

Recent advances of thermal safety of lithium ion battery for energy storage

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ARTICLE INFO

Keywords:

Thermal safety
Lithium ion battery
Energy storage

ABSTRACT

Lithium ion batteries have been widely used in the power-driven system and energy storage system. While thermal safety for lithium ion battery has been constantly concerned all over the world due to the thermal runaway problems occurred in recent years. Lithium ion battery has high temperature sensitivity and the relatively narrow operating temperature range because of the complex electrochemical reactions at different temperatures. And the temperature change, including the global temperature change in different seasons and the local temperature rise that is induced by its self-heating etc., can trigger side reactions and then lead to thermal runaway, which should be further considered to ensure thermal safety of lithium ion battery. This review summarizes the inducements of thermal runaway and relevant solutions, spanning a wide temperature range. The low temperature induced issues, such as capacity fade and lithium plating and dendrite, can cause internal short circuit (ISC), while as the temperature is above the critical temperature, the speeding of side reactions and reduction of lifespan ($T > 40^\circ\text{C}$) and thermal runaway ($T > 90^\circ\text{C}$) will be triggered. In order to solve the thermal issues in batteries, extensive approaches have been investigated to prevent the occurrence, propagation and deterioration of thermal runaway, from the perspective of material to the battery system. The triggered mechanism at a wide temperature range, key factors for thermal safety and the effective heat dissipation strategies are concluded in this review. This review is expected to offer effective thermal safety strategies and promote the development of lithium ion battery with high-energy density.

1. Introduction

The shortage of fossil fuel is a serious problem all over the world. Hence, many technologies and methods are proposed to make the usage of renewable energy more effective, such as the material preparation for high-efficiency photovoltaic [1] and optimization of air foil [2]. There is another, and much simpler way to improve the utilization efficiency of renewable energy, i.e. the energy storage. The energy storage is more like the “agency” to mediate the relation between collection and utilization of renewable energy, removing the discontinuity in space and time. With the mediated property, the energy storage is adopted to peak shaving and valley filling for electric network [3,4], relieving the imbalance between supply and demand.

There are many types of energy that can be stored, such as the thermal

energy, electric energy, mechanical energy or chemical energy [5–8]. The most effective method of energy storage is using the battery, storing energy as electrochemical energy. The battery, especially the lithium-ion battery, is widely used in electrical vehicle, mobile phone, laptop, power grid and so on. However, there is a major problem in the application of lithium-ion battery. The battery generates heat during charge or discharge, increasing the battery temperature. Once the heat cannot be dissipated in time, the battery may get into thermal runaway. Large amount of heat will come out and the temperature could be increased to 200°C [9], resulting in the combustion or even explosion.

The fundamental method to solve the thermal runaway problem is to develop the electrochemical reaction (new battery material) to reduce heat generation or improve thermal stability of function materials of battery [10]. While, restricted by the necessary development process,

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thermal issues cannot be solved easily in the prospective of material, hence, another effective way should be further developed to ensure thermal safety of lithium ion battery, i.e. effective battery thermal management (BTM) strategies. There are many papers to review the progress of BTM [11–13]. However, the fundamental mechanisms from material to system aren't developed up to now. Consequently, this paper is to review the research progress of battery safety, including heat generation and thermal management from perspective of material to system. This review is expected to offer a series of effective approaches to solve thermal runaway problems, leading to a safe, efficient power-driven system and energy storage system.

2. Thermal hazard for lithium ion battery

2.1. Inducements of thermal hazard

Thermal runaway of batteries is the primary thermal hazard for electric vehicles and battery energy storage system, which is concerned by researchers all over the world. In general, the primary abuse conditions for thermal runaway include mechanical abuse, electrical abuse, thermal abuse etc., which may induce ISC in batteries and cause rapid temperature rise and then is thermal runaway [14]. While the thermal runaway can still occur without ISC at high temperature because of the chemical crosstalk between cathode and anode materials as the

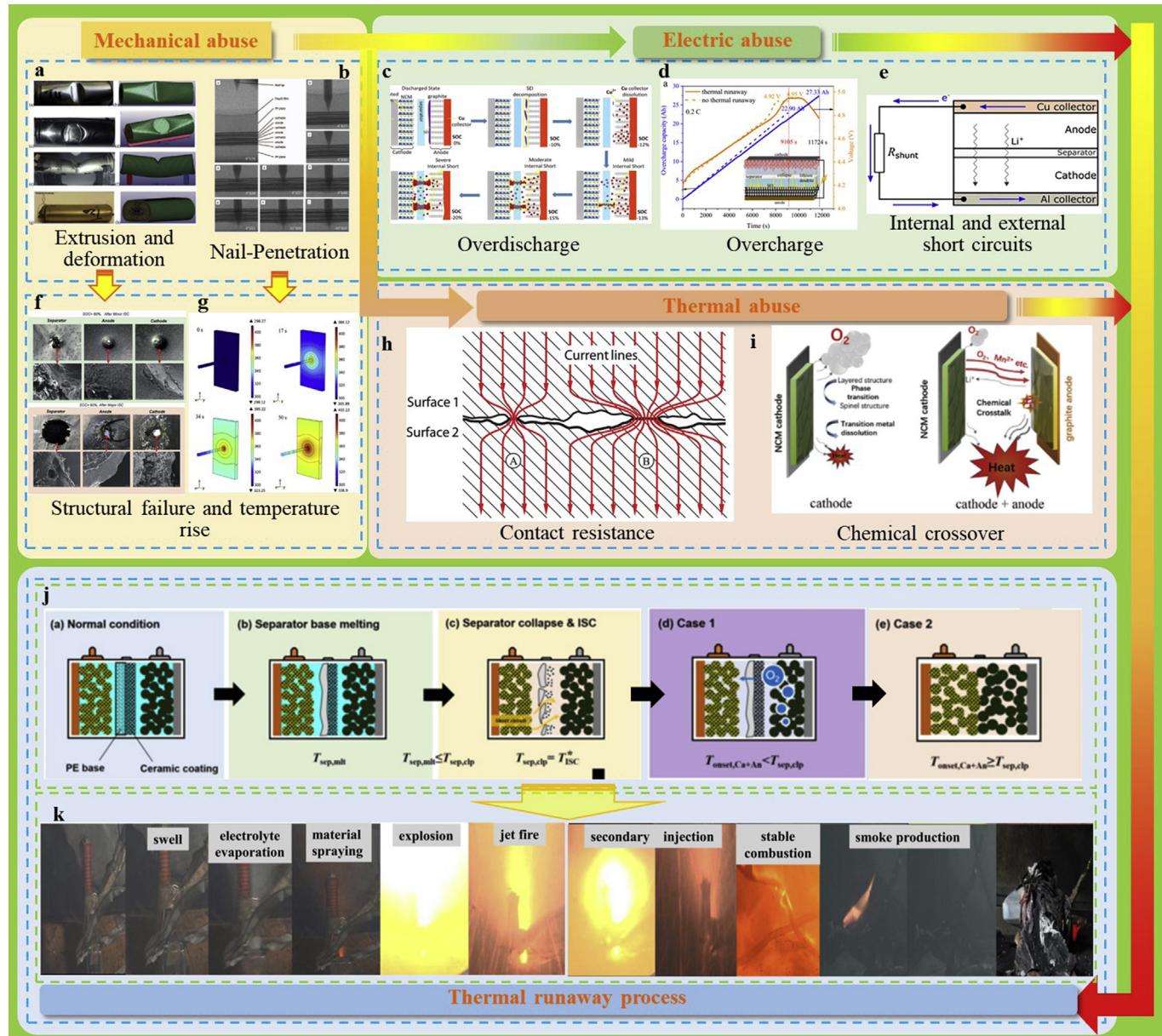


Fig. 1. The abuse conditions and induced thermal runaway process. **a, b**, mechanical abuse conditions in cell. **a** shows cells in quasi-static loading conditions with four test protocols (Rod (a–a), circular punch (a–c), three-point bend (a–e) and flat plate (a–g)). **b** shows nail-penetration in cells. **c–e**, The electric abuse conditions in cells. Overdischarge (**c**), overcharge (**d**), internal and external short circuit (**e**). **f, g**, The structure failure and temperature rise. **f** shows the local internal short circuit triggered by crush. **g** shows the internal short circuit and thermal runaway induced by nail-penetration. **h, i**, The thermal abuse conditions in cells. **h** shows contact resistance for heat generation. **i** shows the thermal runaway without internal short circuit. **j, k**, The thermal runaway process and caused by abuse conditions. Internal structural evolution during temperature rise process (**j**). Combustion process (**k**). Panels reproduced with permission from: **a**, ref. [16] (Elsevier); **b**, ref. [20] (Elsevier); **c, k**, ref. [23] (The Electrochemical Society); **d**, ref. [34] (Nature Research); **e**, ref. [28] (Elsevier); **f**, ref. [36] (RCS); **g**, ref. [19] (Elsevier); **h**, ref. [37] (Elsevier); **i**, ref. [15] (Cell Press); **j**, ref. [18] (ECS).

temperature inside battery is higher enough [15].

Fig. 1 shows the abuse conditions and induced thermal runaway process. As shown in **Fig. 1a** and b, mechanical abuse is mainly caused by mechanical force and commonly occurred in traffic accident, including collision, crush and penetration etc. When the battery or battery pack is collided or crushed by mechanical force, potential damages inside batteries may occur, including rupture of separator and the leakage of flammable electrolyte, causing ISC and then resulting in thermal runaway (**Fig. 1f** and g) [16–18]. While the penetration for lithium ion battery can pierce the positive/negative electrodes and separators and then result in ISC [19,20]. In nail-penetration process, the shapes, size, displacement of nails, nail tip and nail motion all have impacts on ISC process, temperature rise and distribution, as well as the state of charge in lithium ion battery [21,22].

Fig. 1c, d and **1e** shows the electrical abuse conditions that induce the thermal runaway in lithium ion battery. Electrical abuse mainly includes external short circuit (ESC), overcharge and overdischarge [23] etc. The ESC can result in large current and high heat generation in battery, which is primarily caused by ohmic heat generation [24–26]. As the temperature reached up to a highly level (77 °C–121 °C), the cell rupture, venting and electrolyte leakage may be triggered [27,28]. Overcharge process can be divided into three stages. (1) In the first stage, voltage and temperature have no obvious changes. (2) In the second stage, the lithium dendrite deposition will occur at the voltage platform. (3) And in the third stage, the voltage will drop dramatically with high heat generation and gas generation, causing thermal runaway in battery [23,29]. The reconstruction of cathode materials can arise massive heat generation and oxygen release. And the released oxygen can react with electrolyte and cause more side reactions. The decomposition of electrolyte can release much more gas and result in venting of the battery, and then lead to fierce heat generation after venting. Meanwhile, the morphology, structure and thermal stability of cathode and anode become worse with the increase of SOC [29,30]. High current in overcharge process may induce explosion, and there is a relationship between the duration of voltage platform and overcharge rate [23]. Overcharge in lithium ion battery can also induce the growth of Lithium dendrite in anode and pierce the separator, leading to ISC in lithium ion battery [31]. The overdischarge may cause the capacity fade and decomposition of solid electrolyte interphase (SEI), leading to gas release [32]. Meanwhile, the overdischarge may result in the dissolution of copper, and the deposition of the copper can form a copper dendrites, leading to ISC inside the battery [33,34].

Thermal abuse is mainly resulted by overheating, which is shown in **Fig. 1h** and i. The overheating in lithium ion battery may be caused by mechanical abuse, electrical abuse and contact loss of the connector. The contact loss of connector may induce the increase of resistance in battery pack, further leading to local overheat. The insufficient pre-tightening leads to significant increment of contact resistance with massive battery energy loss as heat generation at the interface. Meanwhile, the thermal induced thermal runaway also accompanied with heat generation [35]. The abuse conditions finally result in ISC and cause temperature rise and thermal runaway. Besides, the released oxygen from cathode can react with reductive LiCx accompanied with highly heat generation, which means that the chemical crossover is the potential mechanism of thermal runaway when there is no severe ISC happened [15].

As shown in **Fig. 1j** and k, mechanical, electric and thermal abuse conditions may finally induce the continuous heat generation and temperature rise inside battery, and a series of chain reactions may occur in different stages with the increasing temperature (**Fig. 1j**) [18]. Besides, in thermal runaway process, a series of thermal induced behaviors can be observed by experimental methods. As shown in **Fig. 1k**, the internal high temperature and pressure that is caused by side reactions and electrolyte evaporation may lead to spraying, explosion and jet fire accompanied with gases release [23]. And the jet and injection can cause the propagation of thermal runaway in battery, leading to continuous deterioration

of thermal hazard.

2.2. Temperature sequence map for thermal hazard

Fig. 2 shows a temperature sequence map to demonstrate the potential inducements at different working temperatures. Considering the real application of lithium ion batteries at different temperatures, there are different inducements for thermal runaway and the work temperature range of lithium ion battery is limited. According to different work states, the temperature sequence map can be divided into four states:

- (1) In **State I**, the working temperature is lower than 0 °C. In this situation, the electrochemical reactions will decrease dramatically, and the capacity, rate performance etc. can be further limited. Besides, Li plating and dendrite can be easily triggered during charging process, because of the lithiation reaction of graphite on anode, which can penetrate the separator and then induce ISC inside lithium ion battery [38].
- (2) In **State II**, lithium ion battery is in normal state in this state, and the heating rate is commonly lower than 1 °C/min and the critical temperature is about 90 °C. In this stage, the total heat generation is caused by reversible heat generation (electrochemical reaction), irreversible heat generation (ohmic heat generation and polarized heat generation) and mixed heat generation (heat generation by side reactions). As the temperature is higher than 40 °C, the lifespan of lithium ion battery may shorten due to the speeding side reactions, and when the temperature is near 90 °C or even higher, the decomposition of SEI film can be triggered, which is defined as the beginning of thermal runaway.
- (3) In **State III**, the heating rate is higher than 1 °C/min but no more than 10 °C/min. In this state, side reactions are highly exothermic, mainly including decomposition of SEI film, Li/organic solvents reactions, separator melting, decomposition of cathode materials and so forth.
- (4) In **State IV**, the heating rate is higher than 10 °C/min, and in this state, chemical crosstalk between two electrodes occurs inside battery, the electrolyte starts to burn and even explode [39,40].

