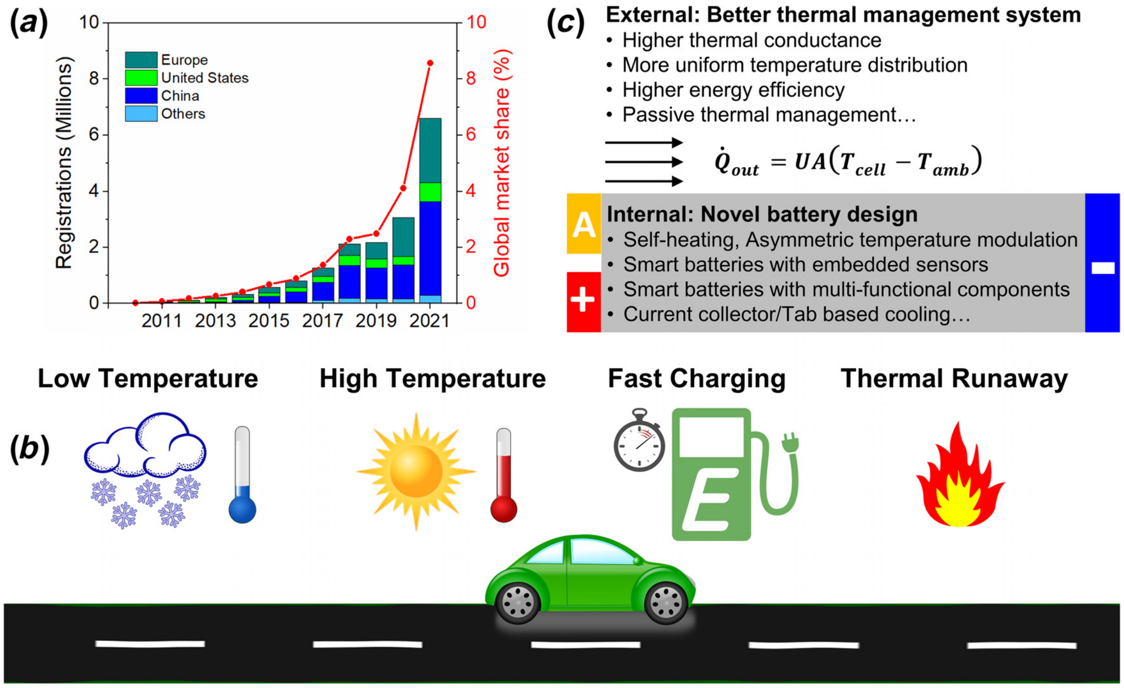

Fig. 1 (*a*) Global sales and market share of electric cars, 2010–2021 (reproduced with data from IEA, Paris under license CC BY 4.0 [1]). (*b*) Schematic of EVs under extreme conditions (made with icons from Openclipart under license CC0 1.0). (*c*) Schematic of progress in battery thermal management.

management under extreme conditions are discussed. Subsequently, progress in battery thermal management is reviewed in two directions. As schematically shown in Fig. 1(*c*), one direction is improving thermal management systems generally applied externally to Li-ion cells, such as increasing thermal conductance and improving temperature uniformity. The other direction is improving internal cell designs, such as batteries with self-thermoregulation and smart batteries with embedded sensors and multifunctional components. Finally, future research to enable EV battery operation under extreme conditions is discussed.

2 Basics of Li-Ion Batteries for Electric Vehicles

2.1 Structure and Components of Electric Vehicle Batteries. As schematically shown in Fig. 2(*a*) [12], a battery pack in an EV typically consists of multiple modules, and each module consists of multiple individual cells. In recent years, the cell-to-pack structure is being actively pursued (Fig. 2(*b*)) [12]. Note that individual cells may have various shapes and dimensions depending on EV models and years, but there are three primary formats for EV applications: cylindrical, prismatic, and pouch. As shown in Fig. 2(*c*) [13], in a cylindrical cell the electrodes and separators are wound into a jelly roll to be sealed in a cylindrical metal can. In a prismatic cell, the electrodes and separators may be wound or stacked and then pressed to be sealed in a prismatic metal can. In a pouch cell, the electrodes are generally stacked together to be sealed in a plastic/aluminum composite pouch.

2.2 Working Principle of Li-Ion Batteries. Although individual Li-ion cells in different EVs may come in different formats and dimensions, their basic components and working principles are similar. As shown in Fig. 3, a typical Li-ion cell has six basic components sealed inside a can or a pouch: negative current collector (Cu foil, ∼10 μm thick), negative electrode (anode), separator, electrolyte, positive electrode (cathode), and positive current collector (Al foil, ∼15 μm thick). The current collectors extend outside the cell through metal tabs or terminals to provide a path of electrons to the external circuit. The anode and the cathode are coated on their respective current collectors and electronically insulated from each other by the separator. Both anode and cathode consist of active materials, binders and additives. Graphite is the dominant anode active material, while silicon is being increasingly used. The cathode active material is more diverse, such as lithium cobalt oxide (LCO, $LiCoO_2$), lithium manganese oxide (LMO, $LiMn_2O_4$), lithium nickel manganese cobalt oxide (NMC, $LiNi_xMn_yCo_zO_2$), lithium nickel cobalt aluminum oxide (NCA, $LiNi_xCo_yAl_zO_2$), and lithium iron phosphate (LFP, $LiFePO_4$) [14]. NMC, NCA, and LFP are the major cathode materials for EV applications. The separator is a porous membrane typically made of polymers such as polyethylene and polypropylene (∼20 μm thick). The electrolyte, typically liquid, fills the micropores of the anode, the cathode, and the separator to provide a path for ionic transport. $LiPF_6$ dissolved in organic solvents has been the dominant electrolyte, but it is highly volatile and flammable. Various solid-state electrolytes are being actively developed, which are safer than liquid electrolytes. Depending on the design, a Li-ion cell may have one pair of anode and cathode that are wound (mostly in cylindrical and prismatic cells) or multiple pairs of electrodes that are stacked (mostly in pouch cells).

When a Li-ion cell is charged, lithium ions (Li^+) and electrons (e^-) leave cathode particles in a half-cell reaction called de-intercalation. While the lithium ions transport to the anode through electrolyte, the electrons transport to the anode through current collectors, tabs, and the external circuit. Then, the lithium ions and electrons enter the anode particles in the other half-cell reaction called intercalation. When a Li-ion cell is discharged, the processes are reversed, with lithium ions and electrons leaving the anode and entering the cathode. Equations (1) and (2) show the half-cell reactions during charge and discharge at cathode and anode, respectively. In the equations, M refers to transitional metal, including Co, Ni, Mn, or a combination of different metals, and C refers to graphite or hard carbon

$$\mathrm{LiMO_2} \underset{\text{DisCharge}}{\overset{\text{Charge}}{\rightleftharpoons}} \mathrm{Li}_{1-x}\mathrm{MO_2} + x\,\mathrm{Li}^+ + xe^- \tag{1}$$

$$\mathrm{C} + x\,\mathrm{Li}^+ + xe^- \underset{\text{DisCharge}}{\overset{\text{Charge}}{\rightleftharpoons}} \mathrm{Li}_x\mathrm{C} \tag{2}$$

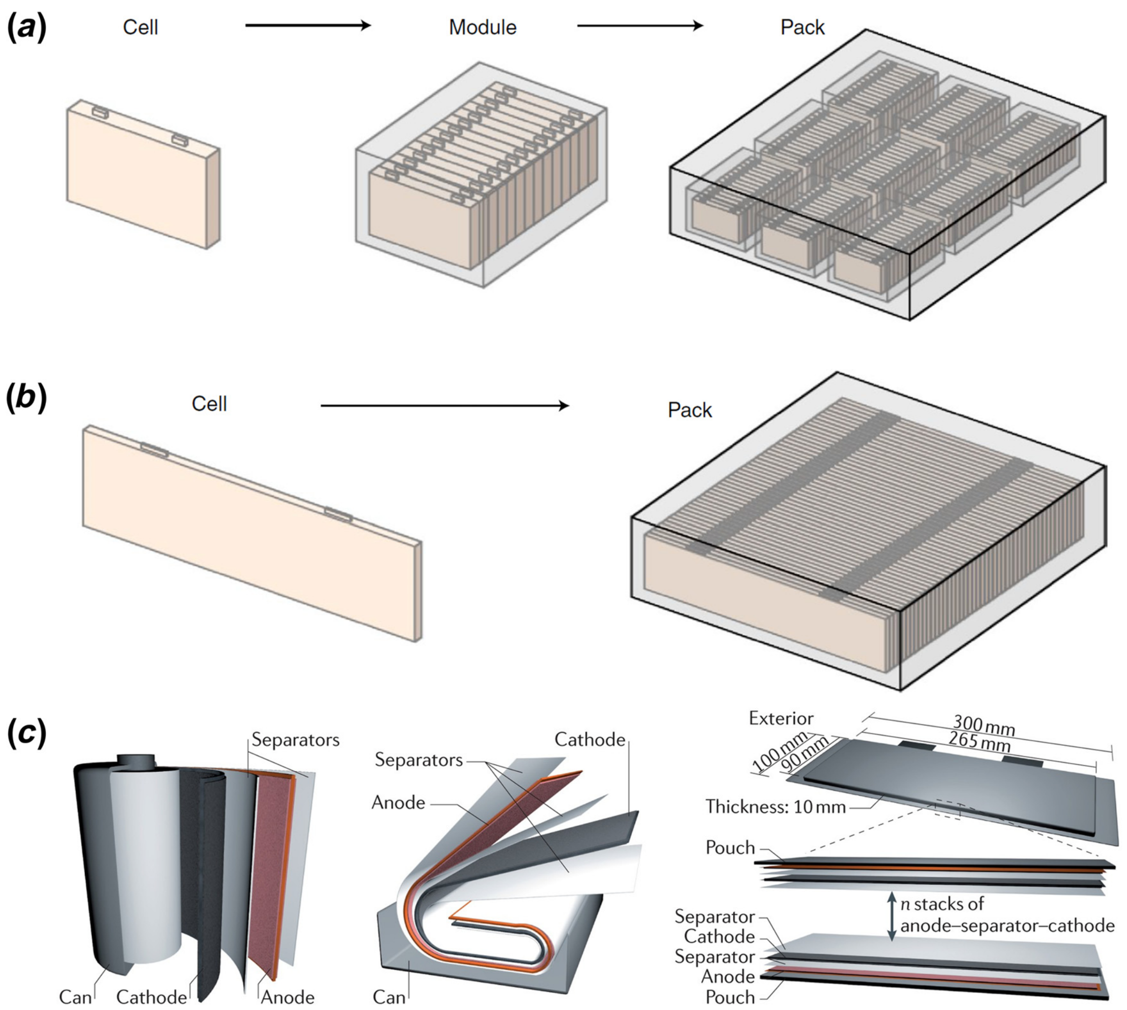

Fig. 2 Schematic of Li-ion battery pack, module and cells. (*a*) Cell-to-module-to-pack structure; (*b*) cell-to-pack structure; [12], and (*c*) different formats of cells [13]. (Reprinted with permission from Springer Nature © 2016).

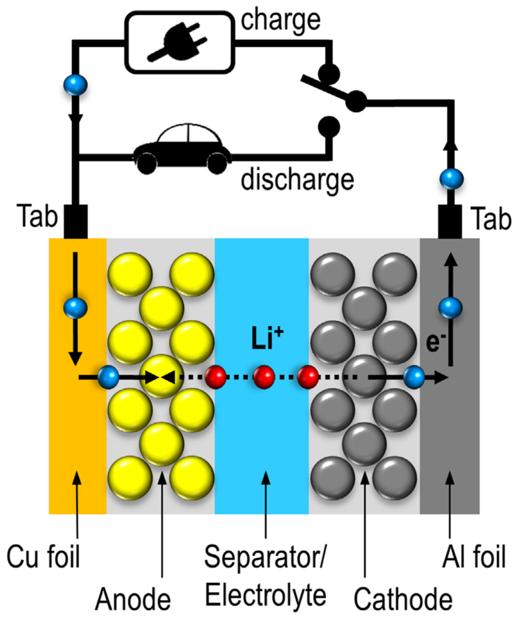

Fig. 3 Schematic of basic components and working principle of a Li-ion cell

Note that lithium ions and electrons may combine at the surface of graphite particles during low-temperature charging and/or fast charging when they cannot intercalate into graphite particles quickly enough. This phenomenon is called lithium plating, with the reaction described in the following equation:

$$x\,\mathrm{Li}^{+} + xe^{-} \xrightarrow{\text{Charge}} x\,\mathrm{Li} \tag{3}$$

2.3 Requirements of Li-Ion Batteries for Electric Vehicles.

Table 1 shows the targets of Li-ion batteries for EV applications set by USABC [5] and the European Council for Automotive R&D (EUCAR) [15]. It can be seen that EV batteries must meet targets of multiple parameters simultaneously. Many of these targets are competing against each other, such as specific energy against specific power and fast charging rate against life. Note that C rate is commonly used to specify charging or discharging rate of batteries. It is defined as the ratio of current (A) to nominal capacity (Ah) of a battery. For example, for a 15-Ah Li-ion battery 1 C discharge rate means that the discharge current is 15 A and 3 C rate means that the current is 45 A. Also note that SOC refers to state of charge, with 100% SOC meaning that a battery is fully charged and 0% SOC meaning that a battery is fully discharged. SOH refers to state of health, which is generally defined as the ratio of a battery's capacity to its original capacity.

Table 1 Targets of Li-ion cells for passenger EVs by USABC [5] and EUCAR [15]

Parameter	Unit	USABC	EUCAR 2030
Baseline range	km	320 km (200 miles)	400 km
Baseline test temperature	°C	30	25
Usable energy of pack	kWh	45 (at the end of life)	60 (estimate)
Specific energy (cell level)[a]	Wh/kg	350	450
Energy density (cell level)[a]	Wh/L	750	1000
Peak discharge specific power	W/kg	700 (30 s pulse)	1800 (10 s pulse at 50% SOC)
Peak discharge power density	W/L	1500 (30 s pulse)	4000 (10 s pulse at 50% SOC)
Fast charging rate	C (1/h)	3.2[b]	3.5
Life[c]	year/cycle	15 yr and 1000 cycles	EV lifetime and 150,000 km
Cell level cost	/kWh	$100	€70
Pack level cost	/kWh	$125	€84
Cell volume per battery pack	%	67 (estimate)	75
Cell weight per battery pack	%	67 (estimate)	80
Operation temperature range	°C	-30 to $+52$	Not specified
Survival temperature range[d]	°C	-40 to $+66$	Not specified
Unassisted operating at low temperature	N/A	>70% useable energy for C/3 discharge at $-20\,^\circ$C	600 W/kg peak specific power for discharge at $-25\,^\circ$C
Safety	N/A	Not specified	≤EUCAR safety level 4 (no fire or flame, no rupture, no explosion)
Self-discharge	%/month	<1	<1

[a] Tested at C/3 discharge rate.
[b] Estimated from the requirement of 80% ΔSOC in 15 min.
[c] SOH reaches 80% at the end of life.
[d] Soaked at $-40\,^\circ$C and $+66\,^\circ$C, respectively, for 24 h following certain procedures.

3 Thermal Issues of Li-Ion Batteries

Thermal issues are critical to Li-ion batteries, especially for electric vehicles. First, Li-ion battery performance, lifetime, and safety are highly dependent on temperature. Second, heat generation associated with Li-ion battery operation can be excessive during high-power charging and discharging. Third, thermal runaway and propagation may occur. Fourth, there exists nonuniform temperature distributions inside a Li-ion cell or between cells.

3.1 Effects of Temperature on Li-Ion Battery Performance, Lifetime, and Safety. As shown schematically in Fig. 4(*a*), the optimal temperature range of conventional Li-ion batteries is quite narrow, about 20–35 °C. Below 20 °C the performance drops due to slower reaction kinetics and diffusion, which cause a decrease in usable capacity and thus driving range of EVs (Fig. 4(*b*)) [16]. More concerning, charging at low temperature can cause lithium plating, which reduces battery life as indicated by SOH (Fig. 4(*c*)) and increases safety risk. Above 35 °C the batteries degrade rapidly due to growth of the solid-electrolyte interface (SEI) [17], even during rest [18]. Above ∼80 °C Li-ion batteries could self-heat from unwanted side reactions [19,20]. Above 135 °C commonly used polyethylene-based separator would melt. Above 160 °C, more side reactions would occur, accelerating self-heating and likely leading to energetic thermal runaway. As shown in Fig. 4(*d*), a 25-Ah pouch cell for EV applications went to thermal runaway after being overheated in an accelerating rate calorimetry (ARC) test [21]. Once the cell was heated to the self-heating temperature, heat generation from side reactions caused further increase of cell temperature under the adiabatic condition during ARC testing. When the cell temperature reached the threshold of thermal runaway, it increased sharply to the peak within a few seconds.

3.2 Heat Generation in Li-Ion Batteries. Heat generation in Li-ion cells comes from multiple sources, including ohmic heating, heating from reactions, entropic heating, and heating from mixing effects. The heat generation from mixing effects is generally negligible, so the heat generation rate $\dot{Q}_{\text{gen}}$ can be estimated by the following equation [22]:

$$\dot{Q}_{\text{gen}} = I(U_o - V_{\text{cell}}) - IT\left(\frac{\partial U_o}{\partial T}\right) \tag{4}$$

in which the first term on the right side describes irreversible heat generation from both ohmic heating and electrochemical reactions. The second term describes reversible entropic heat generation. More specifically, I is the current, U_o is the open-circuit voltage, V_{cell} is the cell voltage, T is the cell temperature, $\partial U_o/\partial T$ is the dependence of open-circuit voltage on temperature named as the entropic coefficient. Note that the irreversible heat generation is always positive during charging or discharging. The entropic heat generation can be positive or negative depending on the SOC and the direction of current (the cell being charged or discharged). Entropic heat generation is typically much less than the irreversible heat generation, especially during high-current charging or discharging [23,24].

To more easily understand the effects of different factors on heat generation, Eq. (4) can be written as

$$\dot{Q}_{\text{gen}} = I^2 R_{\text{cell}} - IT\left(\frac{\partial U_o}{\partial T}\right) \tag{5}$$

where R_{cell} is the direct current (DC) internal resistance of a Li-ion cell, including ohmic resistance, contact resistance, and reaction resistance [25]. The R_{cell} rapidly increases with lower temperature [26], suggesting that heat generation rate is higher at lower temperatures for the same current. Equation (5) also suggests that the heat generation rate increases rapidly with current, as shown in Fig. 5 [27].

3.3 Thermal Runaway. Thermal runaway of Li-ion cells can be triggered not only by thermal abuse (overheating) as described

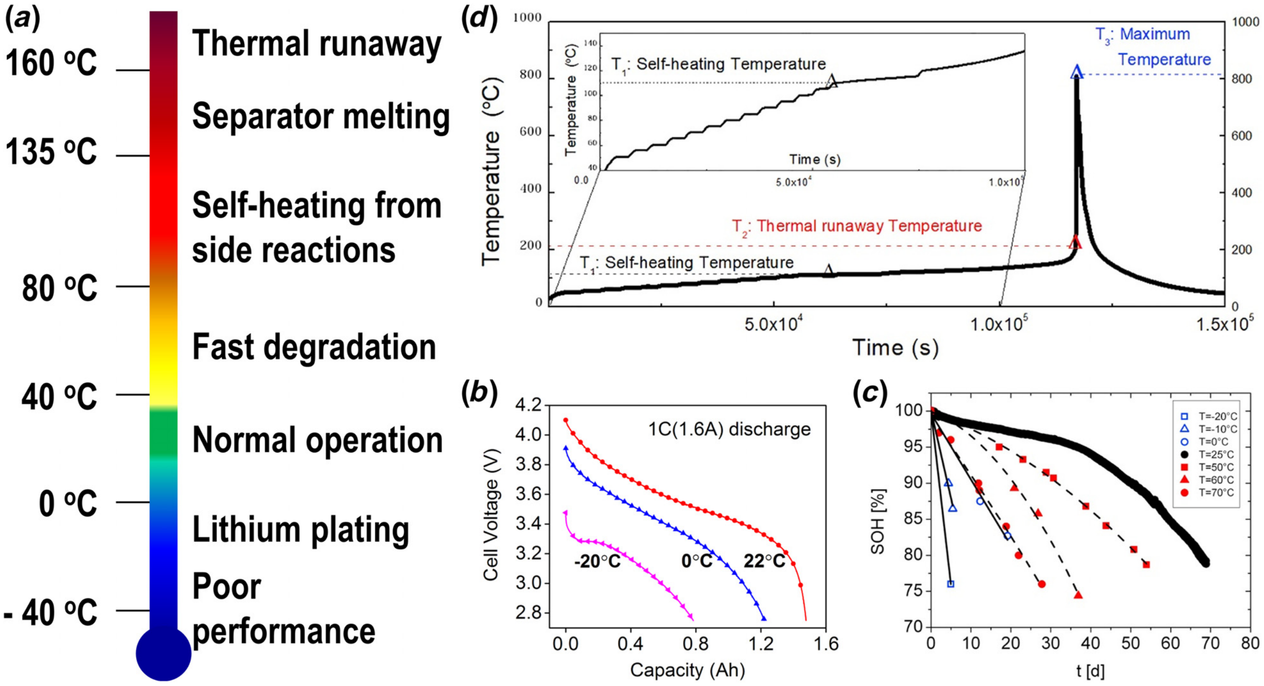

Fig. 4 **Effects of temperature on Li-ion battery. (*a*) Schematic, (*b*) effects on discharge performance [16] (Reprinted with permission from The Electrochemical Society) © 2014, (*c*) effects on lifetime [17] (Reprinted with permission from Elsevier © 2014), and (*d*) thermal runaway of a Li-ion cell triggered by overheating in an ARC test [21] (Reprinted with permission from Elsevier © 2018).**

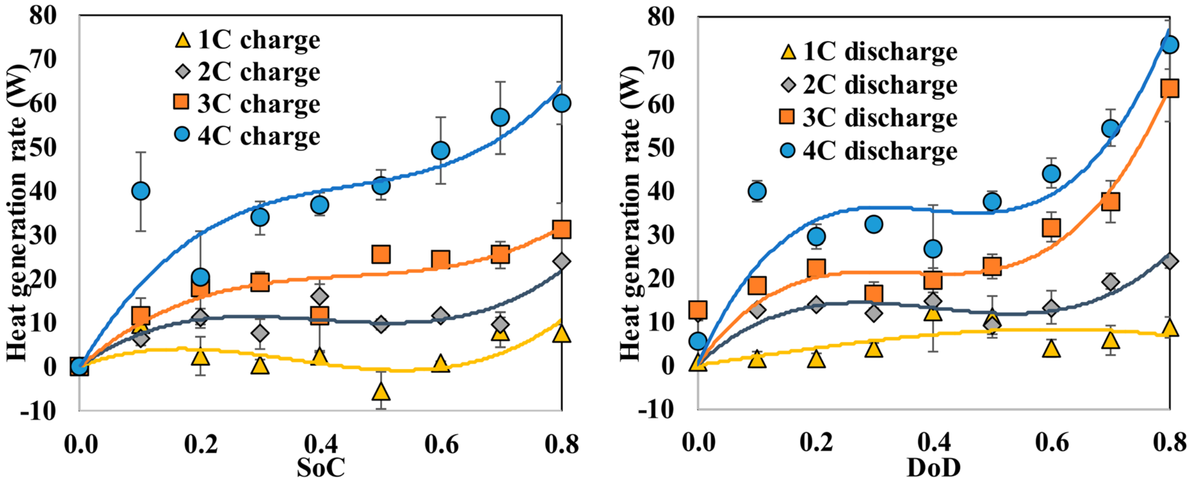

Fig. 5 **Heat generation rate of a commercial Li-ion cell at different charging and discharging rates [27] (Reused under license CC BY 4.0)**

above but also by mechanical abuse (e.g., penetration, indentation, or crush), electric abuse (overcharge, external short circuit, or repeated over discharge followed by charge), and internal short circuit [28]. For example, Fig. 6(*a*) shows the energetic thermal runaway of a 3-Ah cylindrical cell (18,650 format). The cell immediately exploded when penetrated and then caught fire with the surface temperature reaching over 1000 °C within two seconds. In addition to the rapid energy release, various toxic chemicals such as HF and CO can be generated during thermal runaway [29].

Thermal runaway triggered by thermal abuse, mechanical abuse, or electric abuse could generally be expected and prevented by better designs. But thermal runaway triggered by internal short circuit is difficult to detect or prevent. Several high-profile battery fires involving EVs [30,31], airplanes [32], smart phones [33], and grid-scale energy storage systems [34], which suddenly occurred after normal operation for months or years, have been attributed to internal short circuit. It remains a critical challenge that is actively investigated [35–37].

Furthermore, when one cell in a module or a pack goes to thermal runaway, it could trigger thermal runaway of adjacent cells through heat propagation [38,39], as evidenced in real-world EV fires and clearly demonstrated in Fig. 6(*b*) [39].

The large temperature rise during thermal runaway can be understood from Eq. (4). When thermal runaway occurs, all the stored electric energy in a Li-ion cell would convert to thermal energy, which can be estimated by assuming that $V_{\text{cell}} = 0\,\text{V}$. Neglecting the entropic heat generation, the total heat generation, Q_{gen}, can be described by Eq. (6) in which E_{nom} is the stored electric energy

$$Q_{\text{gen}} = E_{\text{nom}} \tag{6}$$

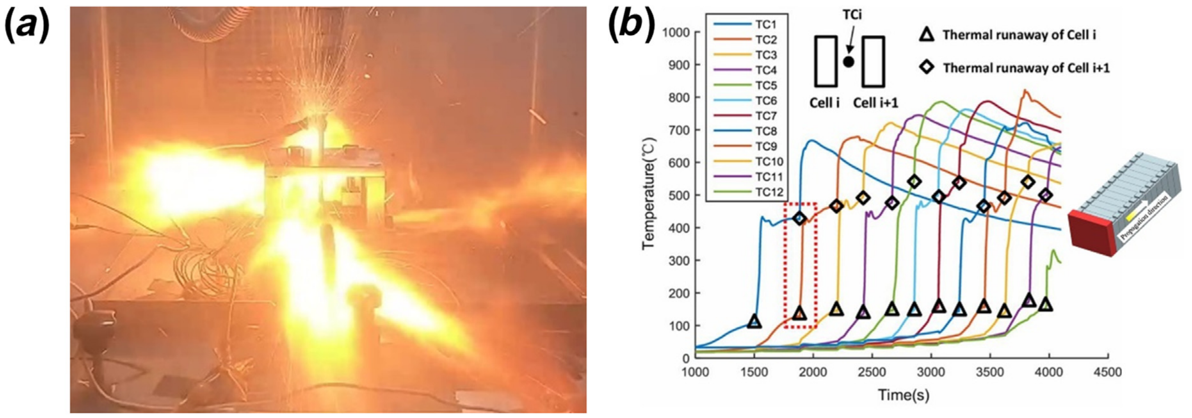

Fig. 6 (*a*) Thermal runaway of a Li-ion cell triggered by nail penetration in Zhang's laboratory and (*b*) propagation of thermal runaway from one cell to other cells in a module (Reprinted with permission from The Electrochemical Society © 2019) [39]

Assuming adiabatic boundary conditions, i.e., heat dissipation is negligible due to the rapid process of thermal runway, the heat generation will cause temperature rise of the cell according to the following equation:

$$Q_{\text{gen}} = mc_{p,\text{cell}}\Delta T_{\text{ad}} \tag{7}$$

In the equation, m is the mass of the cell, c_p is specific heat of the cell, and ΔT_{ad} is the maximum temperature rise of the cell. Then ΔT_{ad} can be estimated by the following equation [40]:

$$\Delta T_{\text{ad}} = \frac{E_{\text{nom}}/m}{c_p} = \frac{\text{SE}}{c_p} \tag{8}$$

in which SE is the specific energy of the Li-ion cell. Assuming $c_p = 1000\,\text{J kg}^{-1}\,\text{K}^{-1}$ [41], a Li-ion cell with a specific energy of 200 Wh/kg would yield a temperature rise of 720 °C during thermal runaway. It should be noted that heat generation during thermal runaway comes not only from the stored electric energy but also from reactions involving SEI decomposition, electrolyte decomposition, electrode decomposition, and electrolyte combustion, etc., [42]. Indeed, the heat generation from these chemical reactions may be several times higher than that from electric energy [29], and the actual temperature during thermal runaway can exceed the melting temperature of copper (1085 °C) [43]. The specific energy of state-of-the-art Li-ion cells is approaching 300 Wh/kg and that of next-generation lithium-metal based cells is approaching 500 Wh/kg. Such high specific energy can cause even higher heat generation than the estimation above.