In **State I** and **State II**, heat generation is quite small compared to thermal runaway process. As shown in **Fig. 2b**, the heat generation at normal state is mainly caused by Li + diffusion in solid and liquid phase, electrochemical reactions in solid-liquid interface and side reactions. The heat generation cause temperature rise and difference inside the battery, which may influence the life and safety of lithium ion battery. And the aging process at high current has an effect on cell safety, while the lithium plating on anode due to low temperature charging shows a significant increase of self-heating at low temperatures [41].

For **State III** and **State IV**, the primary heat generation that causes thermal runaway inside lithium ion battery mainly occur after the onset of overheating. The chain reactions and energy release in thermal runaway process are shown in **Fig. 2**. In fact, for different battery chemistries, the specific exothermic reactions are different, like the reported work by Zheng and Feng et al. [43]. Although the different battery chemistries have different exothermic reactions, the basic processes and reactions, such as decomposition of SEI film, melting of separator, decomposition of cathode materials and side reactions in electrolyte can be occur in the common thermal runaway process. As the temperature is higher than ~90 °C, the SEI decomposition will occur, releasing flammable gases and oxygen. The decomposition of SEI is an exothermic reaction and lead to higher temperature rise inside lithium ion battery. Meanwhile, the onset of overheating has a correlation with the thermal stability of SEI, and the thermal stability of SEI is related to the cycling temperature and aging effect [44,45]. When the temperature is high enough, the reactions between anode and electrolyte will occur. In anode, the intercalated lithium will react with organic solvents accompanied with the releasing of flammable gases, leading to highly

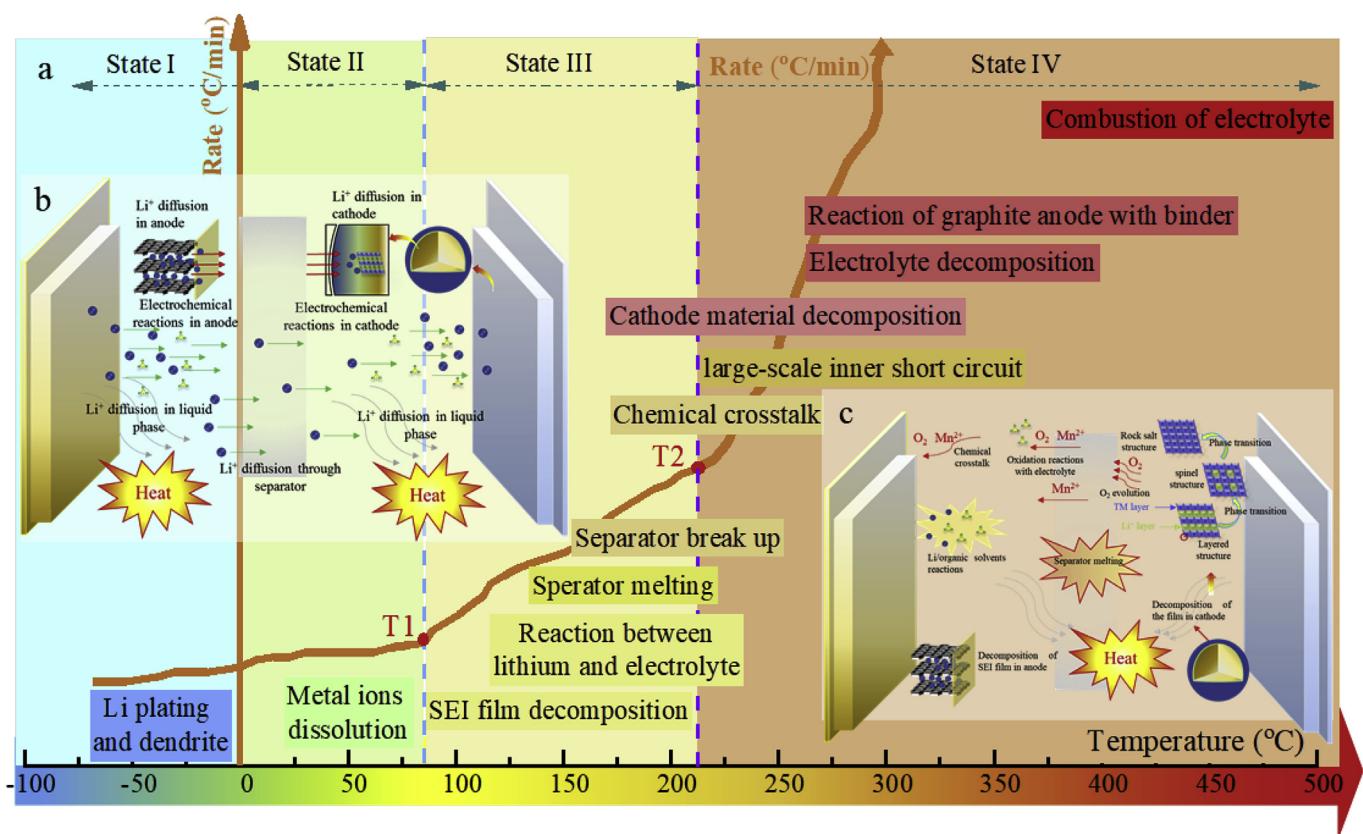


Fig. 2. Main heat generation process in lithium ion battery. **a** shows the exothermic chemical reactions inside cell with increasing temperature. **b, c**, The main electrochemical process inside cell at different temperatures. The electrochemical process and exothermic chemical reactions at normal state (**b**). The electrochemical process and exothermic chemical reactions at high temperature (**c**). Panels reproduced with permission from: **a**, ref. [15] (Cell Press) and ref. [42] (Elsevier).

exothermic and temperature rise. The polyethylene (PE)/polypropylene (PP) separator will melt as the temperature higher than ~ 130 °C, causing ISC and leading to more serious abuse condition. Although the melting of PE/PP separator is a heat adsorption process, the ISC caused by separator melt will further deteriorate the thermal runaway process. The ISC will cause highly exothermic, and as the temperature is higher than ~ 180 °C, decomposition of cathode in lithium ion battery may occur, accompanied with highly heat generation and significant oxygen release. The heat accumulation and gases release (oxygen and flammable gases) will then induce combustion and explosion of lithium ion battery. In thermal runaway process, heat generation caused by ISC is only 2%, and chemical reactions are 98%, including the decomposition of SEI layer, decomposition of electrolyte, reaction of graphite with binder decomposition of cathode materials etc [18].

3. Key factors of lithium ion battery for thermal safety in temperature sequence

Lithium ion batteries have different thermal issues at different ambient temperatures. Hence the internal factors for thermal safety in lithium ion battery deserve to be further reviewed. According to the temperature condition, the temperature sequence can be divided into 3 sections, including **State I**: low temperature (<0 °C), **State II**: normal temperature (0–90 °C) and **State III & IV**: high temperature (>90 °C).

3.1. State I: at low temperature (<0 °C)

Lithium ion battery cannot well work at low temperature because of the limited reaction rate. While the decelerated chemical reactions may induce the Li deposition, plating and dendrite, which is regarded as the main factors that induce the thermal runaway at low temperature. The

issues may cause ISC in the battery and then lead to thermal runaway (Fig. 3a) [41]. Besides, Schappacher et al. [46] reported that the deposition of lithium promotes the reduction and decomposition of carbonate based electrolyte, and the decomposition of electrolyte will produce volatile gaseous products during charge-discharge cycling at low temperature, which lowers the onset temperature of exothermic reactions and leads to an increase of self-heating, showing a different inducement of thermal runaway at low temperature. Ouyang et al. [38] proved that the interaction between plated lithium and electrolyte cause vigorous exothermic reaction (Fig. 3c). Love et al. [47] considered the mechanical deformation at low temperature can accelerate the metal plating and they emphasized the importance of thermo-mechano-electrochemical interplay for the safety of lithium ion battery at low temperature. Zhao et al. [48] investigated swelling failure of lithium ion battery at low temperature by using 2D/3D X-ray computed tomography, and they believed that the slow electrochemical kinetics at low temperature can induce Li dendrites, volume expansion and delamination of active materials, leading to swelling failure inside lithium ion battery (Fig. 3b).

As mentioned above, Li dendrite may penetrate separator and induce ISC and thermal runaway. Hence the detection, prevention of Li dendrite is necessary for thermal safety. Wu and Zhuo et al. [49] designed a bifunctional separator to detect the lithium dendrite inside battery by detecting the voltage change. The proposed advanced separator consists of three layers, including a conducting layer between two conventional separators. The growth of Li dendrite will penetrate the side conventional separator and contact the test layer, which can send a pronounced signal to alert the potential thermal risk that is caused by ISC. Detection is necessary for preventing the ISC and thermal runaway, and there is also other strategies to stop the growth of Li dendrite (Fig. 3d). Adding the functional additives into electrolyte is an effective method to prevent Li plating and dendrite. Zhang et al. [50] employed the p-Toluenesulfonyl

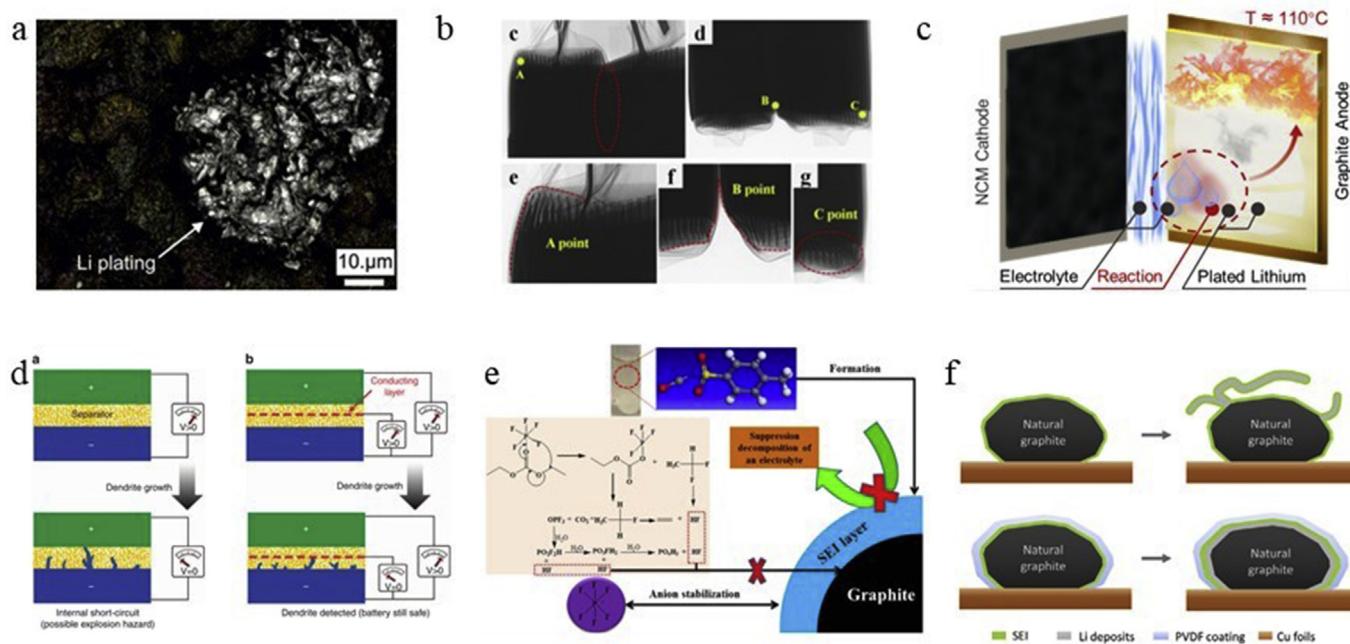


Fig. 3. Inducements of thermal runaway at low temperature. **a** shows the Li plating on the surface of electrode materials. **b**, The swelling failure at low temperature. **c**, Thermal runaway induce by Li plating. **d** shows the novel detection method for ISC. **e** shows the effect of electrolyte additive in lithium ion battery. **f**, coating strategy for preventing lithium dendrite. Panels reproduced with permission from: **a**, ref. [41] (Elsevier), **b**, ref. [48] (Elsevier), **c**, ref. [38] (ACS), **d**, ref. [49] (Nature Research), **e**, ref. [50] (Elsevier), **f**, ref. [51] (Elsevier).

isocyanate as electrolyte additive to reduce the decomposition of carbonate solvents and form a SEI film with more organic compounds and less inorganic compounds, preventing Li plating and dendrite. In addition, coating is another useful method to restrain the occurrence of Li dendrite (Fig. 3e). Wu et al. [51] applied high-polarity β -phase polyvinylidene difluoride as coating material to mitigate Li dendrite formation. The results showed that high-polarity β -phase polyvinylidene difluoride coated graphite exhibited smooth and dendrite-free Li plating and the charge-transfer resistance was reduced, which can effectively improve thermal safety and battery performance (Fig. 3f).

3.2. State II: at normal temperature (0~90 °C)

At normal state, extra heat generation is the main thermal issue, which may induce the beginning of thermal runaway. Fig. 4 shows the main heat generation and heat distribution at electrode. As shown in Fig. 4a, Li(Ni_xCoyMn_z)O₂ (NMC) shows a much higher heat generation rate, compared to that in different kinds of lithium ion battery i.e. LiMn₂O₄ (LMO), LiFePO₄ (LFP), LiCoO₂ (LCO) and NMC. In general, heat generation at normal state in lithium ion battery can be summarized as reversible heat generation, irreversible heat generation, and mixed heat generation. Besides, the state of charge and C-rate also have a different influences on the total heat, irreversible heat and reversible heat. For cathode materials, taking LCO as an example (Fig. 4b), heat generation in lithiation process (discharge) is higher than that in delithiation process (charging process), and, heat generation rate decrease gradually with the increasing state of charge. As known to us all, the heat generation rate increases with increasing C-rate. The irreversible heat is much higher than reversible heat. While for anode material (carbon), as shown in Fig. 4c, heat generation in deintercalation process (discharge) increase gradually with decreasing state of charge, and even higher with higher C-rate.

In intercalation process (charge), heat generation increase with increasing state of charge, and there is fluctuation in the process, not strict linear increase. Fig. 4d also demonstrates the different heat generation rates of different heat generation factors and discharge rates. At

normal state, for commercial lithium ion battery, cathode materials contribute the main heat generation inside the battery, which is much higher than that caused by anode and separator (Fig. 4e). In order to demonstrate the local heat generation and the distribution in cathode materials, Zhu et al. [58] employed a three dimensional cathode structure (Fig. 4j) for lithium ion battery. The calculation results are shown in Fig. 4f, g, 4h and 4i, which indicates the ion resistive heat in electrolyte (Fig. 4f), electronic resistive heat (Fig. 4g), reaction heat (Fig. 4h) and entropic heat (Fig. 4i) in cathode materials etc. The importance of ion resistive heat should be further considered and the potential heat generation calculated by porous-electrode model is underestimated. The different kinds of heat generation factors in electrode show that ion conductivity, electric conductivity, reaction may have an influence on heat generation inside lithium ion battery. Shrotriya et al. [60] conducted a parametric analysis for the heat generation inside lithium ion battery. In fact, the correlation between main factors mentioned above can be directly viewed in the mathematical model that is mainly used for describing internal electrochemical process and heat generation. They analyzed the polarization heating, joule heating, entropic heating and plastic heating inside lithium ion battery by using electrochemical model. The polarization heating is considered to be caused by the over-potential in solid phase (active particle materials), while the solid diffusion coefficient governs the polarization gradient inside solid phase. Besides, according to Shrotriya et al.'s report, due to the influence of solid diffusion coefficient on Li + concentration gradient across the active particle materials, plastic strain may exist inside the particle, and part of the plastic strain energy can be dissipated in the form of heat. Joule heating depends on the electric resistivity in the materials, which can be calculated by Ohm's Law. The entropic heating is caused by entropic property of electrode materials. The reversible heat generation inside lithium ion battery includes reaction heat generation, while irreversible heat generation is mainly consist of polarization heat generation and ohmic heat generation. Polarization in lithium ion battery mainly includes ohmic polarization, electrochemical polarization, concentration polarization.

The reversible heat generation is caused by intercalation and deintercalation of Li + accompanied with the structure reconstruction of

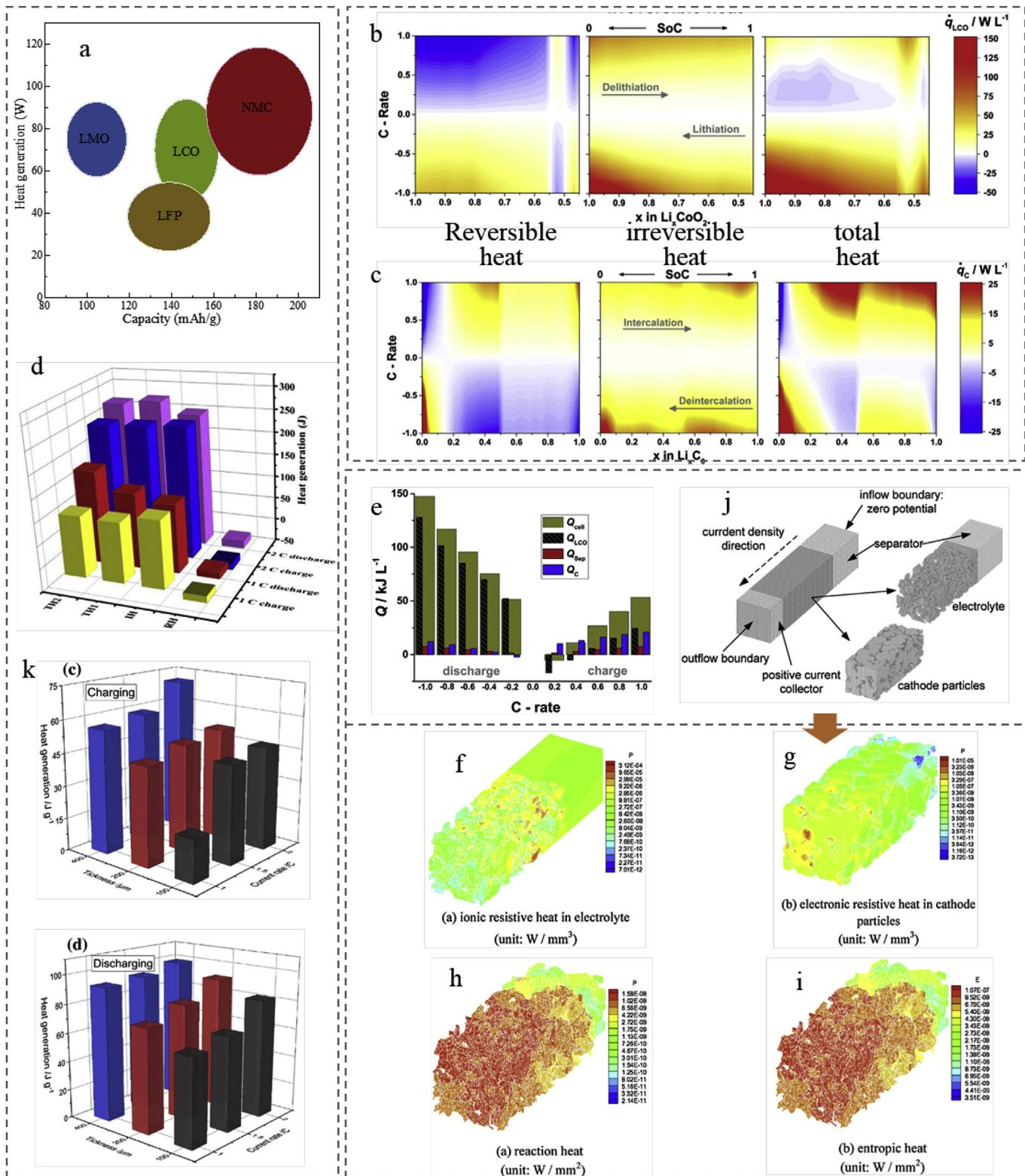


Fig. 4. Heat generation and distribution inside lithium ion battery at normal state. **a** shows the heat generation of different kinds of lithium ion batteries. **b**, **c**, heat generation inside lithium ion batteries (**b**). Different kinds of heat generation in cathode (**c**) Different kinds of heat generation in cathode. **d**, heat generation at different C-rates. **e**, heat generation of different components inside lithium ion battery. **f**, **g**, **h**, **j**. The heat generation is the electrode and electrolyte. **f**. ionic resistive heat in electrolyte. **g**. electronic resistive heat in cathode particles. **h**. reaction heat in electrode. **i**. entropic heat in electrode. **j**. the model for calculation of heat generation in electrode and electrolyte. **k** shows heat generation with different C-rates and sizes of electrode. Panels reproduced with permission from: **a**, refs. [52–55] (Elsevier). **b**, **c**, **e**, ref. [56] (Elsevier). **d**, ref. [57] (Elsevier). **f**, **g**, **h**, **i**, **j**, ref. [58] (Elsevier). **k**, ref. [59] (Springer).

electrode active materials, which is a reversible process with endothermic and exothermic course. The irreversible heat generation mostly includes ohmic heat generation and polarized heat generation [61]. The ohmic heat generation is mainly caused by the resistance of Li⁺ diffusion, electronic conduct in lithium ion battery. While polarization in lithium ion battery mainly includes ohmic polarization, electrochemical polarization, concentration polarization, which demonstrates that the polarized heat generation mainly caused by polarized resistance. Hence, Li⁺ diffusion coefficient, electric conductivity, charge transfer impedance (R_{ct}) and R_{SEI} in cathode, anode, electrolyte and separator have a significant influence in the heat generation rate of lithium ion battery.

Table 1 summarized representative reports pertaining to known cathode and anode materials. **Table 1** shows that Li⁺ diffusion coefficient and electric conductivity in cathode is relatively smaller than that in anode and electrolyte, which also demonstrate the potential higher heat generation rate in cathode in cathode of lithium. Li-ion diffusion inside the porous electrode is a complex process pertaining to multi-scale process. As shown in **Table 1**, Li⁺ diffusion coefficient in cathode materials (solid phase) is much higher than that in liquid phase. Hence Li⁺ diffusion coefficient in solid phase is the key factor for battery

Table 1
The reported reference value for Li-ion battery.

Samples	Li ⁺ diffusion coefficient (cm [2].s ⁻¹)	Electric conductivity (S.cm ⁻¹)	Specific capacity (mAh.g ⁻¹)
Cathode			
NMC111 (298 K) in ref. [62]	$3 \times 10^{-11} - 4 \times 10^{-11}$		~140
NMC111 (324 K) in ref. [63]	2.2×10^{-8}	9.1×10^{-9}	–
NMC111 (334 K) in ref. [63]	5.5×10^{-8}	2.1×10^{-8}	–
NMC442 (298 K) in ref. [62]	$3.5 \times 10^{-11} - 4.2 \times 10^{-11}$		~140
NMC532 (298 K) in ref. [62]	$3 \times 10^{-11} - 5 \times 10^{-11}$		~145
NMC532 (323 K) in ref. [63]	1.5×10^{-8}	8.7×10^{-8}	–
NMC532 (333 K) in ref. [63]	4.5×10^{-8}	9.3×10^{-9}	–
NMC622 (248 K) in ref. [62]	$2 \times 10^{-11} - 4 \times 10^{-11}$		
NMC622 (273 K) in ref. [62]	$3 \times 10^{-11} - 6 \times 10^{-11}$		
NMC622 (298 K) in ref. [62]	$7 \times 10^{-11} - 8 \times 10^{-11}$		~159
NMC622 (323 K) in ref. [62]	$9 \times 10^{-11} - 1 \times 10^{-10}$		
NMC71515 (298 K) in ref. [62]	$4 \times 10^{-11} - 7 \times 10^{-11}$		~161
Pristine-LCO in ref. [64]	$6.5 \times 10^{-11}/4.5 \times 10^{-12}$	–	190
La/Al-doped LCO in ref. [64]	$1.2 \times 10^{-10}/4.7 \times 10^{-11}$	–	
C/LFP in ref. [65]	$10^{-15} - 10^{-9}$	–	166.8
Anode			
C in ref. [66]	10^{-11}	–	
Si/C [67]	$10^{-11} - 10^{-9}$	–	
Electrolyte (298 K)			
0.01 M LiCl in H ₂ O in ref. [68]	$(1.03 + 0.01) \times 10^{-5}$	$(1.1 + 0.02) \times 10^{-3}$	
1 M LiPF ₆ in EC/DMC in ref. [68]	$(2.24 + 0.03) \times 10^{-6}$	$(1.1 + 0.10) \times 10^{-2}$	
0.3MLi-TFSI in MPPyrr-TFSI in ref. [68]	$(5.88 + 0.04) \times 10^{-8}$	$(2.6 + 0.02) \times 10^{-3}$	
0.5MLi-TFSI in MPPyrr-TFSI/EC in ref. [68]	$(5.72 + 0.98) \times 10^{-7}$	$(9.3 + 0.05) \times 10^{-3}$	
1.3MLi-TFSI in DMMA-TFSI/PC in ref. [68]	$(1.46 + 0.03) \times 10^{-7}$	$(2.0 + 0.01) \times 10^{-3}$	

performance and thermal issue in lithium ion battery. The Li-ion diffusion coefficient in solid phase can be tested by galvanostatic intermittent titration technique (GITT), potentiostatic intermittent titration technique (PITT) and electrochemical impedance spectrum (EIS) methods.

The Li-ion diffusion process has a direct relation with the electrode thickness, which linearly decreases with the augment of electrode thickness, especially when the thickness higher than the critical electrode thickness (about 200 μm for NMC622)^[69]. The rapid decrease in Li-ion diffusion coefficient in solid phase is caused by the localized Li-ion oversaturation (in deintercalation process) and over-consumption (in intercalation process). The design parameters (porous and tortuosity etc.) should be further optimized to improve Li-ion diffusion process. As shown in **Table 1**, Li⁺ diffusion coefficient also has a positive correlation with temperature. And among a series of NMC cathode materials, NMC622 has the largest Li-ion diffusion coefficient with a smaller fluctuation with temperature. Meanwhile, the investigation suggested that a larger Li slab space would lead to larger Li-ion diffusion with lower activation energy along the diffusion pathway, and the Li-ion diffusion coefficient would be facilitated with a less temperature dependence, which means that NMC622 is the most promising candidate for lithium ion battery in the multi-temperature applications [62].