The rapid temperature rise during thermal runaway can be understood from Eq. (5). Thermal runaway generally starts when a large area of the separator is damaged by mechanical, electrical, or thermal abuse, causing serious internal short circuit. The internal short circuit current would be very high [44,45], indicating that the heat generation rate is very high. Moreover, heat generation also leads to chemical reactions as noted above, thus further increasing the heat generation and temperature rise rate.

3.4 Nonuniform Temperature Distribution. Due to the low thermal conductivity of the separator, electrolyte, and cathode of Li-ion cells, the effective thermal conductivity of Li-ion cells in the through-plane direction is very low, typically less than $1\,\text{W m}^{-1}\,\text{K}^{-1}$. In comparison, the in-plane direction thermal conductivity is about 20–40 $\text{W m}^{-1}\,\text{K}^{-1}$ [41,46]. As a result, the temperature distribution through the thickness of a Li-ion cell would be nonuniform, which can be understood from the Biot number (Bi)

$$\text{Bi} = \frac{hL_c}{k_{\text{thru}}} \tag{9}$$

in which h is the convection coefficient, L_c is the characteristic length of a Li-ion cell, and k_{thru} is the effective through-plane thermal conductivity. For EV Li-ion batteries, h should be above $100\,\text{W m}^{-2}\,\text{K}^{-1}$ for practical air cooling and above $700\,\text{W m}^{-2}\,\text{K}^{-1}$ for practical liquid cooling [47]. Based on the data of a commercial pouch-type Li-ion cell for EV [46], with has a nominal capacity of 41 Ah, a thickness of 0.008 m (L_c of 0.004 m), and k_{thru} of $0.77\,\text{W m}^{-1}\,\text{K}^{-1}$, the Biot number would be 0.5 and 3.6 for $h = 100\,\text{W m}^{-2}\,\text{K}^{-1}$ and $h = 700\,\text{W m}^{-2}\,\text{K}^{-1}$. Such Biot number is greater than the typical criterion of 0.1 for temperature gradient to be neglected. Indeed, measurement of nonuniform temperature distributions in Li-ion cells has been actively pursued [16]. As shown in Fig. 7(*a*), the maximum temperature difference along the radial direction of a small cylindrical cell (18,650 format)

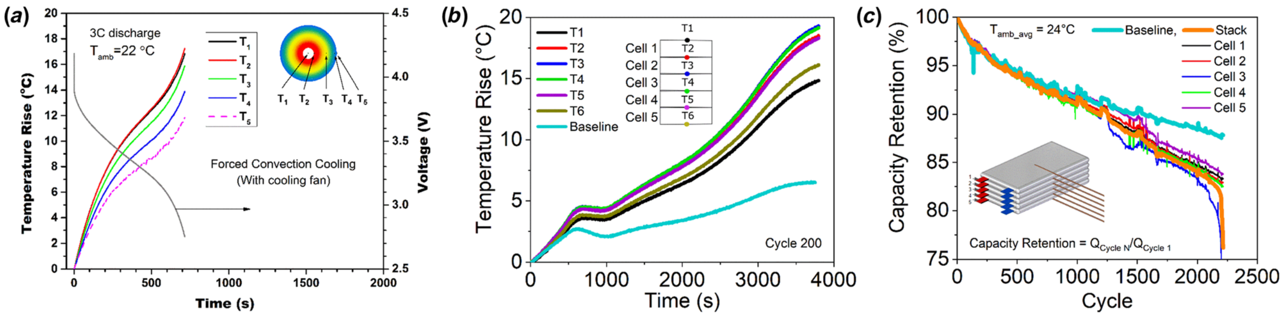

Fig. 7 Nonuniform temperature distribution in Li-ion cells. (*a*) Radial temperature distribution in a cylindrical cell [16] (Reprinted with permission from The Electrochemical Society © 2014); (*b*) temperature distribution across the thickness of a stack consisting of five parallel-connected pouch cells; and (*c*) nonuniform degradation of cells in the stack [49].

Table 2 USABC Li-ion battery thermal management system requirements [51]

Key parameter	Parameter details	Unit	Target
Operational life	At 30 °C	Years	15
Operating environment	Temperature range[a]	°C	−30 to +52
Pack temperature uniformity	ΔT: cell-to-cell	°C	<3
Cell temperature uniformity	ΔT: cell surface	°C	<3
System efficiency	Ambient (unconditioned)[b]	Ratio[d]	>15
	Active[c]		>4
Weight	Pack components	kg	<5.3
Volume	Pack components	L	<13.5
System cost	At 250 k units	$	<112

[a] System survival temperature range is −40 °C to +66 °C.

[b] In ambient (unconditioned) systems, electric power is only used for moving thermal fluids by pumps, fans, etc.

[c] In active systems, electric power is used directly for cell/battery thermal control and for moving thermal fluids.

[d] The ratio of heat transfer rate (removed, in Watts) versus electrical power (in Watts).

reached nearly 6 °C by the end of 3 C discharge. Note that temperature distribution can also be nonuniform in the in-plane direction of a Li-ion cell [48] and from cell to cell in a battery pack due to nonuniform heat generation and nonuniform cooling.

Since temperature greatly influences Li-ion cell performance and durability, nonuniform temperature distribution inside a cell or a module would cause nonuniform and accelerated degradation. This hypothesis was recently confirmed experimentally [49,50]. As shown in Figs. 7(*b*) and 7(*c*) [49], the middle cell in a stack degraded much faster than other cells in the stack and the baseline cell.

4 Requirements of Electric Vehicle Battery Thermal Management and Technological Gaps

Thermal management is critical for efficient, durable, and safe operation of Li-ion batteries in EV applications. Table 2 lists the USABC Li-ion battery thermal management system requirements [51]. The goals are to enable EVs to operate in a wide range of temperature (−30 to +52 °C) while minimizing temperature nonuniformity, electric power consumption, weight, volume, and cost.

All commercial EVs have battery thermal management systems. Table 3 lists key features of thermal management systems in three representative state-of-the-art mass-market commercial EVs with comparable battery capacities. Note that liquid-based thermal management is dominant in commercial EVs.

Table 3 Battery thermal management of representative mass-market EVs

EV model	Nominal battery capacity [52]	Max. charging power (C rate)	Survival temp. range	Battery warranty	Battery cooling	Battery heating
Nissan Leaf e+ 2022 [53,54]	62 kWh	100 kW (1.6 C)	−25 °C to 49 °C	8 yr/160,000 km (≥75% retention)	Air, passive	Resistive[a]
Tesla Model 3 RW 2021 [55]	60 kWh	170 kW (2.8 C) [52]	−30 °C to 60 °C	8 yr/160,000 km (≥70% retention)	Liquid + refrigeration	Heat pump
VW ID.3 Pro-Performance 2020 [56,57]	62 kWh	100 kW (1.6 C)	−30 °C to 60 °C	8 yr/160,000 km (≥70% retention)	Liquid + refrigeration[b]	Heat pump + resistive

[a] Automatically on below −20 °C and off above −18 °C [53].

[b] Automatically on above 32.5 °C [56].

Great progress has been made in EV batteries in the past decade. However, there are still many gaps between commercial EVs and the USABC or EUCAR targets, especially under extreme conditions. First, the maximum charging power of commercial EVs is generally lower than 3 C even under ideal conditions. The average charging power of these EVs would be even lower, especially at low temperatures [58,59]. Second, the survival temperature range is generally −30 °C to 60 °C for liquid-based thermal management and −25 °C to 49 °C for air-based thermal management. This range is still narrower than the USABC target range of −40 °C to 66 °C. Third, electricity consumption is greatly increased at extreme temperatures [60], which reduces driving range of EVs [61,62]. Note that the dramatic drop of driving range at low temperatures is not only from energy consumption by cabin heating. The higher resistance at lower temperature would cause Li-ion cells to reach cutoff voltage earlier, reducing usable capacity as shown in Fig. 4(*b*). Furthermore, the power of regenerative braking (amounting to 20–25% of cruise range) is dramatically reduced below 10 °C and becomes almost completely disabled below −5 °C [63]. Fourth, battery warranty is typically

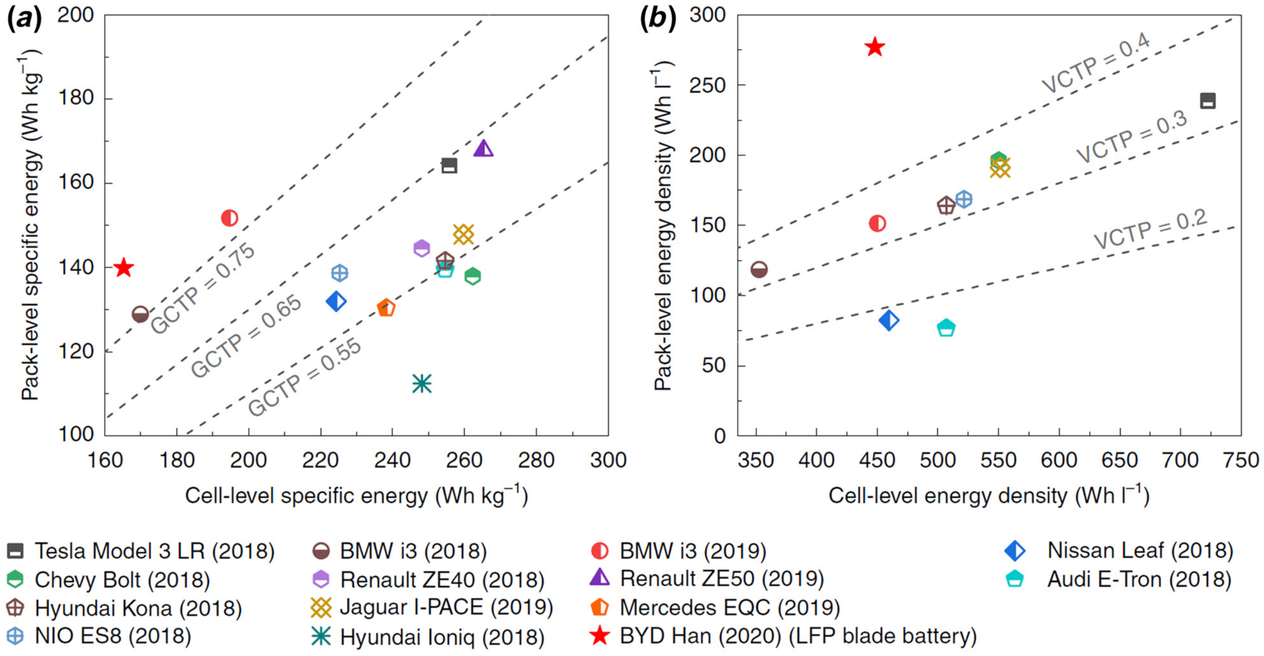

Fig. 8 Cell-level and pack-level specific energy and energy density of some commercial EVs [12] (Reprinted with permission from Springer Nature © 2021)

8 yr or 160,000 km (about 400 cycles for a 60-kWh battery) whichever comes first, and the battery capacity retention could drop to as low as 70% by the end of the warranty. These are lower than the life target of USABC. Fifth, specific energy at cell and pack levels is still significantly lower than USABC and EUCAR target, especially at pack level, as shown in Fig. 8. The figure also shows that cell-to-pack ratio is still lower than the targets for most commercial EVs, especially the volumetric cell-to-pack. Last but not least, EV battery fire has become a critical challenge as discussed earlier.

To fill the gaps between commercial EV batteries and the automotive targets, especially those related to thermal issues, it is important to fundamentally understand the challenges of battery thermal management under extreme conditions, as analyzed and discussed in Sec. 5.

5 Challenges of Thermal Management for Li-Ion Batteries Under Extreme Conditions

The challenges for battery thermal management under extreme conditions are discussed below, focusing on four areas: cooling at high temperatures, heating at low temperatures, fast charging, and thermal runaway.

5.1 Challenges for Battery Cooling at High Temperatures. Electric vehicle battery cooling has received the most attention. Various cooling methods have been developed, such as air cooling, direct liquid cooling (immersion cooling), indirect liquid cooling, and phase change material (PCM) cooling, as schematically shown in Fig. 9 [64].