In order to improve Li-ion diffusion coefficient and battery performance of layered oxide framework cathode materials, doping with different elements and surface modification would be useful method. Liu et al. [64] developed a La/Al doped LCO cathode materials. The La works as a pillar while Al works as a positively charged center to facilitate Li-ion diffusion, to stabilize the layered structure and to suppress phase transition of LCO cathode materials. The Li-ion diffusion coefficient of La/Al-doped LCO cathode at the initial charge state and the final discharge state is much higher than that of pristine LCO cathode at the same state (**Table 1**). Wang et al. [70] prepared a TiO₂-coated LCO cathode material. The TiO₂-coating on the surface of pristine LCO cathode material would reduce surface oxygen activity and surface oxygen loss to suppress the electrolyte decomposition and layered structure degradation. Meanwhile, the increased Li slab space and decreased Li-ion transfer activation energy barrier would facilitate Li-ion diffusion in crystal structure and Li-ion transfer through the solid electrolyte interphase.

The electronic conductivity of NMC cathode materials increases with increasing SOC (magnitude is from 10^{-7} S.cm⁻¹– 10^{-2} S cm⁻¹), while the Li-ion diffusion coefficient deceases with deceasing SOC. Meanwhile, the Li-ion diffusion coefficient increases with increasing temperature, while the electric conductivity deceases with increasing temperature [63]. The solid-state diffusion coefficient decreases with decreasing temperature because of the increased solid-state diffusion activation energy. Meanwhile, the charge-transfer resistance and SEI resistance increases at low temperature, causing severe kinetic limitations. The Li-ion diffusion coefficient for K⁺ in graphite anode is much smaller than that of Li⁺ (two orders of magnitude smaller). The graphite anode exhibits a good cycling stability at 40 °C [66]. The carbon-rich silicon-oxy carbide ceramic is a promising anode for lithium ion battery. Compared to graphite and disordered carbons, Li-ion diffusion coefficient in carbon-rich silicon-oxy carbide ceramic is less potential dependent, which is caused by the particular morphology of the free carbon phase [67]. The Li-ion diffusion coefficient is one of key parameters to evaluate the electrolyte. As shown in **Table 1**, the electric conductivity is similar in different electrolytes. The Li-ion diffusion coefficient is in the range of 10^{-8} to 10^{-5} cm [2].s⁻¹, while the electric conductivity is in the range of 10^{-3} to 10^{-2} S cm⁻¹⁶⁸. The drastic differences in Li-ion diffusion coefficient can influence the battery performance.

Besides, the thickness of electrode also has an influence on heat generation rate, as shown in Fig. 4k and heat generation rate increases significantly with increasing thickness of electrode both in charging and discharging process [59]. Hence the reconstruction of electrode is an efficient way to decrease heat generation and improve battery performance. An ideal structure of electrode is necessary for increasing

electrochemical performance and decreasing heat generation, and there are also some methods to design a novel architecture of electrode. Chiang et al. [71] developed an electrode construction method to improve electrode kinetics. Similarly, Cui et al. [72] employed wood template to construct an architecture electrode, and the Li-ion conductivity and electron conductivity is significantly improved. Besides, magnetic template also can be employed to construct ordered cathode and anode electrode, including the magnetic alignment of LCO cathode [73], and

magnetically aligned graphite electrodes [74]. The magnetic field method is proved to be an effective method to align the cathode and anode materials.

3.3. State III: at high temperature (>90 °C)

State III is recognized as thermal runaway process, and Fig. 5 shows the chain reactions and the relevant energy release of each reactions in

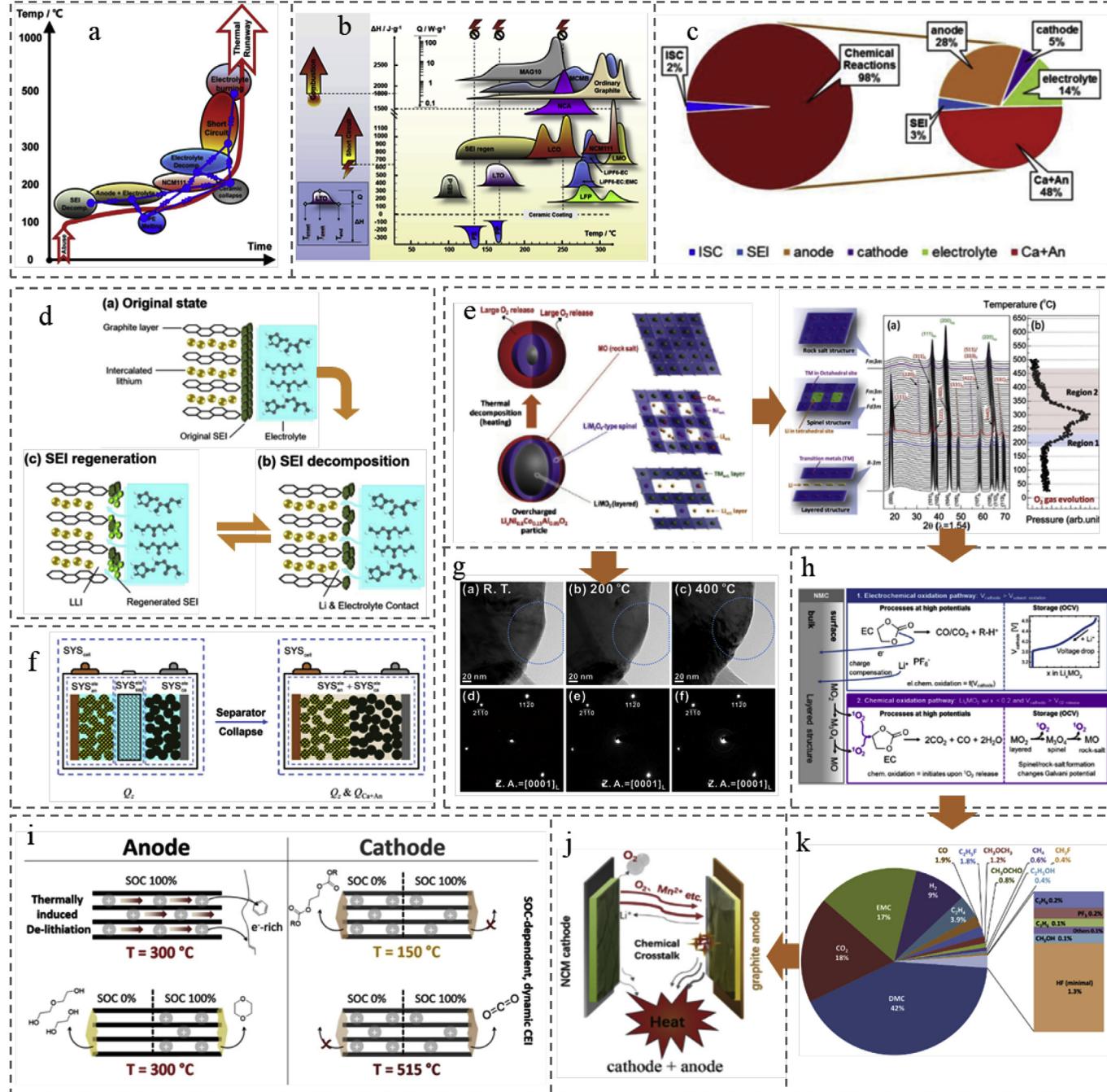


Fig. 5. Heat generation of chain reactions and the key issues in thermal runaway process. **a** shows chain reactions in thermal runaway process. **b** shows main heat generation in thermal runaway process. **c** shows the percentage of total heat generation during thermal runaway process. **d**, **f**, The key points in thermal runaway process, the decomposition of SEI layer (**d**) and the melting of separator (**f**). **c**, **d**, The key points in thermal runaway process, the decomposition of SEI layer (**c**) and the melting of separator (**d**). **e** shows the structure evolution with gas release of NMC cathode materials during heating process and the oxygen release mechanism of NMC cathode materials from overcharge state during heating up to 500 °C. **g** shows the structure evolution at different temperatures. **h** shows the two mechanistic electrolyte oxidation pathways. **i** shows the side reactions of anode and cathode with electrolyte. **j** shows the thermal runaway without internal short circuit. **k** shows global chemical composition of the vent gas. Panels reproduced with permissions from: a, ref. [14] (Elsevier); b, ref. [43] (Elsevier); c, d, f, ref. [18] (ECS). c, ref. [82] (Wiley); e, ref. [80] (Wiley); g, ref. [81] (ACS); h, ref. [83] (ECS); i, ref. [84] (Elsevier); j, ref. [15] (Cell Press); k, ref. [29] (Elsevier).

thermal runaway. As shown in Fig. 5a, the decomposition of SEI film is considered as the beginning of thermal runaway, and after that, a series of exothermic reactions will be triggered. Feng et al. [14] summarized the chain reactions (Fig. 5a) and energy release diagram of each reactions (Fig. 5b) in thermal runaway process. Fig. 5c shows the main heat generation aspects and its proportion in the total thermal release in thermal runaway process. The largest proportion of heat generation is caused by rapid oxidation-reduction reaction between the cathode and anode ($\text{Ca} + \text{An}$), about 48%, while the heat generation of other chemical reactions in anode, cathode and electrolyte is much smaller. The smallest heat generation is decomposition of SEI film [18].

3.3.1. Propagation and accumulation of thermal hazard inside lithium ion battery

Decomposition of SEI film. The decomposition of SEI film is the key point for the beginning of thermal runaway, which means that the corresponding temperature is considered as critical temperature in thermal runaway process (Fig. 5d) [18]. As the temperature is over the critical temperature, the chain reactions will be induced and side reactions will become violent [75]. Besides, the decomposition of SEI layer will cause the side reaction between anode and electrolyte with large heat generation [76]. Meanwhile, almost all of the induced chain reactions are highly exothermic, which will lead to fast temperature rise and then cause thermal runaway.

Phase transition and decomposition of cathode materials. As shown in Fig. 5e, The onset temperature of phase transition of Ni-rich NMC materials is quite low (about 100 °C) due to the existing thermally induced reduction of Ni ions, and an additional redox reaction at 4.2 V exacerbate thermal unstable of Ni-rich NMC materials [77]. Meanwhile, metal dissolution rate of NMC materials increases as the cutoff voltage is higher than 4.6 V [78]. The phase transformation in active materials accompanied with exothermal behavior. The stronger oxidation capability of Ni^{4+} in NMC materials causes higher heat generation and oxygen release compared to the Mn^{4+} in LMO materials [79]. The overlap between $\text{Co}^{3+/4+}$ 3d t_{2g} band and O^{2-} 2p band leads to severe oxygen removal and phase transformation, accompanied with micro-crack in NMC materials (Fig. 5e and g) [80,81].

Decomposition of separator. The separator is also important for preventing thermal runaway. As shown in Fig. 5b, the PE/PP separator melts at about 150 °C, and the melting of separator will induce ISC inside battery, which will cause higher temperature rise rate, leading to uncontrollable thermal runaway. Besides, exothermic reactions inside the battery will cause a higher temperature rise, and the flammable liquid electrolyte accompanied with released gases (especially O_2) also will lead to the fire and explosion.

Gas emission and decomposition of electrolyte. Gas emission is accompanied with overheating and thermal runaway process, and the released gases are widely concerned by researchers due to the toxicity, flammability and the risk of gas explosion [85]. Gas release is a common phenomenon in lithium ion battery, which is mainly caused by structure reconstruction of cathode (Fig. 5e) [80] and side reactions in electrode and electrolyte (Fig. 5h) [83]. The global chemical compositions of vent gas are shown in Fig. 5k, which demonstrate that the main compositions of gas emission is caused by flammable volatile solvents [29]. The oxygen release in cathode materials will lead to structural degradation and rapid capacity fade, triggering the parasitic thermal runaway. Oxygen release occurs at the surface of cathode materials with the valence state reduction and structural reconstruction, leading to inter/intragranular cracking in the bulk of cathode materials, which is a complex reaction at diverse abuse conditions and is influenced by various parameters [86]. The extracted oxygen species will oxidize the electrolyte solvents and generate acidic species, and then attack the solid electrolyte interface, causing a high electrode resistance [87]. Meanwhile, the released oxidative gas will transfer to anode and reacts with reductive LiC_x at 274.2 °C, accompanied with tremendous heat generation (Fig. 5j) [15].

Jung and Gasteiger et al. [83,88] conducted a series of work about

oxygen release in NMC cathode materials, including the influence of temperature and cycling stability. Their research shows that the surface structures propagate towards to the core, and the rock-salt phase becomes the dominant structure at high temperature. The oxygen release in layered cathode materials is caused by H2–H3 phase transition, and the released lattice oxygen will have a chemical reaction with electrolyte, leading to the evolution of CO_2 and CO. Besides, they find that the oxygen release will occur as the about 81% of Li-ions remove from layered cathode materials, which is caused by the formation of surface spinel and/or rock-salt phases as the state of charge reaches 81% in charging process [88]. The amount of oxygen release increases with increasing temperature, as well as CO_2 and CO. While the CO_2 evolution before oxygen release is caused by ethylene carbonate hydrolysis and electrolyte impurity oxidation [73]. The O_1 structure at the surface of NMC, this phase transforms into the Co_3O_4 -type spinel structure as stays as such to high temperature [80].

Other exothermic reactions. In the overcharge and thermal abuse process, the main composition is volatile solvents and flammable gases and temperature is the key parameters for gases evolution [29]. The probability of thermal runaway in lithium ion battery grows with number increase of charge/discharge cycles and increase of cells of SOC. With the number growth of cells charge/discharge of cycles, there is an obvious decline of initiation of exothermic reactions of thermal runaway and increase of release energy. Meanwhile, in the course of cells cycling, hydrogen release occurs in anode graphite, and it is a powerful exothermic reaction, which increases the released energy in the beginning of the thermal runaway and decreases the temperature of initiation of thermal runaway [89]. Meanwhile, as heating up to a high level (650 °C), the electrolyte will release organic compounds, toxic decomposition products [85]. Besides, Thermal induced side reactions between electrophilic lithium ion in anode/cathode and electrolyte may occur at extremely high temperature, which may have a further influence on the thermal safety of electrode materials. The thermally induced de-lithiation in anode materials will occur at 300 °C, while for cathode materials, the side reactions may happen on the surface at 150 °C [84].

3.3.2. Relevant solutions for preventing exothermic reactions

Side reactions inside lithium ion battery can be prevented by adding relevant additives in the electrolyte and coating materials on the surface of active materials, and the design and preparation of novel alternative materials lithium ion battery is also an effective way to improve for thermal safety. Fig. 6 shows the reported solutions for preventing exothermic reactions inside lithium ion battery. Stabilizing additives is a promising candidate for improving thermal stability of electrolyte and electrode. As shown in Fig. 6a, Dimethylacetamide (DMAc) is proved to an efficient additive for improving thermal stability of electrolyte and the SEI on graphite anode. Xu et al. [90] demonstrated that 1% DMAc in electrolyte can effectively keep the thermal stability of electrolyte and SEI layer at 85 °C for a long period. Other additives, such as cresyl diphenyl phosphate (CDP), can also be a promising candidate for improving thermal stability and thermal safety of lithium ion battery (Fig. 6e) [91]. Besides, novel safety electrolyte can be another choice for improving thermal safety of lithium ion battery. A liquid-like poly (ionic liquid) was synthesized, which is proved to be an excellent choice as electrolyte inside lithium ion battery to improve thermal safety of batteries (Fig. 6c) [92]. For electrode materials, surface coating and novel design are common methods to improve thermal stability. Neudeck et al. [93] employed organophosphates as coating materials to modify the NCM622 cathode materials, and the battery performance, thermal stability and thermal safety are further improved (Fig. 6b).

In order to enhance thermal safety, separator can be coated by other materials. As shown in Fig. 6d, Kim et al. [94] coated separator by amino-functionalized SiO_2 particles, which is proved to be a promising separator for a more safe battery. Apart from modification strategies, novel promising materials for thermal stability and safety are also be further investigated in the reported researches. Fig. 6f shows a new

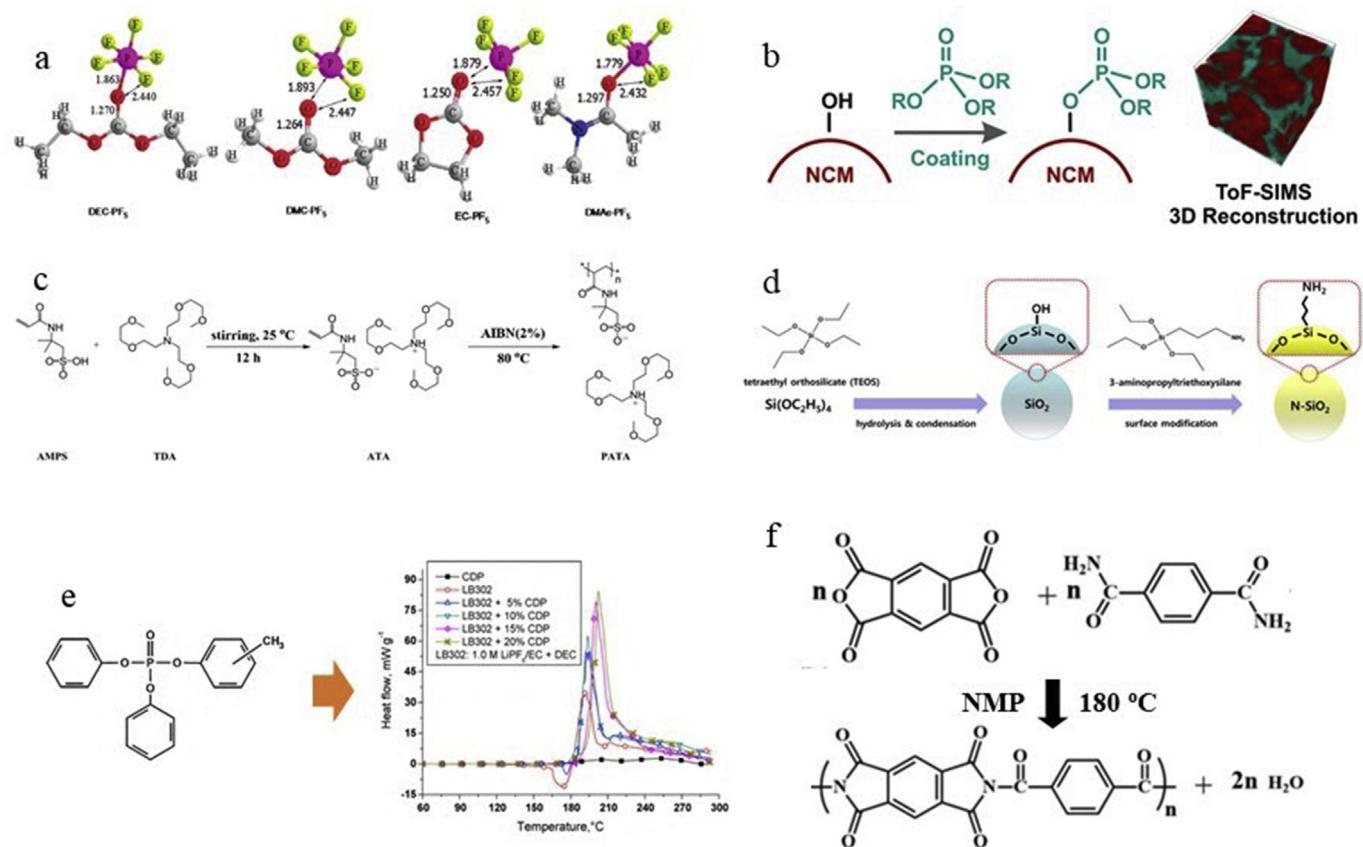


Fig. 6. Solutions for preventing exothermic reactions at high temperature. **a** shows the additives for improving thermal stability of electrolyte and graphite anode. **b** shows surface coating for cathode to improve thermal safety. **c** shows liquid-like poly (ionic liquid) as novel electrolyte. The modified separator by coating amino-functionalized SiO₂ particles (**d**). The additives for improving thermal stability of electrolyte (**e**). polyimide as novel anode (**f**). Panels reproduced with permission from: **a**, ref. [90] (ACS). **b**, ref. [93] (ACS). **c**, ref. [92] (ACS). **d**, ref. [94] (Elsevier). **e**, ref. [92] (ACS). **f**, ref. [91] (Elsevier).

anode by using polyimide. The heat release of polyimide in decomposition process is much smaller than that of graphite, which shows that the polyimide is much more safety than graphite anode.

3.3.3. Theoretical analysis from materials perspectives to batteries

In order to investigate the thermal stability and thermal effect in battery materials, theoretical calculation is conducted to analyze the properties of battery materials (Fig. 7). Based on the proposed theories by Ong and Ceder [95,96], the released free energies when the oxides form corresponding cathode materials (Gibbs formation energies) were utilized to characterize the thermal stability. The results showed that LFP and LiMnPO₄ are easier to decompose into the oxides during the charge or discharge process than the element phase. And the LFP and LiMnPO₄ are more thermodynamically stable with regard to the oxides than LCO, LiNiO₂ (LNO), and LMO [97]. Ceder et al. [98] performed high-throughput DFT computations to assess the connection between voltage and thermal safety (evaluated by temperatures when O₂ was thermodynamically released) influenced by the polyanion group, redox metal, as well as ratio of oxygen to counter-cation for more than 1409 cathode materials. The results showed that the strong inverse relation exists between voltage and thermal safety though not all oxygen release only are put down to the high voltage. In addition, this paper suggested that only a fraction of compounds on the basis of limited redox couple/polyanion combinations demonstrate both high voltage and high safety. Kim et al. [99] investigated the thermal stability of layer-structured cathode materials introduced with intrinsic point defects with first principle simulations. Shahbazian-Yassar et al. [100] performed Ab initio molecular dynamics simulations (AIMD) to provide an explanation for the inhomogeneous oxygen release from LCO cathode

crystals at high temperatures. The simulated results showed that oxygen evolution behavior lies on the exposed Miller index facets. The facets with lower surface energy, such as [0 1 2] and [1 0 4], will produce oxygen more easily (the oxygen release occurs at the [0 1 2] and [1 0 4] facets at 300 and 400 °C within time range from 400 to 900 fs) while those with higher surface energy, such as [0 0 1], will release oxygen more difficultly (no oxygen evolution at time spans of about 2 ps at 300 and 400 °C). Pan et al. [101] designed new layered NMC ($x+y+z=1$) materials to enhance the thermal stability and utilized ab initio calculations to investigate the tuning mechanism of the thermal stability. They successfully clarified the tuning mechanism and methods of the most unstable oxygen on the thermal stability of NMC and pointed out that thermal stability of NMC was determined by the oxygen local coordination structure unit (TM(Ni, Mn, Co)₃-OLi_{3-x}). The influence of lithium content, valence states of Ni, contents of Ni, Mn, and Co, and Ni/Li disorder on the sites, content, and the release temperature of the most unstable oxygen were also elucidated. They proposed that the development of Li[Ni_{0.5}Mn_{0.5}]_{1-x}CoxO₂ (0 < x ≤ 0.33) system will be a prospective method to realize the optimization of thermal stability of NMC with high thermal stability and the fast Li-ion diffusivity as well as high reversible capacity for “Ni=Mn” group. Han and Kang et al. [102] constructed archetype material LNO surface coated with ultrathin Al₂O₃ (0001) as Li de-/intercalation channel in order to improve thermal stability of high Ni compositional cathodes. They carried out density functional theory calculations to capture the phase transition of Al₂O₃ from the corundum-type crystalline (c-Al₂O₃) to amorphous (a-Al₂O₃) when the layer number of Al₂O₃ equals 3. And then they found the inhibition of a-Al₂O₃ on oxygen gas evolution at 400 K by performing the ab initio molecular dynamic simulations owing to strong ionic bonding

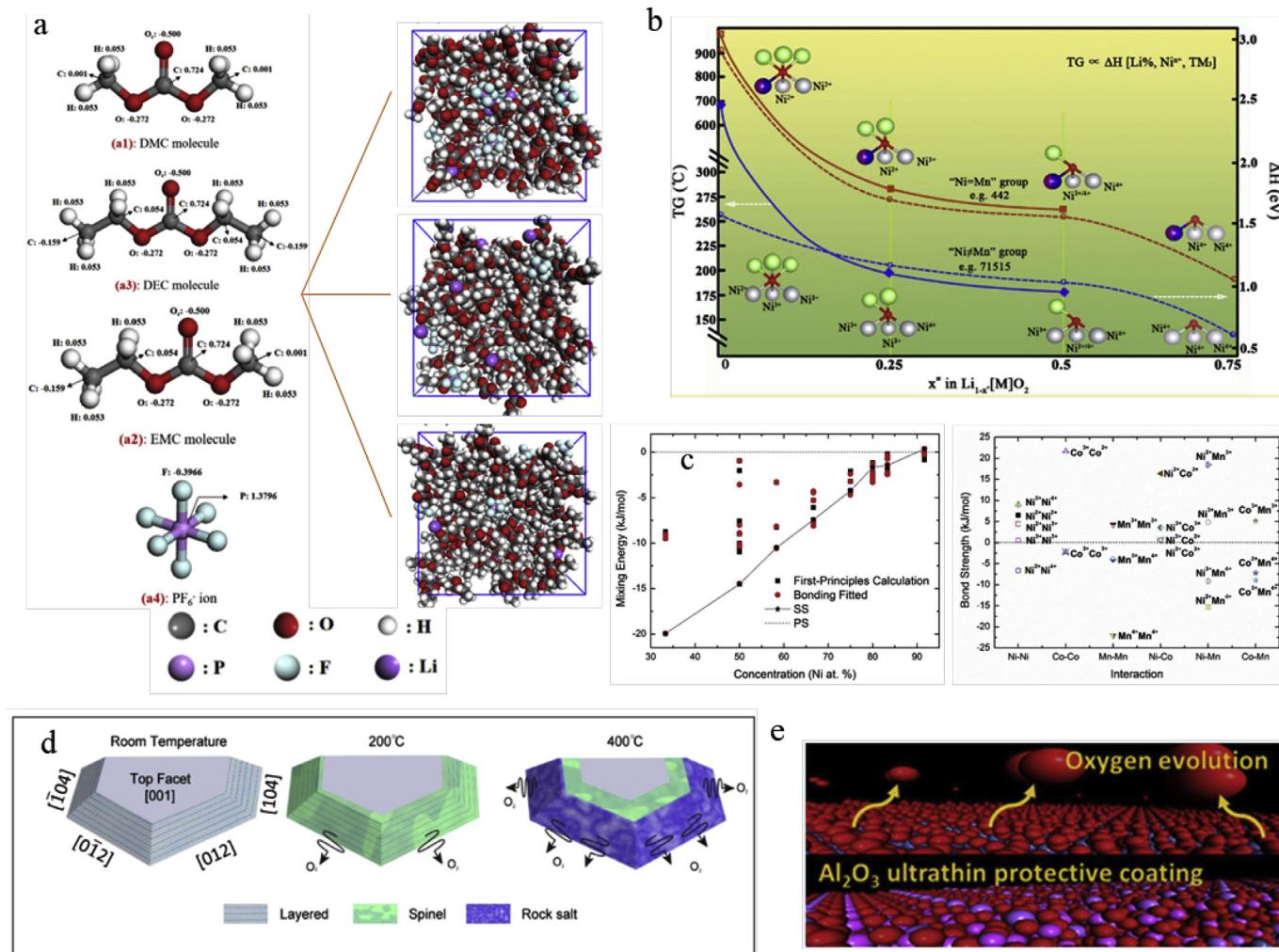


Fig. 7. The calculation for thermal stability of battery materials. **a** shows the calculation of thermal degradation of electrolyte. **b** shows the thermal stability analysis of Ni-Rich LiNi1-2xCoMnO_2 cathode materials. The theory analysis of thermal stability of LCO (**d**). Thermal stability analysis of LiNiO_2 Materials Coated by Amorphous Al_2O_3 (**e**). Panels reproduced with permission from: **a**, ref. [113] (Elsevier), **b**, ref. [114] (ACS), **c**, ref. [115] (ACS), **d**, ref. [116] (ACS), **e**, ref. [117] (ACS).

characteristic between Al and O at the interface. In addition, the simulated results showed that the thermodynamic stability of a- Al_2O_3 increases when there are Li atoms in the layer.