The performance of air and liquid cooling can be evaluated through the following equation:

$$\dot{Q}_{\mathrm{out}} = UA(T_{\mathrm{cell}} - T_{\mathrm{amb}}) \tag{10}$$

in which $\dot{Q}_{\mathrm{out}}$ is the heat dissipation rate of cells, U is the effective heat transfer coefficient, A is the effective heat transfer area, T_{amb} is the ambient temperature. The product of UA is also referred to as thermal conductance. Equation (10) implies three ways of enhancing cooling: increasing U, increasing A, or increasing temperature difference ($T_{\mathrm{cell}} - T_{\mathrm{amb}}$). The heat transfer area A could be increased by using fins, but this approach will reduce usable pack volume. U could be increased by using active liquid cooling instead of passive air cooling, but it increases the cost/mass of the overall battery system (pumps, valves, heat exchangers, etc.), increases parasitic energy consumption, and has reliability and safety concerns [65,66]. Moreover, increasing U would increase Biot number according to Eq. (9) and thus nonuniformity of temperature distribution inside Li-ion cells. The temperature difference could be increased by either reducing T_{amb} or increasing T_{cell}. T_{cell} must be always higher than T_{amb} for cooling. If T_{amb} is high, e.g., 45 °C in heatwaves, T_{cell} would be higher and cause fast degradation for conventional Li-ion cells as discussed earlier. While integration of liquid cooling with refrigeration can be used to reduce T_{amb}, as employed in many commercial EVs, refrigeration consumes extra electric energy. The energy consumption increases significantly with higher environmental temperature due to the decrease of coefficient of performance of refrigeration systems. Further, keeping refrigeration on when vehicles are parked is impractical.

Another issue of battery cooling is the temperature variation of coolant. It can be described by the following equation which is derived from energy balance:

$$\dot{Q}_{\mathrm{out}} = \rho \dot{V} c_{p,\mathrm{coolant}}(T_{\mathrm{out,coolant}} - T_{\mathrm{in,coolant}}) \tag{11}$$

In the equation, ρ, $\dot{V}$, and $c_{p,\mathrm{coolant}}$ are density, volumetric flowrate, and specific heat of coolant. $T_{\mathrm{out,coolant}}$ and $T_{\mathrm{in,coolant}}$ are the outlet and inlet temperature of the coolant. For air-based cooling, the density and specific heat of air are small, so the volumetric flowrate needs to be large and air flow properly regulated [47,67,68] to control the coolant temperature rise and its influence on cell-to-cell temperature distribution.

Assuming that all the PCM melts, neglecting temperature gradient inside the PCM, and neglecting heat transfer from the PCM to ambient, the performance of PCM cooling can be evaluated by the following equation:

$$Q_{\mathrm{out}} = m_{\mathrm{PCM}}\lambda_{\mathrm{PCM}} + m_{\mathrm{PCM}}c_{p,\mathrm{PCM,eff}}(T_{\mathrm{PCM},f} - T_{\mathrm{PCM},i}) \tag{12}$$

where Q_{out} is the total heat transfer from cells to PCM, m_{PCM} is the mass of PCM, λ_{PCM} is the latent heat of fusion of PCM per

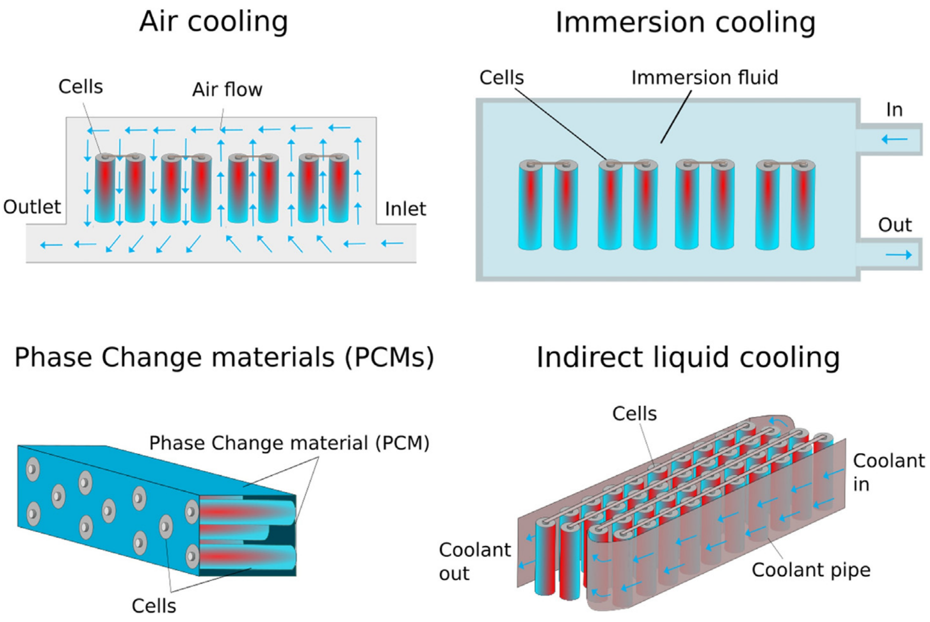

Fig. 9 Schematic of common cooling methods for Li-ion batteries [64] (Reused under license CC BY 4.0)

unit mass, $c_{p,\mathrm{PCM,eff}}$ is the average effective specific heat of PCM, $T_{\mathrm{PCM},f}$ is the final temperature of PCM, and $T_{\mathrm{PCM},i}$ is the initial temperature of PCM. The advantage of PCM lies in its latent heat of fusion, which could enable passive cooling and simplify the thermal management system. Therefore, PCM-based thermal management has received significant attention in recent years [69,70]. However, several challenges still exist. First, as pointed out by Longchamps et al. [40], the latent heat of fusion of commonly proposed PCM is still not high enough for EV battery thermal management, especially for next generation high-energy-density battery packs. Although PCM cooling can be combined with other cooling methods [71] to enhance cooling capability, the combination increases system complexity. Second, common PCMs has low thermal conductivity, which limits their cooling performance [72]. Third, many PCMs are combustible, which poses a safety concern in the event of PCM leakage or battery thermal runaway. In addition, PCM could greatly increase energy consumption for heating Li-ion batteries from low temperatures, as to be discussed below.

5.2 Challenges for Battery Heating at Low Temperatures.

Electric vehicle battery heating has not received much attention until recent years. As indicated in the commercial EV battery thermal management systems, preheating the batteries at low ambient temperature is a primary strategy. Various preheating methods have been used for EVs, such as positive temperature coefficient (PTC) heater (plus air or liquid coolant), heat pump, thermal energy storage, or an integration of these methods [73]. Energy consumption and heating time are two important factors in consideration of battery heating at low temperatures. Energy consumption for heating can be evaluated through the following equation:

$$\Delta E_{\mathrm{st,cell}} = m_{\mathrm{cell}} c_{p,\mathrm{cell}} \left(T_{\mathrm{cell,\,final}} - T_{\mathrm{cell,\,initial}}\right) \tag{13}$$

Then the ratio of energy consumption to stored electric energy can be described by the following equation:

$$\frac{\Delta E_{\mathrm{st,cell}}}{E_{\mathrm{nom,cell}}} = \frac{m_{\mathrm{cell}} c_{p,\mathrm{cell}} \left(T_{\mathrm{cell,\,final}} - T_{\mathrm{cell,\,initial}}\right)}{m_{\mathrm{cell}}\mathrm{SE}} = \frac{c_{p,\mathrm{cell}} \left(T_{\mathrm{cell,\,final}} - T_{\mathrm{cell,\,initial}}\right)}{\mathrm{SE}} \tag{14}$$

Assuming that $c_{p,\mathrm{cell}} = 1000\,\mathrm{J/kg\ K}$, $T_{\mathrm{cell,\,initial}} = -30\,^{\circ}\mathrm{C}$, $T_{\mathrm{cell,\,final}} = 0\,^{\circ}\mathrm{C}$, SE = 250 Wh/kg and that all the electric energy is used to heat the Li-ion cells, the consumed electric energy would be 8.3 Wh/kg, or 3.3% of stored electric energy. Such a percentage of energy consumption is not high considering the benefits of heating Li-ion cells to 0 °C, including enhanced driving range, faster charging, enabled regenerative braking, and longer life. However, it is important to note that if the heating is through liquid as in commercial EVs, the liquid in the loop must be also heated, which would consume more energy.

Heating time can be estimated from energy consumption and heating power using the following equation:

$$t_{\mathrm{heating}} = \frac{N_{\mathrm{cells}} m_{\mathrm{cell}} c_{p,\mathrm{cell}} \left(T_{\mathrm{cell,\,final}} - T_{\mathrm{cell,\,initial}}\right)}{P_{\mathrm{heating}}} = \frac{\left(\frac{E_{\mathrm{tot}}}{\mathrm{SE}}\right) c_{p,\mathrm{cell}} \left(T_{\mathrm{cell,\,final}} - T_{\mathrm{cell,\,initial}}\right)}{P_{\mathrm{heating}}} \tag{15}$$

where N_{cells} is the number of cells in the pack, E_{tot} is the total stored electric energy in the pack, and P_{heating} is the heating power. For example, when a PTC heater with a maximum power of 5.5 kW is used to heat a 60-kWh battery from −30 °C to 0 °C, assuming $c_{p,\mathrm{cell}} = 1000\,\mathrm{J/kg\ K}$ and SE = 250 Wh/kg, it would take 0.36 h or 22 min even if all the heat generated by the PTC is used to heat only the cells. The heating time would be longer if thermal mass of the coolant is considered. Heat pumps have been increasingly used in commercial EVs to reduce electric energy consumption of battery heating. However, the coefficient of performance of a heat pump decreases significantly at lower temperature [74], and it usually does not work well at temperatures below −10 °C [75].

5.3 Challenges of Thermal Management for Fast Charging.

Thermal management for fast charging is quite complicated. As shown in Fig. 10 [76], for fast charging at low ambient temperatures, the battery must be heated to prevent degradation from lithium plating. For fast charging at high ambient temperatures, the battery must be cooled to reduce degradation caused by SEI growth and to prevent overheating. A recent study, which involved 18 models of EVs for amounts of degradation over 1 yr,

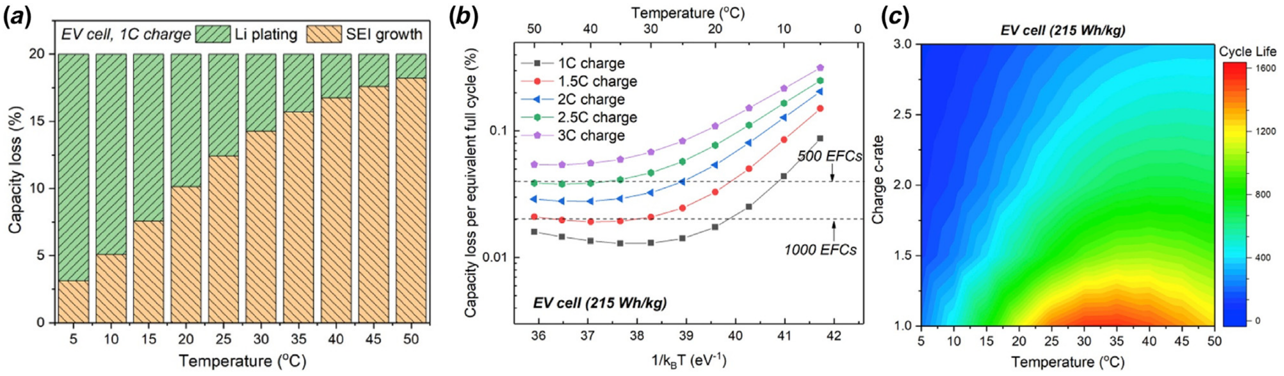

Fig. 10 (*a*) Breakdown of total capacity loss at the end of life of an EV cell with 1 C charge; (*b*) aging rate versus temperature for the EV cell at various charge rates; and (*c*) contour plots showing the impacts of charge rate and temperature on the cycle life of EV cells. The aging rate and cycle life are defined at 20% total capacity loss [76]. (Reprinted with permission from Elsevier © 2018).

suggested that fast charging caused much faster degradation than normal charging [77]. In addition, temperature nonuniformity inside a Li-ion cell would be much larger during fast charging than during normal charging [23], which can exacerbate lithium plating [78,79].

5.4 Challenges of Thermal Management for Thermal Runaway. Thermal runaway of EV batteries, in the form of smoke, fire, or/and explosion, has received considerable attention in recent years due to its costly consequences. There are several challenges for thermal runway from a thermal management perspective. First, a large amount of heat is generated quickly during thermal runway of Li-ion cells which could cause temperature increase of more than 800 °C in a few seconds or faster [7], as shown in Fig. 6(*a*). Such rapid heat generation and temperature rise, as well as the low thermal conductivity of Li-ion battery (LIB) materials, make it impractical to stop thermal runway once started even by liquid nitrogen [80]. Second, thermal runaway can propagate from one cell to adjacent cells (Fig. 6(*b*)) and EV battery fires can reignite after being put out. Therefore, in some EV firefighting practice an entire EV was put into a large container of water to prevent fire reignition [11]. Third, thermal runaway can occur due to various causes, such as mechanical abuse, electric abuse, thermal abuse, or internal short circuit. In particular, thermal runaway due to internal short circuit is the likely cause of EV fires under noncrash conditions. It remains a challenge due to difficulties in early detection, prevention, and mitigation [35]. Fourth, temperature distribution is highly nonuniform during thermal runaway [37,44,45], which makes surface temperature measurement ineffective in early detection. For example, Figs. 11(*a*) and 11(*b*) shows temperatures of a 3-Ah Li-ion cell during thermal runaway triggered by nail penetration [36]. The internal temperature at the internal short circuit location, as measured at the nail tip, showed multiple peaks that greatly exceeded the safety limit. However, the surface temperature showed little increase until the onset of thermal runaway. The measured cell voltage also showed little change during early stages of internal short circuit.