The artificially constructed SEI layer is formed by the reaction between the ethylene oxides and graphite edge plane with functional group of -OH. This kind of graphite SEI layer can effectively prevent solvent from being co-intercalated into graphite layers. In addition, the thermal stability of the designed SEI is attributed to the covalent bonds between PEO and the graphite in comparison with the physical coating [103].

The additives can improve the thermal stability of electrolyte. Li et al. [104] reported the additive vinyltriethoxysilane (VTES) that was used to keep the flammability of a carbonate based electrolyte down. They found that this kind of additive not only suppress the flammability of the electrolyte, but also improve the thermal stability of the electrolyte and the cathode material LCO due to formation of a stable surface layer. The authors utilized ab initio simulations to explain the formation mechanism of the surface layer. The results showed that the stable components will be preferably produced through the reaction between the vinyl silyl radical of VTES and the oxygen atoms of solvent molecules.

Thermal stability of anode materials is also important for battery thermal safety. molecular dynamics and density functional theory simulations also can be used to predict thermomechanical properties of two-dimensional psi (ψ)-graphene, which are actually defect-filled

graphene-like membranes including 5-, 6-, and 7-membered carbon rings. The simulations showed that ψ -graphene demonstrated a non-positive linear thermal expansion coefficient, a good specific heat capacity, and excellent elastic constants, which is beneficial to avoid the thermal runaway due to overheating and prevent lattice damage during the drastic volume expansion of the lithium ion batteries [105]. Yi et al. [106] performed DFT calculations to understand the safety of $\text{Li}_4\text{Ti}_5\text{O}_12$. They found the low formation enthalpy is the reason for high thermodynamic stability of $\text{Li}_4\text{Ti}_5\text{O}_12$. The good thermal stability of $\text{Li}_4\text{Ti}_5\text{O}_12$ is associated with the highly covalent bonding between Ti and O on the basis of the electron density difference diagram.

The change of oxidation state and site preference of TM (i.e. Ni, Co, Mn) in NMC materials will determine the origin of thermal decomposition of NMC materials [107]. Thermal decomposition temperature is a key parameter to evaluate thermal safety of cathode active materials. The Ni-rich NMC cathode materials ($x + y < 0.5$) mainly consist of $\text{Ni}^{3+}/\text{Mn}^{4+}$ and $\text{Ni}^{3+}/\text{Ni}^{3+}$, while the major ionic bond in common NMC materials ($x + y > 0.5$) are $\text{Ni}^{2+}/\text{Mn}^{4+}$ and $\text{Co}^{3+}/\text{Mn}^{4+}$. The thermal stability of Ni-rich NMC is not as good as common NMC materials, because the bond strength of $\text{Ni}^{3+}/\text{Mn}^{4+}$ and $\text{Ni}^{3+}/\text{Ni}^{3+}$ is weaker than that of $\text{Ni}^{2+}/\text{Mn}^{4+}$ and $\text{Co}^{3+}/\text{Mn}^{4+}$ [108]. In the thermal decomposition process, Mn ion keeps in 3a octahedral sites of layer structure, and Co will migrate to 8a tetrahedral sites of spinel structure, which plays a crucial

role in thermal stability [107]. The thermal stability of NMC cathode materials decreases with the increasing Ni content [109]. As the Ni content rise up to 0.95, the phase transition temperature decrease to 180 °C, a quit low thermal stability [110]. The structural reconstruction (formation of a surface reduced layer) and chemical evolution (formation of a surface reaction layer) will occur at the surface of NMC particles in delithiated process, accompanied with oxygen release and transition metal (TM) migration. The surface reconstruction forms along with lithium diffusion channels, exhibited a strong anisotropic characteristic, and the surface reaction layer consist of lithium fluoride embedded in a complex organic matrix. The formation of surface reduced layer and surface reaction will lead to impedance buildup and capacity fade [111]. Otherwise, the formation of surface reduced layer and the type of transformation have a strong relationship with cutoff voltage condition. The oxygen loss from the surface structure will occur in highly oxidative environment (higher voltage condition), causing dominant formation of rock salt phase in the surface of the materials [82]. The formation of inertia phase (rock salt phase) is ionically insulating phase and lead to sluggish kinetics, causing the charge transfer resistance increase.

The coupled thermal and mechanical effect electrochemically triggered aggravate the failure of NMC cathode. The thermal stress (due to electrochemically induced phase inhomogeneity) and internal pressure (caused by oxygen release) mainly lead to intragranular cracking [112].

For further investigating heat generation in lithium ion battery, plenty of battery models and calculation methods is proposed in the past decades, such as empirical and semi-empirical model, electrochemical-thermal (ECT) model, equivalent circuit model, electro-thermal model and Bernardi heat generation model [13,118]. Empirical or semi-empirical model is mainly based on the experimental result, while ECT model is based on the internal electrochemical reactions, Li ion diffusion and electron conductivity inside lithium ion battery, which is commonly used for analyzing the heat generation and electrochemical performance in batteries^{119–55}. Equivalent circuit model is built by considering the battery as electric circuit, including several electronic components. Besides, electro-thermal model is mainly used for analyzing the heat generation caused by ohmic resistance [120]. And Bernardi model is used for calculating the heat generation caused by reaction heat and ohmic heat, which is commonly used for the overall thermal performance in battery pack, and to evaluate BTM performance [121].

Heat generation in lithium ion battery mainly consist of reversible heat generation, irreversible heat generation and mixed heat generation (caused by side reactions). Irreversible heat generation is much higher than reversible and mixed heat generation in lithium ion battery, and the mixed heat generation is lower than reversible heat generation [122]. Heat generation rate is different with different discharged rates and temperatures. Heat generation rate increases with the increasing charge/discharge rates, but with the increasing temperature, the heat generation rate decreases gradually [52,55]. Heat generation rate has a relationship with its specific capacity [52–55]. NMC has a much higher specific capacity (especially Ni-rich NMC cathodes) with a high heat generation rate. While the LFP has a relatively high specific capacity with a much lower heat generation rate, which means that the LFP has much higher safety at normal state.

4. Cell design for better electric and thermal performance

The cell design is another important part for the electric and thermal performance of lithium ion battery. The design and arrangement from electrode to the overall battery perspectives is further considered by researchers all over the world. Wang et al. [123,124] conducted a series of work for a better cell design. From the internal aspects, they considered the influence of electrode design parameters, such as electrode thickness, volume fraction of active materials and particle size, on the battery performance. Besides, the tab dimension optimization is considered for decreasing heat generation and temperature, keeping the thermal safety of lithium ion battery. Hosseinzadeh et al. [125] developed a

multi-layered electrode with different porosities to improve the electric and thermal performance. The influence of microstructure of electrode on the thermal performance also should be considered. Sangros et al. [126] investigated the effect of electrode structure on thermal conduction of electrode. The thermal conductivity of electrode, and the heat generation rate at different porosity and thicknesses were further analyzed. Li et al. [127] developed an electric-thermal model for the thermal design of pouch-type large format lithium ion battery. The electric distribution and thermal performance were considered in the design process.

5. Propagation of thermal issue in battery and battery pack

In general, the temperature distribution of lithium ion battery is caused by a comprehensive effect of internal heat generation, internal heat conduction and external heat dissipation. Thermal behavior and temperature distribution inside lithium ion battery is important for the electric and thermal performance for batteries. Jia and An et al. [128] investigated the thermal behaviors and lithium ion transport inside the batteries, which has a closely relationship with battery performance. They developed a 2D electrochemical-thermal model to analyze the heat and mass transfer in lithium ion battery (Fig. 8a). The results show that the non-uniformity of current and potential distribution on the geometrical structure can lead to an uneven temperature distribution. Besides, the heating effect of tab will influence the overall temperature distribution inside lithium ion battery. Yang and Liu et al. [129] developed a multi-node electro-thermal model to investigate the non-uniformity of temperature distribution inside prismatic lithium ion batteries (Fig. 8b). Moreover, the complex internal structure may induce uneven temperature distribution, as shown in Fig. 8d. While for battery pack, the internal temperature distribution in lithium ion battery can be assessed by Bi number ($Bi = hl/\lambda$), Where the h is the external heat transfer coefficient, l is the characteristic length and λ is thermal conductivity.

As shown in Fig. 8e, only when the Bi number is lower than 0.1, the temperature inside the battery can be regarded as uniform distribution. While the heat convection outside the battery will influence the total heat generation and thermal propagation in battery and battery pack [130]. And the external forced convection can effectively reduce the temperature of battery in normal state and thermal runaway process [132]. The cylindrical battery has a complex structure inside the battery can, inducing an anisotropy of thermal conductivity inside cylindrical battery. Hence the heat mainly will conduct along with the axial direction [131]. At normal state, the heat generation mainly occurred on the aluminium tab of battery (Fig. 8d and e), because of the higher current density in tab compared with the body of battery. While in thermal runaway process, the thermal reactions (side reactions) occurred one after another with highly exothermic, inducing a much higher self-heating rate. As a consequence, the temperature gradient inside the battery is much larger than that at normal state [120]. As the local ISC occurred, the temperature will increase to a quite high level at few minute, and the thermal runaway propagation will spread fast inside the cell [132].

The thermal runaway propagation will induce severe thermal hazard in battery pack. The limited contact between batteries (cylindrical battery) will be less prone to propagate thermal failure, while the electrical connectivity will impel the propagation of thermal failure in single battery. For pouch type battery, the well contact between batteries will induce a faster heat transfer in battery pack [133]. The heat transfer in prismatic battery pack mainly consists of three parts, i.e. heat transfer through battery shell, pole connector and fire spread. The heat transfer by battery shell is the principal thermal propagation process, and heat transfer by pole connector is about 1/10 of that. The fire is mainly caused by the burst out electrolyte, and the fire can scorch adjacent batteries (Fig. 9a), but fire spread is also not the main heat transfer path for thermal propagation. The TR propagation in battery pack is a sequence process, which is mainly caused by heat transfer and voltage response in the battery [9].

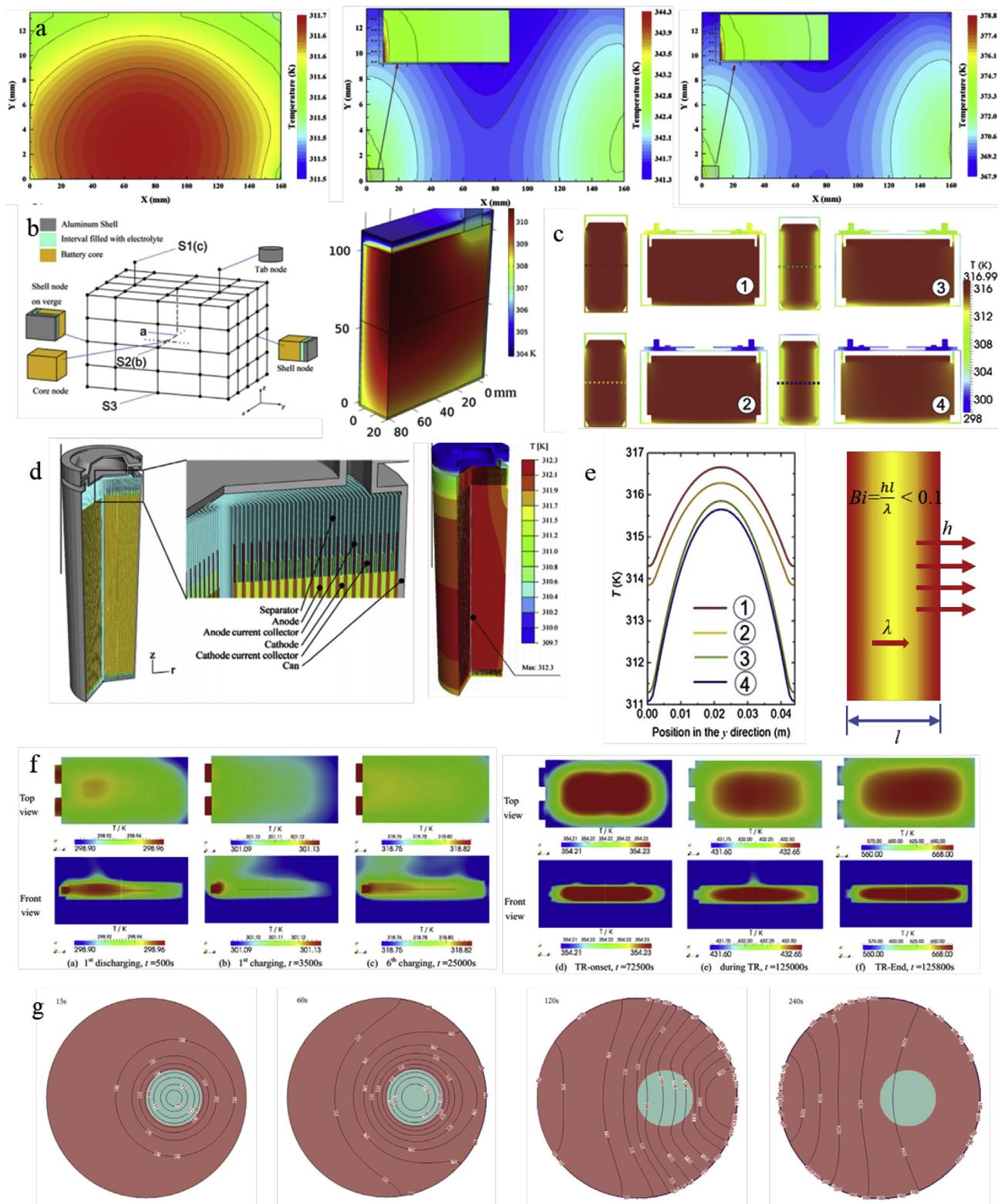


Fig. 8. The heat transfer and temperature distribution inside and outside the cell. **a** shows the internal temperature distribution inside shows the internal temperature distribution of cell and the evaluation standard for temperature distribution. **b** shows 3D temperature distribution inside lithium ion battery by using multi-node electro-thermal model. **c** shows the effects of different boundary conditions on the local temperature distribution. The internal structure and temperature distribution of cylindrical battery (**d**). The internal temperature distribution of cell and the evaluation standard for temperature distribution (**e**) The temperature distribution inside and outside the battery at normal state and thermal runaway process (**f**). The heat transfer and isotherms under nail-penetration condition (**g**). Panels reproduced with permission from: **a**, ref. [128] (Elsevier); **b**, ref. [129] (Elsevier); **c, e**, ref. [130] (Elsevier); **d**, ref. [131] (Elsevier); **f**, ref. [120] (Elsevier); **g**, ref. [132] (Elsevier).