To address the challenge of EV battery thermal management under extreme conditions, various efforts been made and can be broadly categorized into two directions. The first direction is improving thermal management system, which is generally external to Li-ion cells. The other direction is improving battery design, which is generally internal to Li-ion cells. These two directions are discussed below.

6 Progress on Enhancing Current Thermal Management Systems

Taking inspirations from various heat transfer techniques is a natural approach to improving battery thermal management for extreme conditions. Numerous efforts have been made in this direction and a few are highlighted below.

6.1 Air-Based Thermal Management. As discussed earlier, air-based thermal management has the advantage of low cost, simplicity, high reliability, and great electric insulation, but it also has the disadvantage of lower thermal conductance (*UA*). The efforts on air-based thermal management have been focusing on increasing thermal conductance. For example, Han et al. [47] proposed an air-cooling design with winglet arrays, which can enhance the heat transfer coefficient and thus the thermal conductance. Li et al. [81] proposed adding fins to cells to increase heat

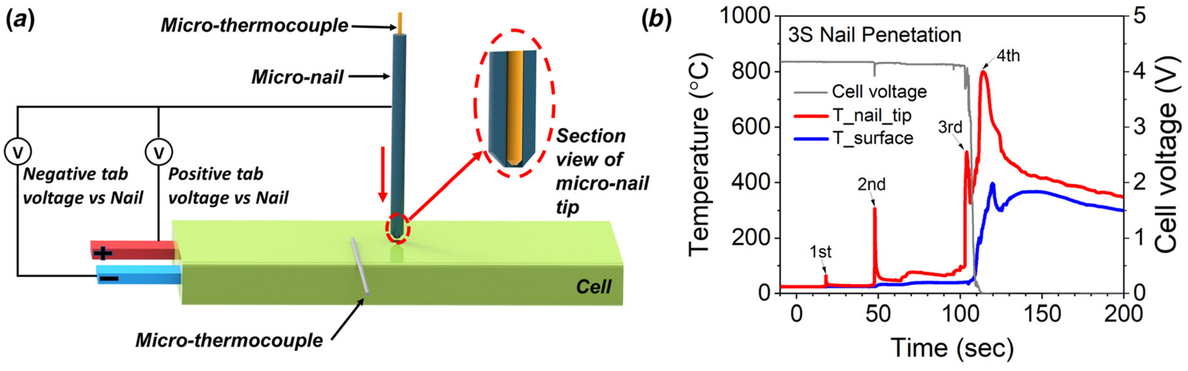

Fig. 11 Nonuniform temperature distribution during thermal runaway of Li-ion cells triggered by nail penetration. (*a*) Schematic of experimental setup and (*b*) measured temperatures and cell voltage [36] (Reused under license CC BY 4.0).

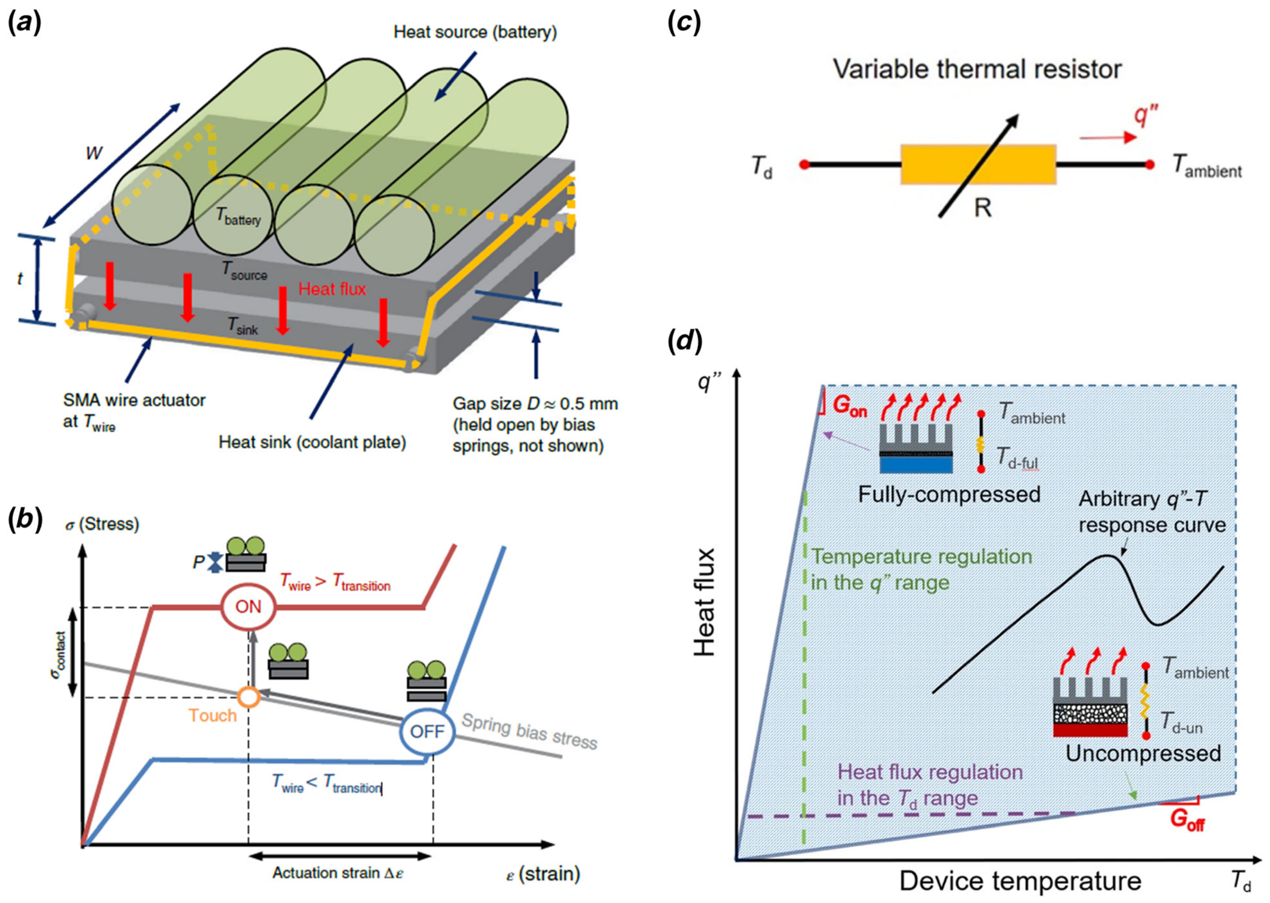

Fig. 12 Regulation of interfacial thermal resistance between Li-ion batteries and heat sink by a thermal switch. (*a*) and (*b*) An ON–OFF thermal switch based on a shape memory alloy [91] (Reprinted with permission from Springer Nature © 2018); (*c*) and (*d*) a continuously tunable thermal switch based on compressible graphene composite foams [92] (Reused under license CC BY 4.0).

transfer area. Ren et al. [82] used a U-shaped microheat pipe array to enhance temperature uniformity, but the array is still thick. There are also many other types of heat pipes, such as vapor chamber and oscillating heat pipe, but every type has its challenges for practical EV applications [83].

6.2 Liquid-Based Thermal Management. Liquid-based thermal management has been the most widely used strategy in commercial EVs due to its stronger cooling capability than air cooling. However, liquid-based systems are prone to leakage, which greatly heightens the risk of electric short circuit in high-voltage (400 or 800 V) systems [65,66]. Liquid-based systems are much more complicated and expensive than air-based systems. They also have the potential risk of flow channel clogging. To enhance safety, immersion cooling, or direct cooling, using dielectric and flame-retardant coolant has been actively investigated in recent years [64]. However, its effects on energy density, low temperature performance, and durability of EV batteries remain to be justified. The heating time and energy consumption for preheating at low temperature could be significantly increased due to the additional thermal mass of liquid. Moreover, the flow and thermal behaviors of a two-phase immersion cooling system are more complicated than a single-phase system.

6.3 Phase Change Material Based Thermal Management. While PCM-based thermal management has many advantages as a passive cooling, it has challenges of insufficient latent heat of fusion, low thermal conductivity, combustibility, and low temperature performance. Great efforts have been made to address these challenges [84,85]. For example, metal or graphite-based fins, foam, or nanoparticle additives are used to enhance effective thermal conductivity of PCM. Heat pipes are also used to enhance effective thermal conductivity of a PCM-based system. To reduce safety concerns, flame retardant additives are added to the PCM. However, the latent heat of fusion still needs to be greatly enhanced [40] and the heating time dramatically reduced [86] to be practical for EV applications.

6.4 Emergency Cooling. While the progress in battery cooling is mostly for normal conditions, there are increased efforts in emergency cooling for thermal runaway conditions. Kritzer et al. [87] proposed an emergency cooling method that is integrated with carbon dioxide (CO_2) based mobile air conditioning systems. The proposed battery system consists of cells equipped with thermocouples and a cooling system. Each thermocouple is linked with a CO_2 tube and a corresponding outlet valve to enable directed release of the cooling medium into exact areas where the overheating is detected. The concept was demonstrated in a module of four pouch cells that was overcharged without thermal runaway. More recently, emergency cooling using refrigerants that can be integrated with an EV heat pump system is reported [88]. In addition, emergency cooling using water mist [89] and dielectric fire-suppressing fluid [90] are also reported. However, emergency cooling devices lower the pack energy density too much; as such, its applicability in EV battery packs remains to be seen.

6.5 Thermal Switch for Regulation of Interfacial Thermal Resistance. While the interfacial thermal resistance between Li-ion cells and cooling channels is typically fixed, the concept of regulating the resistance according to operating temperature, sometimes called thermal switch, has been pursued in recent years. Hao et al. [91] used a shape memory alloy-based regulator to change the interfacial thermal resistance. As shown in Figs. 12(*a*) and 12(*b*), cells and heat sink will be separated (OFF state) when the temperature is low to increase interfacial thermal resistance and reduce heat loss. When the temperature is high, the cells and heat sink will be tightly connected (ON state) to reduce interfacial resistance and enable effective cooling. More recently, Du et al. [92] reported a method based on compressible graphene composite foams that allow a wide range of continuously tunable and fast thermal switching as shown schematically in Figs. 12(*c*) and 12(*d*). This thermal switching concept could be difficult for implementation in an EV battery due to the increase of system complexity and the risk of mechanical failures during long-term cycling.

7 Progress in the Direction of Novel Battery Designs

While the conventional approach focuses on improving external thermal management systems, an alternative approach focuses on novel battery structure designs capable of intracell thermoregulation. As shown in some highlighted examples below, this approach could reduce or even eliminate the dilemmas faced by conventional battery designs, and thus greatly simply or eliminate external thermal management. This approach has received increased attention in recent years. Note that there are also numerous efforts from the perspective of developing new materials. They have been extensively reviewed [93–96], and thus are not discussed here.

7.1 Self-Heating Li-Ion Battery for Superior Low-Temperature Performance. As discussed earlier, conventional efforts of improving the low-temperature performance of EV Li-ion batteries focus on externally heating the cells by an electric heater or heat pump through circulation of air or liquid coolant in addition to electrolyte improvement. This conventional approach suffers the problems of long heating time, e.g., more than 20 min for heating from −30 °C to 0 °C, and low heating efficiency [97]. To address this problem, Wang et al. [98,99] developed a novel self-heating Li-ion battery (SHLB), also called All-Climate Battery, which can rapidly and efficiently self-heat from −30 °C to 0 °C within 20 s. After the rapid self-heating, the power performance can increase by ∼10 times, which not only extends EV driving range at low temperature [100] but also enables fast charging of EV at temperatures as low as −50 °C [101,102]. Moreover, this design is simple and material-agnostic, as has been successfully demonstrated in EVs.

Figures 13(*a*) and 13(*b*) show schematically the structure and working principle of the SHLB [98,99]. One or multiple layers of nickel foil is embedded inside the SHLB cell. One end of the nickel foil is welded to the negative terminal of the cell and the other end extends outside the cell as a third terminal, called the activation terminal. A temperature-controlled switch is placed between the activation terminal and the positive terminal. When the switch is turned on at a low temperature, current flows through the nickel foil and generates heat internally to warm up the cell (self-heating mode) as shown in Fig. 13(*c*). When the cell temperature rises to a set value with cell performance recovered, the switch turns off automatically, reverting the cell to the baseline mode where an external load is connected between the positive and negative terminals, and no current flows through the nickel foil. Since the nickel foil is embedded inside the cell, all energy consumed during the self-heating process is efficiently used to warm up the cell. Furthermore, it has been demonstrated that energy from regenerative breaking during EV driving at low

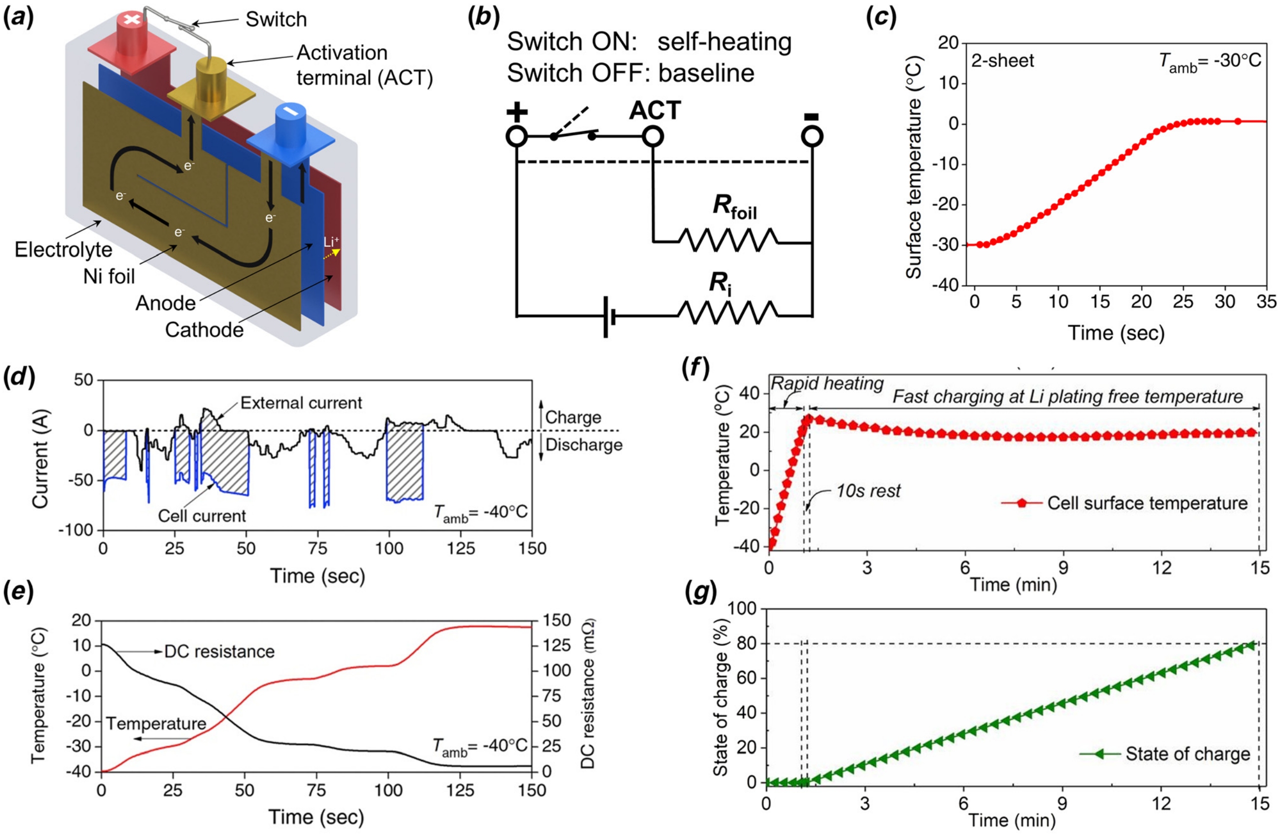

Fig. 13 Self-heating Li-ion battery (All-Climate Battery). (*a*) Schematic of cell structure; (*b*) electric circuit representation of self-heating mode and baseline mode; (*c*) cell temperature during self-heating from −30 °C [98,99] (Reprinted with permission from Elsevier © 2016); (*d*) and (*e*) current, cell temperature and resistance during "battery heating while driving" at −40 °C in US06 driving cycle [100] (Reprinted with permission from Elsevier © 2017); and (*f*) and (*g*) cell temperature and SOC during rapid heating and then fast charging at −40 °C [101] (Reused under the PNAS license).

temperatures can be used for battery self-heating, as shown in Figs. 13(*d*) and 13(*e*). This "battery heating while driving" strategy not only makes use of regenerative breaking energy that would otherwise be wasted but also eliminates the need of waiting time for preheating. As a result, it can dramatically extend driving range at low temperatures, e.g., by ∼50% at −40 °C [100]. Moreover, the SHLB structure and principle can be used prior to fast charging at low temperatures [101] as the energy for heating comes from an external charger. Figures 13(*f*) and 13(*g*) showed that a Li-ion cell was rapidly heated from −40 °C and then charged from 0% to 80% SOC, with a total time of 15 min.

7.2 Asymmetric Temperature Modulation for Fast Charging of Energy-Dense Li-Ion Batteries.

Building on the SHLB concept, Wang's group [103,104] further developed the asymmetric temperature modulation (ATM) concept for extreme fast charging of energy-dense Li-ion batteries. Extreme fast charging capability and high energy density are competing factors for conventional Li-ion batteries, so they must be compromised. Li-ion cells with the ATM concept avoided this compromise. As schematically shown in Fig. 14(*a*), a Li-ion cell with high energy density is rapidly preheated to and charged at high temperature (60 °C or 65 °C), which dramatically enhances kinetics and transport and hence eliminates Li plating. After fast charging, the cell will cool to ambient temperature, reducing SEI growth. It was demonstrated that an energy-dense cell (209 Wh/kg) retains 91.7% capacity after 2500 cycles of 10-min extreme fast charging, far exceeding the U.S. Department of Energy (DOE) target, as shown in Fig. 14(*b*). More recently, the ATM concept was combined with a thermally stable dual-salt electrolyte ($LiPF_6$–LiFSI) and higher anode porosity, enabling extreme fast charging of Li-ion cells with further higher specific energy, 265 Wh/kg, while maintaining excellent cycle life as shown in Figs. 14(*c*)–14(*e*). Furthermore, electrochemical-thermal coupled simulation suggests that passive air cooling would be sufficient for such cells in EV applications by making use of their high stability temperature and the high thermal conductivity along the in-plane direction of electrode assembly, as shown in Fig. 15. Such air-based cooling and ATM-based self-heating would dramatically simplify the thermal management system as compared to liquid cooling system dominant in state-of-the-art EVs.

7.3 Energy-Dense Battery for Durable and Safe Operation at High Temperatures.

To address the challenge of degradation and safety failures at high temperatures, Wang's group [105,106] developed a heat-tolerant battery, also known as safe, energy dense battery (SEB), by passivating a Li-ion cell. As schematically shown in Fig. 16(*a*), an enhanced SEI layer and a cathode-electrolyte-interface layer are formed by adding a small amount of triallyl phosphate in conventional electrolytes. The enhanced SEI layer and cathode-electrolyte-interface layer increased resistance of such passivated cells by ∼5 times, as shown in Fig. 16(*b*), thereby ensuring high safety and thermal stability when the cells are not in use (majority of the cells life). When high power and thus low resistance is needed upon operation, the cells can be self-heated to an elevated temperature such as 60 °C within tens of seconds using the SHLB method described above. As shown in Figs. 16(*c*)–16(*e*), the SEB cells were demonstrated to have superior cycling stability at 60 °C and safety during nail penetration tests as compared with baseline Li-ion cells.

Similar heat-tolerant battery designs can use graphite anode materials of low surface area or large particles [12], thermally stable salts such as LiFSI [104], and lower-voltage cathode operation [107] to provide exceptional stability of Li-ion cells operated at 65–70 °C or higher with no gas generation and little impedance growth. It appears that a "hot" paradigm of Li-ion batteries tolerant to heat and resistant to thermal runaway has emerged.

7.4 Smart Batteries With Sensing and Early Detection.

Since temperature distributions and reactions are nonuniform in Li-ion cells under extreme conditions [16,23,37], as discussed earlier, it is helpful for Li-ion cells to be equipped with interior sensors for early detection of abnormal behaviors. As shown in

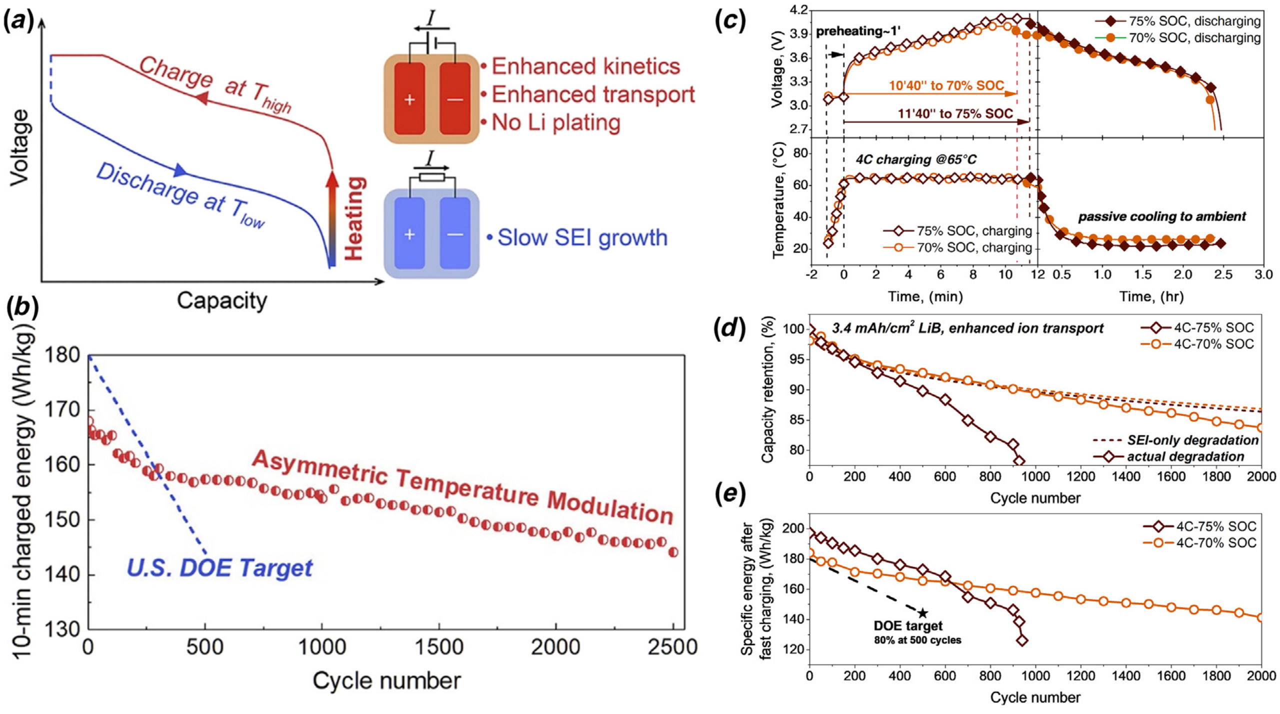

Fig. 14 ATM for fast charging of energy-dense Li-ion batteries. (*a*) Schematic of ATM cell operation [103] (Reprinted with permission from Elsevier © 2019); (*b*)–(*d*) fast charging of energy-dense (265 Wh/kg) Li-ion cells with thermally stable dual-salt electrolyte [104] (Reprinted with permission from Springer Nature © 2022).

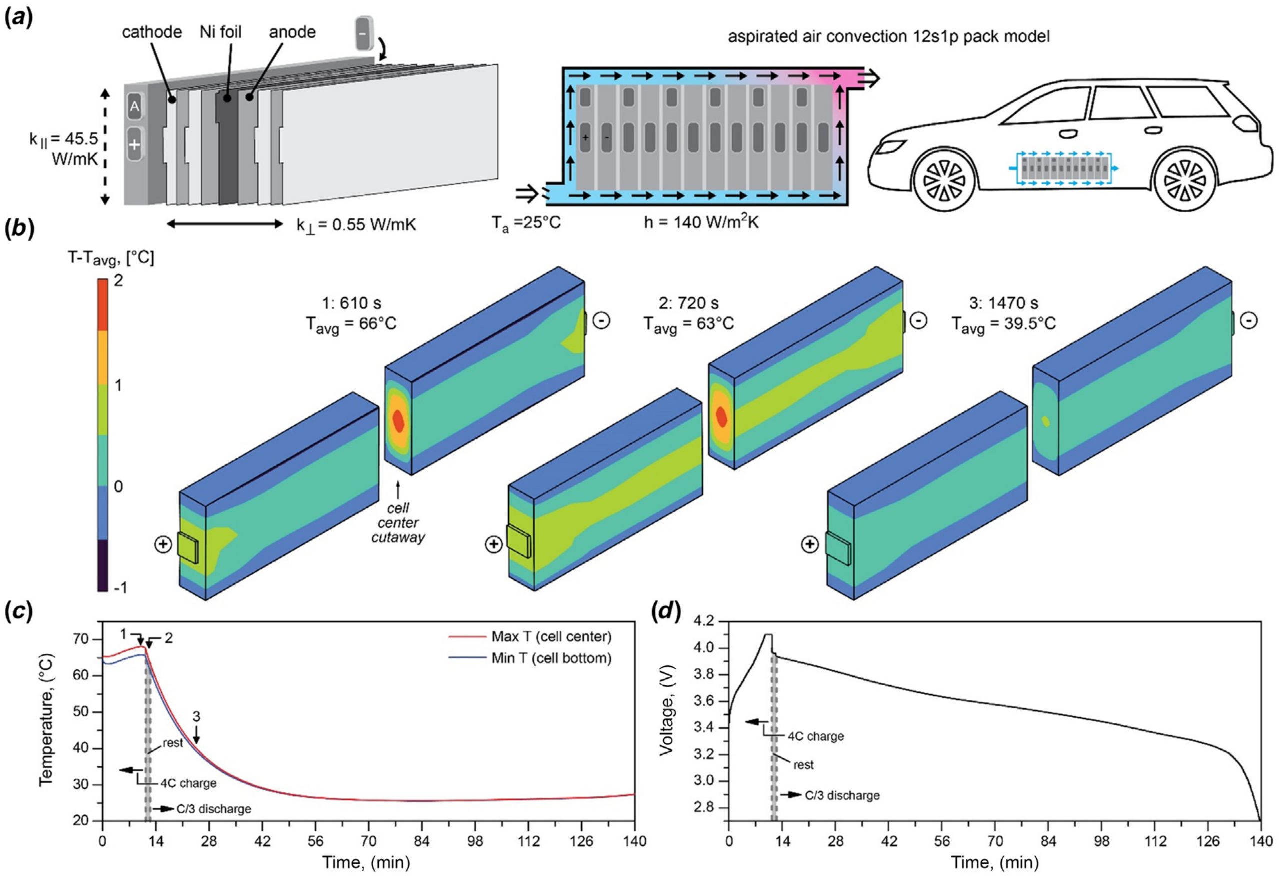

Fig. 15 Electrochemical–thermal coupled simulations of a 12S1P pack of 150 Ah prismatic cells with ATM and thermally stable electrolyte. (*a*) Cell construction, pack model and thermal conditions under aspirated air convection; (*b*) 3D temperature difference contours in 150 Ah prismatic cells at three representative time instants during 4 C charge–C/3 discharge cycling; (*c*) evolution of maximum and minimum temperatures in the prismatic cell; and (*d*) cell voltage evolution during cycling [104] (Reprinted with permission from Springer Nature © 2022).