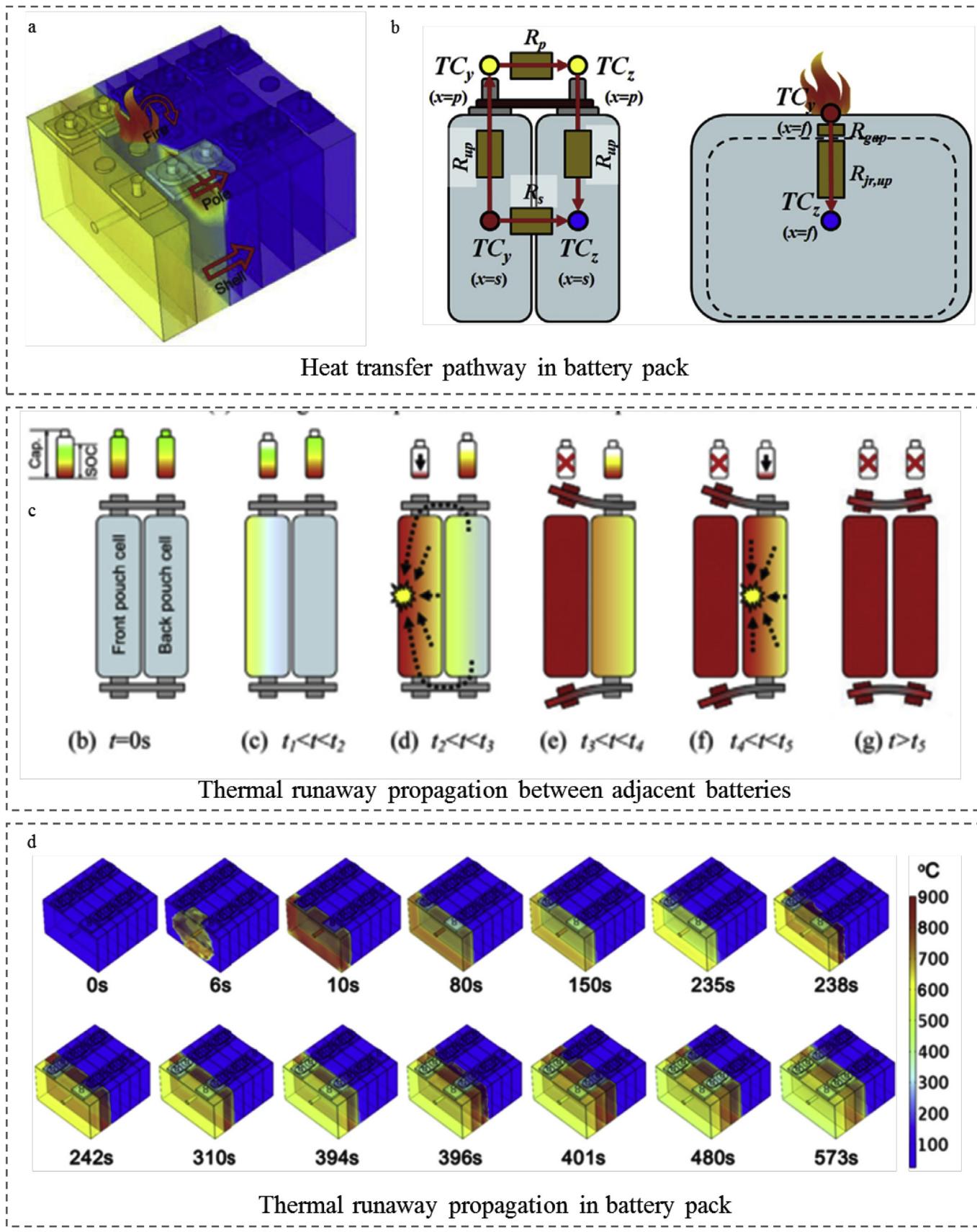


Fig. 9. The heat transfer and thermal runaway propagation in battery and battery pack. **a-b**, Heat transfer paths during thermal runaway propagation. Heat transfer paths (**a**), thermal resistance network representation (**b**). Thermal runaway propagation between adjacent batteries (**c**). Thermal runaway propagation in battery pack (**d**). Panels reproduced with permission from: **a, b, c**, ref. [9] (Elsevier); **d**, ref. [134] (Elsevier).

6. BTM for lithium ion battery

Thermal issues such as thermal runaway, subzero temperature battery performance and heat generation in battery are key factors for the application of lithium ion battery. And in order to investigate the thermal issue and thermal safety performance of lithium ion battery, the battery thermal model should be developed and coupled with thermal management strategies [135]. The BTM is an effective way to control the temperature rise in charge/discharge process, which can improve thermal stability and safety of lithium ion battery [136]. The common methods for BTM mainly include air, liquid, solid-liquid phase change, heat pipe and boiling based strategies [118]. Air based BTM has a simple structure, while liquid based BTM system has great potential in energy-saving and efficiency promotion. Phase change materials can be used to keep the temperature of battery into operating range when integrated in BTM [11, 137]. The heat pipe and boiling based BTM strategies are novel methods for battery packs, which has a better performance in temperature control [138].

6.1. Heating strategy at low temperature

As mentioned above, Li plating and dendrite can be easily triggered at low temperature, which may induce ISC inside lithium ion battery. Hence, heat strategy for lithium ion battery at low temperature is an important way to prevent thermal runaway at low temperature. Alternating current (AC) heating is regarded as an effective method to heat lithium ion battery from low temperature. For AC heating, the influence of current frequency, amplitudes and waveforms should be further considered. Dai and Zhu et al. [139] analyzed the effect of AC heating method on the Li plating behavior (Fig. 10a). They reported that AC heating with low frequencies and large current can cause complex side reactions and the Li plating can be triggered on the surface of graphite anode, while high frequencies will not induce Li plating. Their subsequent work considered the effect of AC heating on the state of health [140]. The result shows that high amplitudes can facilitate the heat accumulation and the AC heating will not aggravate the degradation of lithium ion battery even with a low frequency range. Xiong and Guo et al.

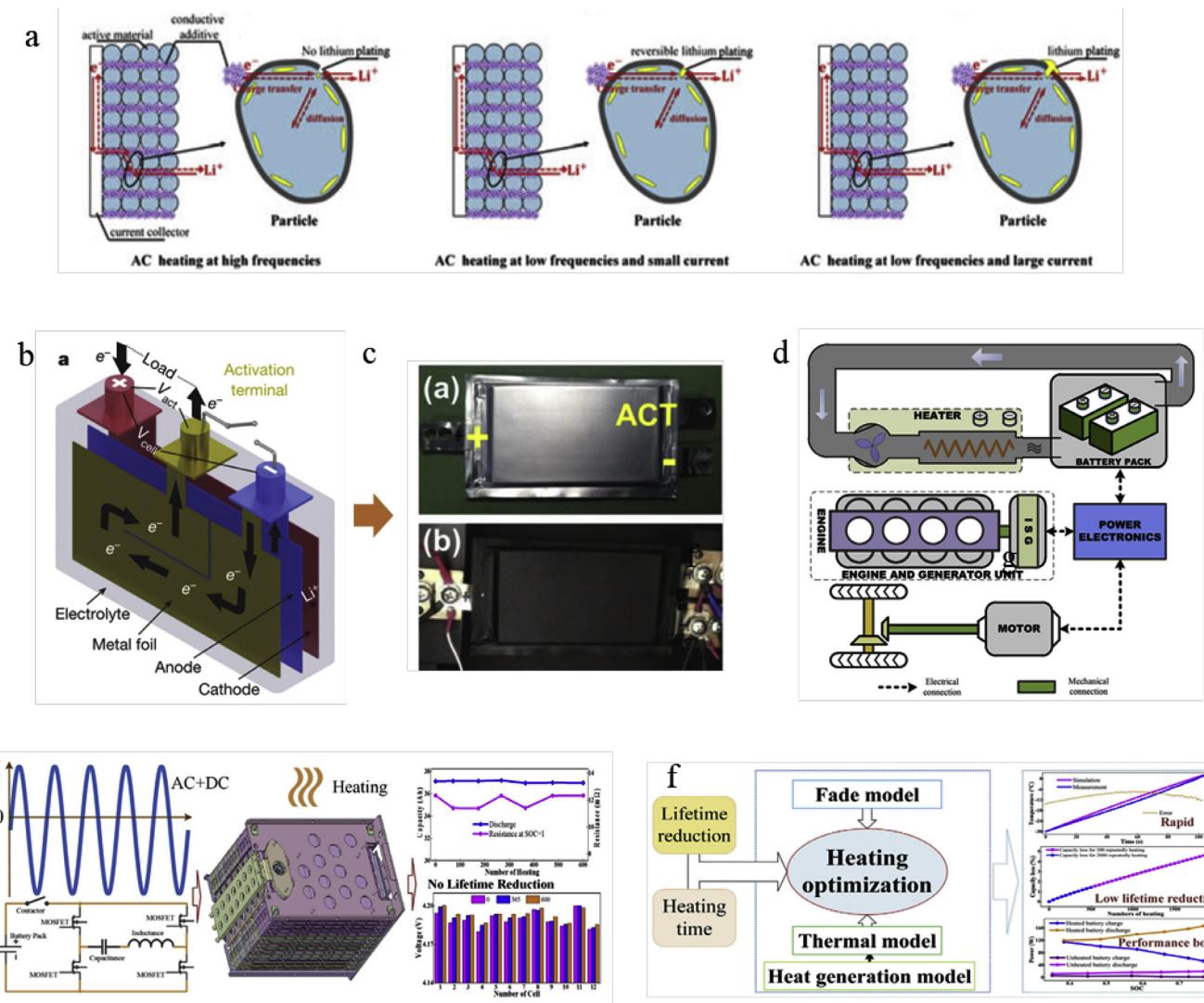


Fig. 10. Heating strategy for lithium ion battery at low temperature. **a**, The influence of AC heating on Li plating. **b, c**, self-heating strategy. **d**, heating strategy in plug-in hybrid electric vehicles. **e**, AC + DC heating for battery pack. **f**, AC heating and optimization by multi-objective genetic algorithm Panels reproduced with permission from: **a**, ref. [139] (Wiley); **b**, ref. [144] (Nature research); **c**, ref. [145] (Elsevier); **d**, ref. [146] (Elsevier); **e**, ref. [142] (Elsevier); **f**, ref. [143] (Elsevier).

[141] developed an echelon internal heating strategy based on AC heating strategy, which can balance the heat generation rate and degradation on battery lifetime. Direct current (DC) heating is also an efficient way to heat batteries, due to the simple implementation and high heat generation. Ruan and Sun et al. [142] proposed AC + DC heating strategy to prevent lithium ion deposition (Fig. 10e). In their work, a soft-switching circuit was designed to heat the battery pack, and the lifetime was further enhanced. After that, they optimized the internal-heating strategy for lithium ion battery at low temperature by using multi-objective genetic algorithm (Fig. 10f), and the performance is still excellent [143]. Wang et al. [144] proposed a novel self-heating strategy to heat lithium ion battery by using a third electrode. They designed a novel structure inside lithium ion battery to heat batteries to normal temperature with only a few seconds. And after that, a series of works were conducted to investigate the performance of self-heating strategy (Fig. 10b and c) [145]. Wang and song et al. [146] evaluated the fuel cost electricity cost and

battery degradation cost of plug-in hybrid electric vehicles at low temperature (Fig. 10d). Their work exhibited that the preheating can decrease the operating cost in the driving cycles.