Figs. 17(*a*) and 17(*b*), Zhang et al. [108] first demonstrated this concept by embedding a temperature sensor in a cylindrical Li-ion cell. The internal temperature sensor, labeled as reaction temperature sensor (RTS) in the schematic, could detect abnormal temperature rise much faster and more accurately than the surface temperature sensor. As a result, the internal-temperature-sensor-based control enables early detection and termination of a short-circuit event and prevents cell overheating. This work of embedding temperature sensors has started an active pursuit of smart batteries around the world recently [109–113], including using strain sensors or gas sensors, and using wireless sensors to eliminate the inconvenience of wire connection [109,110] as shown schematically in Fig. 17(*c*). However, wireless sensors in reported studies are still too big in size, and their durability in Li-ion cell environments remains to be seen [109,110]. Further efforts are still needed in developing smart cells with embedded microsensors that are durable, low cost, and accurate. The demonstration of smart cells in large modules or packs is also needed.

7.5 Smart Batteries With Multifunctional Components for Enhanced Safety. In addition to embedding sensors for early detection of abnormal behaviors, smart batteries can also be made by using multifunctional components. For example, as shown in Fig. 18, Wu et al. [114] reported a smart separator that sandwiches a thin conducting layer. Such a smart separator works not only as a separator but also as a sensor for early detection of lithium dendrite growth that can cause internal short circuit. Another example is the nickel foil in the SHLB cell described above. As shown in Fig. 19, the nickel foil used for rapid heating can also be used as a temperature sensor due to the dependence of foil resistance on temperature [99].

More recently, smart current collectors that can easily break to mitigate internal short circuit during mechanical impact [115–117] or overheating [117,118] to prevent thermal runaway are actively pursued. Figure 20(*a*) shows schematically a notched current collector design by Wang et al. [115] that can easily break along the notches during mechanical impact so that the impacted region can be detached from the main region of the cell. Figure 20(*b*) shows schematically the predetermined breakable pattern in battery electrodes by Naguib et al. [116]. In this design both the current collector and electrode are partially slitted. The mechanically impacted part of the battery will be separated upon prescribed deformation from the rest of the battery. Figure 20(*c*) shows schematically a metalized current collector by Pham et al. [117] that will retreat from internal short circuit caused by nail penetration. Figure 20(*d*) shows schematically a copper-coated-polyimide-based current collector embedded with triphenyl phosphate flame retardant by Ye et al. [118]. This design aims to simultaneously minimize the "dead weight" within the cell and improves fire safety. The recent studies by Zhang's group [36,37] on internal short circuit and thermal runaway supported the working mechanism of these multifunctional current collectors, and further revealed that it is the aluminum current collector that matters most. Note that these smart current collectors are mostly tested in coin cells or small pouch cells except the study by Pham et al. in which 18,650-formate cells are made [117]. In addition, the effects of current collector modification on large-format cell manufacturing, power performance, and durability need to be further investigated.

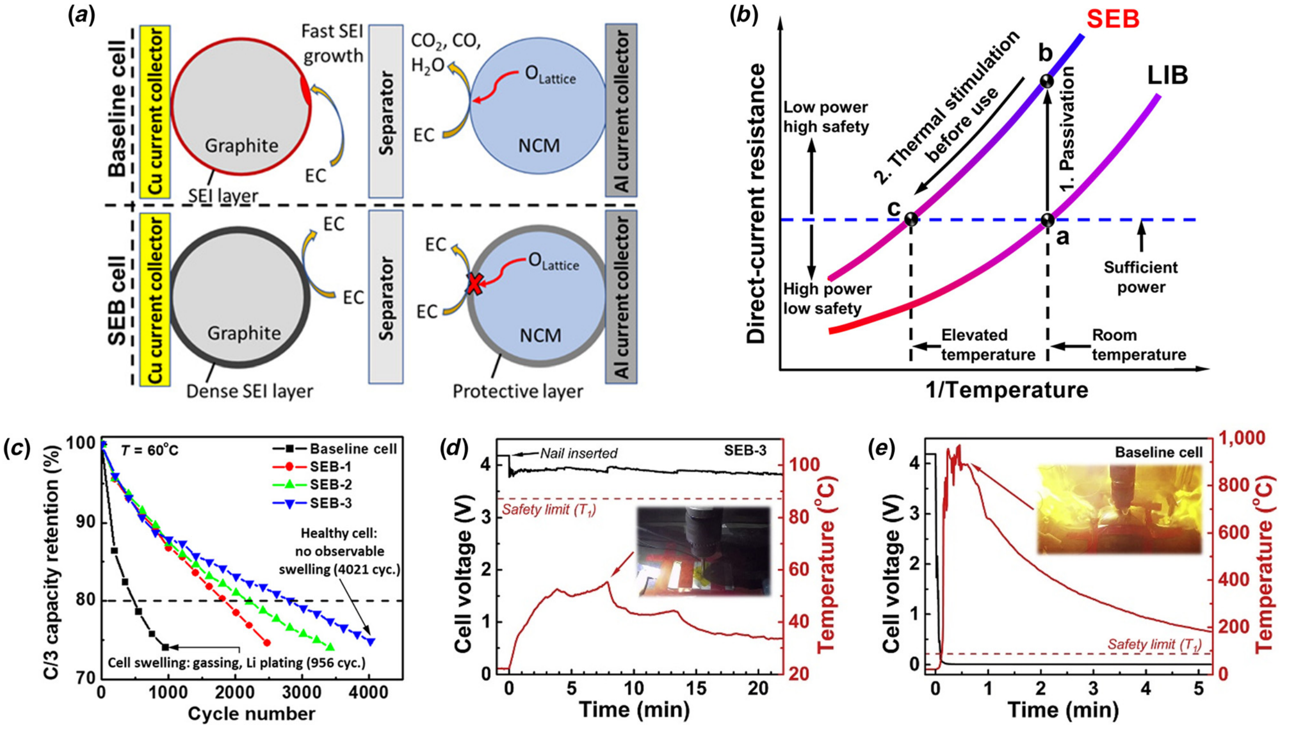

Fig. 16 Principle and advantages of a SEB cell versus a conventional LIB cell. (*a*) Schematic showing enhanced interfacial layers. (*b*) DC resistance varies with the inverse of temperature, where the upper curve for the SEB is always safer due to higher DC resistance. The SEB can, however, achieve a similar power output to the LIB by thermal stimulation before operation, shown as going from point b to c. (*c*) Comparison of cycling stability at 60 °C [105]. (Reused under license CC BY-NC 4.0). (*d*) and (*e*) Comparison of cell voltage and temperature during nail penetration tests [106]. (Reused under license CC BY-NC-ND 4.0).

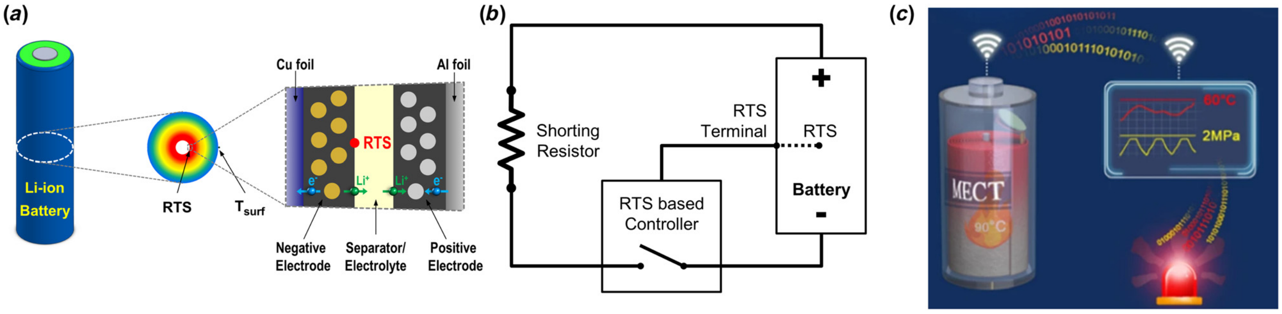

Fig. 17 Smart cells with embedded sensors. (*a*) and (*b*) Schematic of a smart cell with embedded internal temperature sensor for enhanced safety [108]; (*c*) schematic of a smart cell with embedded wireless sensors [109]. (Reused under license CC BY 4.0).

7.6 Current Collector/Tab Cooled Li-Ion Cells. As discussed in Sec. 3.4, the through-plane direction thermal conductivity of Li-ion cells is low, typically less than 1 W m^{-1} K^{-1}, which can cause nonuniform temperature distribution through the thickness of the cells under extreme conditions. It is also noted that the in-plane direction thermal conductivity is much higher, about 20–40 W m^{-1} K^{-1} [41,46], mainly due to the high thermal conductivity of Cu foil and Al foil current collectors. Therefore, making use of the in-plane direction heat transfer could enhance the uniformity of temperature distribution. One approach is to widen cell tabs and use current collector/tab-based cooling instead of surface cooling [119]. A simulation on pouch cells [120] shows that both temperature distribution and current distribution in a tab-cooled cell are more uniform than that in a conventional surface-cooled cell (Figs. 21(*a*) and 21(*b*)). However, the simulation also shows that the average temperature can increase more in a tab-cooled cell than that in a surface-cooled cell due to the limited heat transfer area of tab cooling even with a wide tab. More recent simulations (Fig. 21(*c*)) on cylindrical cells [121,122] show that tab cooling with a continuous-tab design, also referred to as all-tab design or tables design [123], could not only enhance uniformity of temperature distribution and current distribution but also reduce heat generation and average temperature rise. This simulation is consistent with earlier simulations on the effects of tab designs [124,125]. Moreover, cells with a continuous tab design could be made large and integrated with the cell-to-pack structure based on both cylindrical cells [126] and prismatic cells [104], instead of cell-to-module-to-pack structure. Such cell-to-pack structure could significantly enhance the cell-to-pack ratio and thus is actively pursued by the EV and battery industry [12,126,127]. However, the manufacturing of wide-tab pouch cells and continuous-tab cylindrical cells is more complicated than conventional designs. Further investigations are still needed.

8 Future Research

Great efforts are still needed to enable safe, reliable, and efficient operation of EV batteries under various extreme conditions.

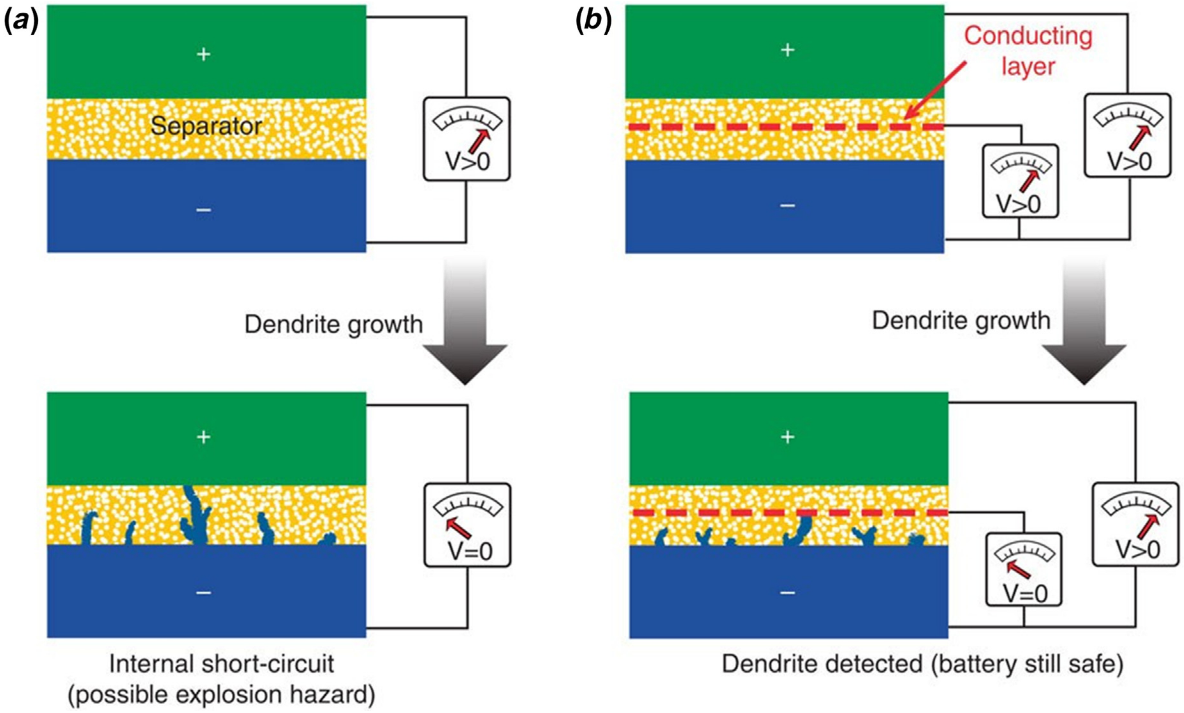

Fig. 18 Smart cells with a bifunctional separator for early detection of lithium dendrite growth [114] (Reprinted with permission from Springer Nature © 2014)

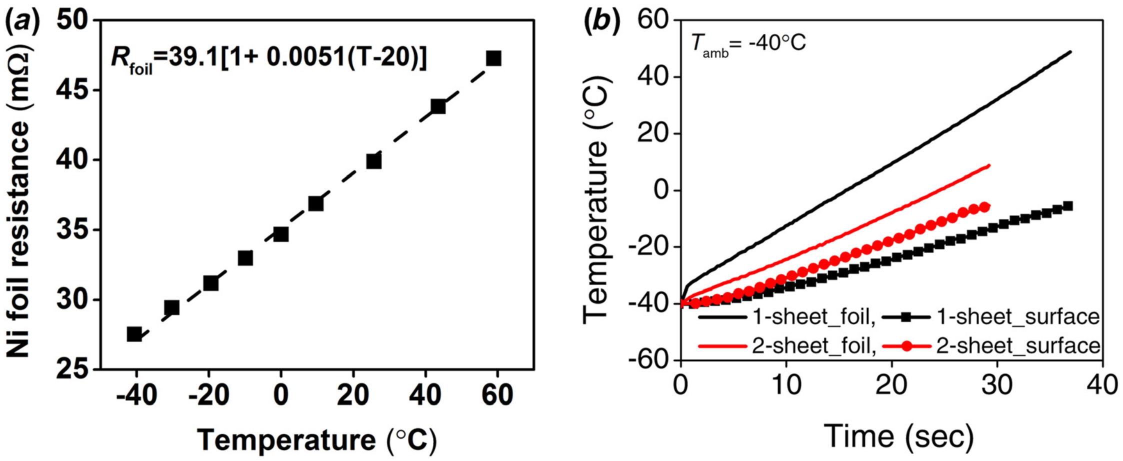

Fig. 19 Bifunctional use of the nickel foil in SHLB cell as both a heater and temperature sensor. (*a*) Dependence of the foil resistance on temperature and (*b*) measured foil temperature and surface temperature with different SHLB designs [99] (Reprinted with permission from Elsevier © 2016).

In addition to the extreme conditions discussed in this review, scenarios special to EVs are emerging, such as vehicle-to-grid (V2G) and vehicle-to-vehicle (V2V) connection [128], structural batteries that are part of vehicle body [129], and battery recycling [130]. Above all, EVs must be more affordable and energy efficient. These emerging scenarios will create additional challenges and opportunities for battery thermal management. For example, the V2G and V2V scenarios require bidirectional (two-way) charging of EVs, which means much more frequent charging and discharging of batteries than conventional one-way charging. In such scenarios, durability and fast charging capability of batteries, and therefore thermal management to enable them, become even more important. For structural batteries, mechanical strength of batteries needs to be greatly enhanced, and the shape needs to be more flexible. These additional requirements would make complicated active thermal management more difficult. Simpler and passive thermal management based on the concepts of heat-tolerant batteries and smart batteries would be more desirable. Battery recycling [130] has become increasingly important due to the growth of retired EVs and the limitation of battery raw materials. But sophisticated designs of cells, modules, and packs make battery recycling difficult and costly. To enable more sustainable development, battery recycling must be taken into consideration in future designs of EV batteries. For example, simpler cell and pack structures [130] and simpler thermal management systems (e.g., air-based) or management-free designs will facilitate battery recycling. Finally, reducing the cost of battery and operation is important. An often overlooked approach is to use smaller packs with fast charging capability and low-cost, air-based thermal management [104]. Table 4 summarizes some of these challenges and potential strategies to inspire further discussion and investigation. The challenges already discussed in this review are also included.

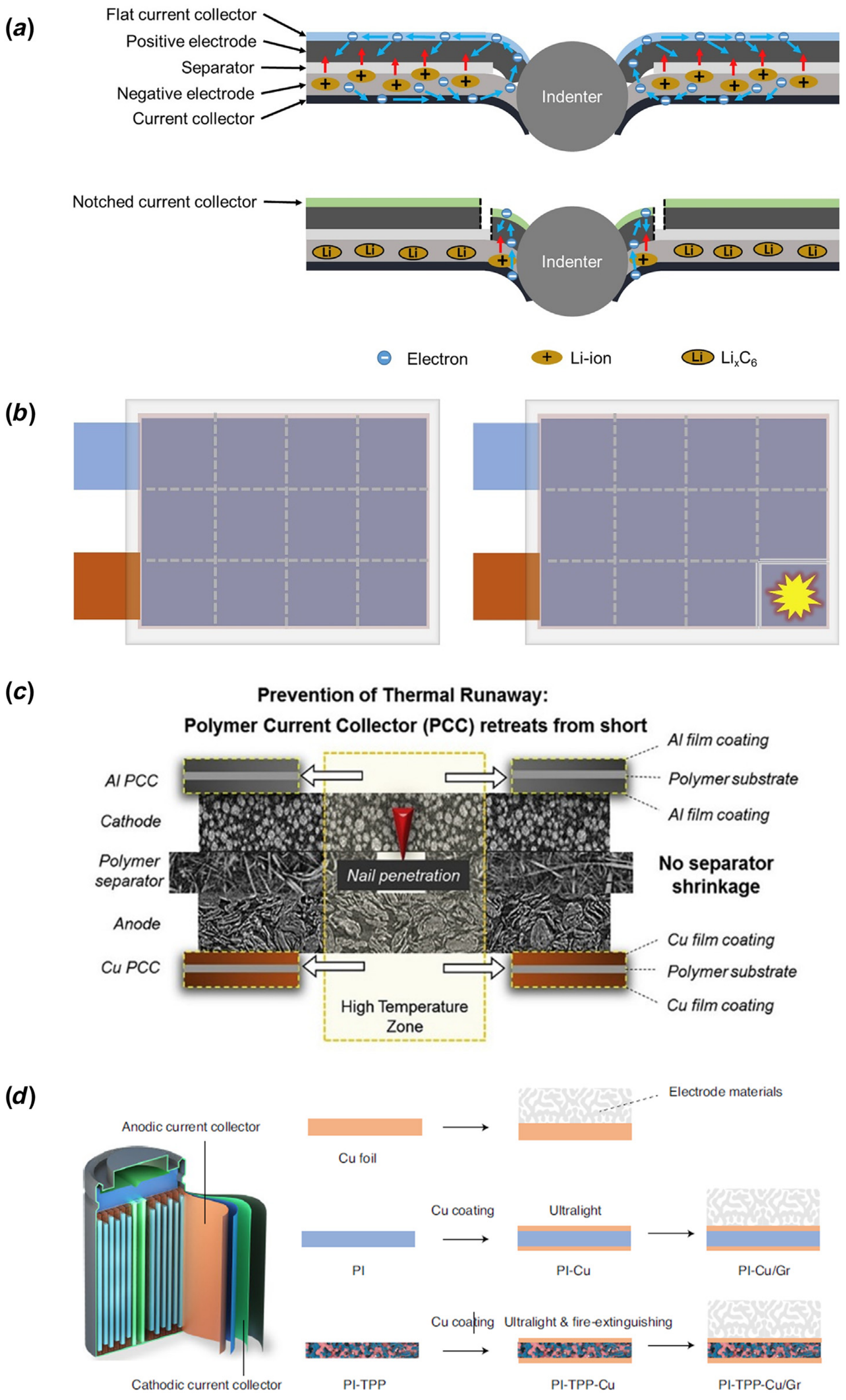

Fig. 20 Schematics of smart cells with multifunctional current collectors. (*a*) A notched current collector easily breaks upon mechanical impact [115] (Reprinted with permission from Elsevier © 2017). (*b*) Battery electrodes with predetermined breakable pattern [116] (Reprinted with permission from Elsevier © 2018). (*c*) Metalized polymer current collector [117] (Reused under license CC BY 4.0). (*d*) Metalized polymer current collector with embedded flame retardant [118] (Reprinted with permission from Springer Nature © 2020).

In order to address the comprehensive and mostly competing demands of EV batteries under extreme conditions and emerging scenarios, it is necessary to take an integrated approach from both the perspective of developing better thermal management systems and the perspective of designing novel structured batteries. Therefore, the information exchange and collaboration among different research communities, e.g., heat transfer community, materials community, battery community, and EV community, becomes critical.

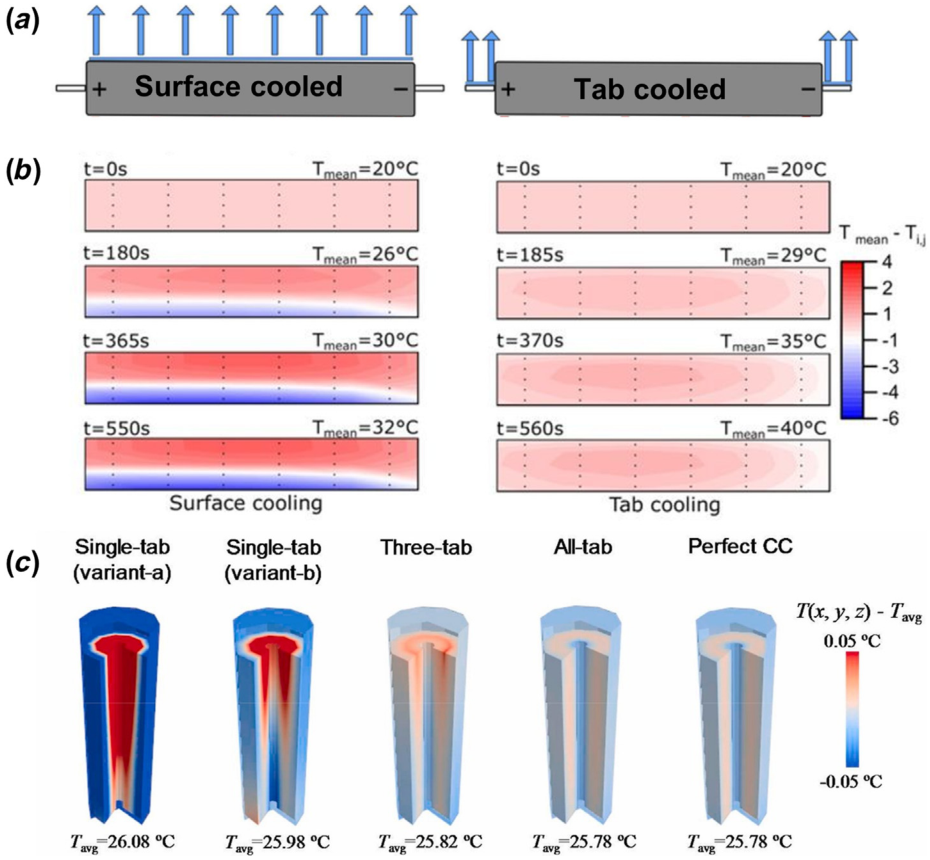

Fig. 21 **Tab-cooled Li-ion cells with enhanced uniformity of temperature distribution. (*a*) and (*b*) Schematic of surface-cooled and tab-cooled pouch cells and simulated temperature distributions [120] (Reused under license CC BY 4.0). (*c*) Simulated temperature distributions in cylindrical cells with different tab designs after the first 60 s of 1 C discharge [121] (Permission to reprint from Elsevier © 2021).**

Table 4 **A brief summary of challenges and innovative strategies for EV batteries under extreme conditions and emerging scenarios**

Conditions/scenarios	Challenges for batteries	Examples of innovative strategies
Low temperature	Poor performance, lithium plating, degradation, efficiency	Self-heating battery
High temperature	Degradation, safety, efficiency	Heat-tolerant battery; novel external cooling
Fast charging	Lithium plating, overheating, degradation, safety, efficiency	Asymmetric temperature modulation; heat-tolerant battery
Thermal runaway	Spontaneous fire due to internal short circuit, thermal runaway propagation, stranded energy	Heat-tolerant battery; smart battery with embedded sensors and multifunctional components; emergency cooling; thermal switch
V2X (V2V, V2G…)	Degradation, fast charging, smart communication	Heat-tolerant battery; smart battery with embedded sensors
Structural battery	Battery strength, flexible shape, thermal management	Cell-to-pack structure; all-tab design with current collector/tab-based cooling; passive thermal management
Battery recycling	Sorting, disassembly, safety, cost	Simple pack structure (e.g., cell-to-pack); simple thermal management (e.g., air-based)
Affordability	Cost of battery pack, cost of operation	Smaller packs with fast charging; low-cost thermal management (e.g., air cooling, passive cooling)

9 Conclusions

With relentless pursuit of transport decarbonization, EVs are increasingly used under extreme conditions such as low temperature, high temperature, and fast charging. As dominant power sources for EVs, Li-ion batteries face various challenges under these extreme conditions. Moreover, spontaneous fire has become a pressing issue for EVs which can be attributed to internal short circuit of Li-ion batteries. Thermal management plays a critical role in addressing these challenges due to the interactions between thermal, electrochemical, and chemical processes in Li-ion batteries. However, due to the competing requirements of EV batteries, simply improving current thermal management systems is not enough to address the dilemmas faced by conventional Li-ion batteries. Instead, novel battery designs capable of intracell thermoregulation could reduce or even eliminate these dilemmas, thus greatly simplifying battery thermal management under extreme conditions. To address grand challenges and cross-fertilize innovations, it is necessary to integrate efforts from both the perspective of improving thermal management systems and the perspective of designing novel structured batteries from cells to packs.

Data Availability Statement

The datasets generated and supporting the findings of this article are obtainable from the corresponding author upon reasonable request.

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