6.2. BTM at normal state

6.2.1. Traditional strategy for BTM

Air cooling. Considering the cost and space limitations, air cooling has been widely used to control the maximum temperature and local temperature difference of battery pack in some automotive companies [147,148]. The effects of cooling conditions and pack configuration on the temperature of prismatic battery pack should be further investigate how to maintain operating temperature by designing proper battery configuration and choosing proper cooling systems [149]. As shown in Fig. 11, air cooling, including natural air cooling and forced air cooling, is proved to be an effective method for BTM in lithium ion battery pack

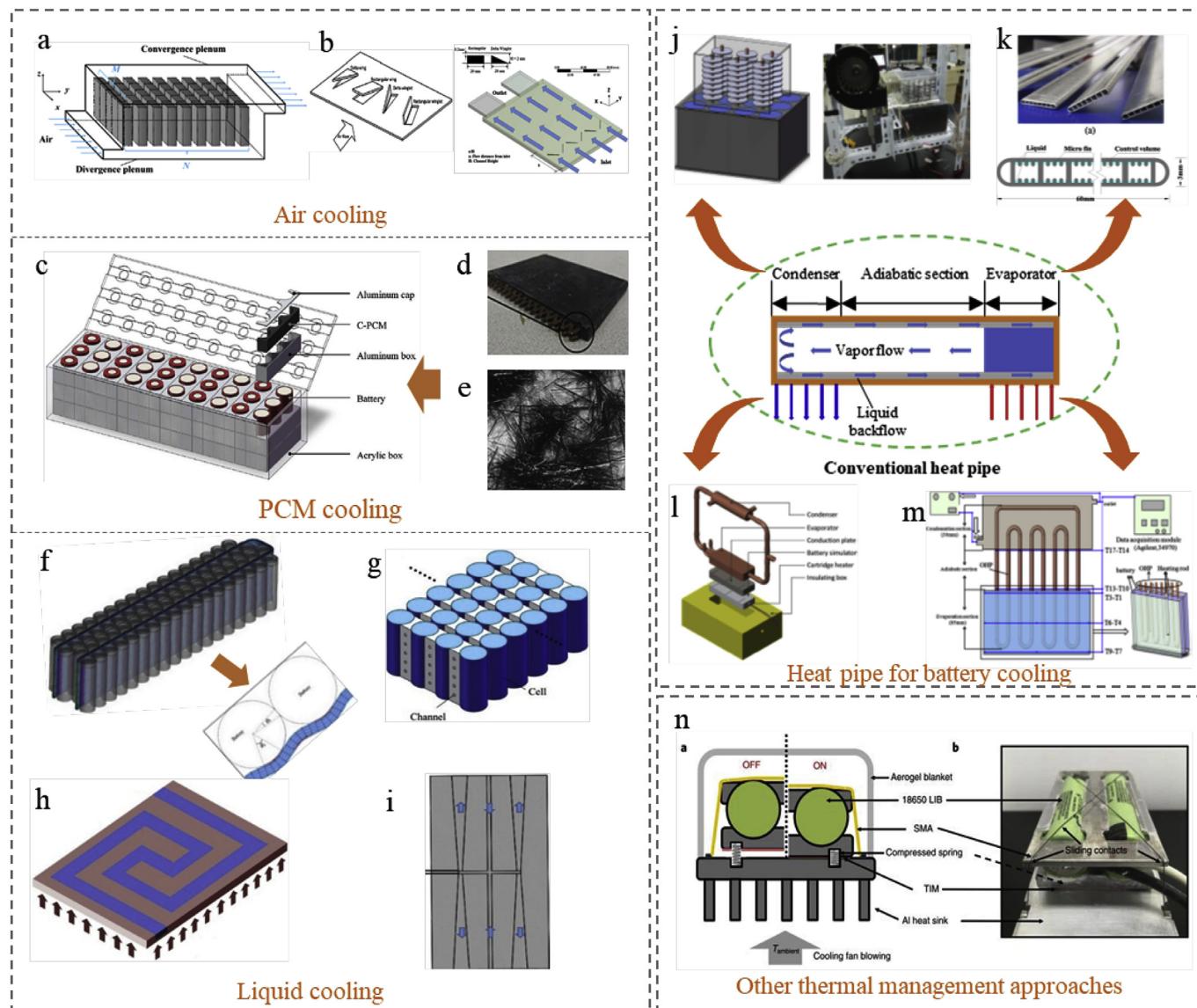


Fig. 11. Traditional strategy for BTM. **a**, Schematic of the parallel air-cooled BTM system. **b**, Vortex generator for enhancing air cooling performance. **c**, Passive BTM system (using phase change materials). **d**, Phase change materials plate. **e**, Carbon fibers in phase change materials. **f**, U-tube channel. **g**, The microchannels with changing contact surface Al block. **h**, Serpentine-channel. **i**, wedge-shape channels. **j**, heat pipe BTM coupled with phase change materials. **k**, Micro heat pipe array. **l**, Loop heat pipe for BTM. **m**, oscillating heat pipe. **n**, Passive battery thermal regulator based on shape memory alloy. Panels reproduced with permission from: **a**, ref. [155] (MDPI); **b**, ref. [204] (Elsevier); **c**, ref [176] (Elsevier); **d**, ref [185] (Elsevier); **e**, ref [186] (Elsevier); **f**, ref [121] (Elsevier); **g**, ref [156] (Elsevier); **h**, ref [192,193] (Elsevier); **i**, ref [201] (Elsevier); **j**, ref [205] (Elsevier); **k**, ref [206,207] (Elsevier); **l**, ref [208] (Elsevier); **m**, ref [209] (Elsevier); **n**, ref [210] (Nature Research).

[150]. Designing an effective way to control thermal performance channel, inlet and outlet in battery is necessary for controlling temperature rise and temperature difference [151]. Zhou et al. [152] developed an air distribution pipe to control the air flow inside battery pack, and the orifice parameters were optimized. Besides, in order to optimize the thermal management performance, the parameters, such as gap between cells [153] and spatial configurations [154], are deserved to be further developed in battery pack. Chen and Wang et al. [155–158] conducted a series of work to optimize the structure of battery pack, including arranging the widths of inlet and outlet, flow rate, the angles of plenums, and the cell spacing. Their work is meaningful for the further development of air cooling strategies in a more smart way (Fig. 11a). Fan et al. [159] and Mahamud et al. [160] studied the effects of several parameters on rectangular and cylindrical battery modules through 3D and 2D numerical simulation, respectively. Among which, Mahamud proposed a novel reciprocating air cooling style, contributing to reduction of temperature difference. For decreasing temperature difference in battery pack, flow in the channel also can be carefully controlled. Kang et al. [161] designed a reverse layered air flow channel to improve temperature consistency of battery pack. In order to improve heat transfer efficiency, metal foam was employed in the air flow channel and on the two sides of battery to enhance heat transfer and realize temperature control [162,163]. And apart from metal foam, heat sink is also a better choice for enhancing heat transfer and controlling temperature rise [164].

PCM cooling. The purpose of using PCM for BTM is to absorb the heat generated through latent heat passively and efficiently. The temperature of PCM can be maintained in small range before the finish of phase transformation, which can improve the heat transfer temperature difference and therefore enhance the heat transfer rate. Related research status of PCM based BTM has been reviewed by Ref. [165] and Ref. [11] systematically and comprehensively. Al-Hallaj group [166–168] firstly put forward the method of PCM based BTM and verified the feasibility through testing cylindrical battery pack with PCM under different conditions. Dinner's group [169–172] paid much attention on the field of PCM based thermal management for prismatic batteries and carried out a large number of experimental studies and modeling researches. Also, a pack level passive thermal management system has been designed and experimentally studied. Rao group [173–177] took advantage of experiment or simulation method to study BTM with PCM from different research ideas, which includes different PCM, cooling or heating methods, battery monomer and battery module, different battery shapes and so on. In particular, an improved BTM system based on the composite phase change material was developed to solve the temperature uniformity issue and leakage problem [176]. Zhang group [178–181] focused on the performance of PCM used for BTM and continuously optimize the properties of PCM, including anti-leakage, anti-volume-change, and low density. Battery pack with those PCMs was set up and tested as well. Parhizi et al. [182] carries out iterative heat transfer analysis of PCM based BTM and pointed out the key design trade-off between discharge rate and energy storage density.

In order to enhance the heat transfer performance of PCM based BTM, the thermal conductance enhancement of PCM became the research hotspot. Many porous materials or added particles with high conductive are tried to composite with PCM (Fig. 11d and e). Porous materials, such as copper foam [131], nickel foam [183,184] and copper mesh [185], can increase thermal management performance of those BTM systems. Carbon materials, like carbon fiber^{186 187}, graphite and carbon nanotube [188], can be good additives due to stable properties and relatively high thermal conductivity. Other strategies, such as silica and Al-honeycomb panels [189], skeleton structure [190] and copper fiber [191], can be used to enhance thermal conductivity of PCM for a better thermal management performance.

Liquid cooling. Liquid cooling has the advantage of high heat transfer rate in spite of complex system configuration. It has the potential of solving the heat dissipation problem of battery pack under extreme conditions. Related research status of Liquid based BTM has been

reviewed by Refs. [137] systematically and comprehensively. Conventional liquid cooling is realized by working fluid flowing through a heat absorbing plate, or called cooling plate, in contact with the heat source. Most work focused on the optimization of flow structure inside the cooling plate. Some representative work will be introduced below. For prismatic battery, the cooling plate is easy to contact with the flat surface. The sandwich structure for the cold plate and the battery monomer as shown in Fig. 11f–i is the common configuration. Jarrett et al. [192,193] designed a serpentine-channel cooling plate and accessed the working performance through computational fluid dynamics method. Optimization is also carried out to make a tradeoff between the heat transfer rate and pressure drop loss. Dincer et al. [194,195] investigated the temperature distributions of a prismatic Li-ion battery cooled by liquid cooling plate and then established a series connected battery pack assembled with the same cooling plate to reveal the thermal management performance in pack level. The battery pack can be maintained within the required temperature range under a wide range of conditions. Zhang et al. [196,197] designed and developed a new liquid cooling strategy based on thermal silica plates for large capacity power battery pack. Similarly, the performance of monomer level and pack level were both explored. Deng et al. [198,199] designed a serpentine-channel cooling plate and a straight wide-flat-channel cooling plate for BTM. For the straight-channel case, parametric study using orthogonal array method was performed to analyze the influence of four parameters on the thermal management performance. It was found that the channel number has most obvious effect, coolant velocity is second, and pipe height has the minimal effect. Lee et al. [200] proposed a ultra-thin mini-channel cooling plate with novel channel structure. The simple configuration of oblique cuts across the straight fins can destroy the developed hydrodynamic boundary layer and enhance the heat transfer ability with minimal extra pressure drop loss. Rao et al. [12,201] paid attention to straight mini-channel cooling plate and discussed the influence of internal flow direction. Furthermore, the configuration of channel was optimized. For cylindrical battery, the cooling surface is difficult to contact with the curved surface of the battery. Related research on the indirect liquid cooling of cylindrical battery is relative little. Rao et al. [201–203] successively proposed two cooling structures for cylindrical BTM, the mini-channel liquid cooled cylinder structure and the aluminum cooling block with variable contact surface and straight mini channel. In general, increasing the contacting area can significantly drop the maximum temperature but reduce the temperature uniformity.

Comparative analysis for different types of batteries. Liquid cooling is considered as a much more efficiency strategy for BTM, but for different types of batteries, the BTM method can be different. (I) Cylindrical battery. There will be obvious gap between cylindrical batteries, and the geometric size of cylindrical batteries is relatively small. Hence air cooling is a suitable choice for cylindrical battery [151,152,211]. PCM with enhanced thermal properties is a potential candidate of BTM for cylindrical battery, because of its flexible construction for battery pack and the excellent ability of temperature regulation and control [212]. However the sustainable temperature control performance of PCM is insufficient, which means that other methods should be applied to adjust the heat accumulation in PCM and battery pack [169,205]. Liquid cooling method for cylindrical battery should be further considered the contact area between ducts and batteries. Hence, the flexible channels can be an alternative candidate for effective thermal management in battery pack [211,213,214]. (II) Prismatic battery and pouch-type battery. Prismatic and pouch-type battery have similar geometric construction and internal structure. Hence this two kinds of batteries have similar consideration for thermal management strategies [171]. About air cooling, like cylindrical batteries, the gap between batteries, site of the batteries should be considered for a better thermal management performance [157,158]. PCM cooling strategy is also a promising candidate for prismatic and pouch-type batteries due to its flexibility [181,215]. The prismatic and pouch-type battery has a more regular geometric structure. Hence liquid cooling is much suitable for BTM in

prismatic and pouch-type battery pack [194,195]. However, considering the necessity of overall high energy density and long driving mileage, the weight and heat transfer coefficient should be further developed. Hence, mini-channel or ultra-thin mini-channel is more useful for improving both electric performance and thermal management performance [200, 216,217]. In fact, single BTM strategy is not sufficient for overall thermal management. Hence, coupled BTM strategy, such as air-PCM coupled BTM, liquid-PCM coupled BTM etc. deserves to be further developed in battery pack for a much more effective thermal management performance [205].

6.2.2. Heat pipe based BTM

As a high effective heat transfer device, heat pipe has been used in BTM system for its high thermal conductivity, flexibility and low cost. The work principle of conventional heat pipes is shown in Fig. 11. The conventional heat pipe has a closed-shell with wick structure inside. When heat pipes were heated, the working fluid inside evaporated at evaporation section and condensed at condensation section, and then the condensed fluid would be transferred back to evaporator through wick structure. Large amount of heating could be transferred through this cyclic process. As compared to conventional liquid cooled systems, BTMS based on heat pipes could provide better cell/module temperature uniformity, less complicated design and a safer system (no leakage issues in high voltage areas). Because of these advantages shown above, many researchers did related works about thermal management of single battery cell, battery unit and battery system.

Due to poor thermal conductivity of Li-ion cells, traditional cooling methods like air cooling on the cell surface do not effectively access and cool the core [218]. Shah et al. [218] have proposed a heat pipe cooling method for single cell by inserting a heat pipe in the axis to achieve core cooling. The results showed that this method could achieve 18–20 °C temperature reduction. Worwood et al. [219] have also proposed a similar approach to lower the thermal resistance for heat transport through the inside of the cell by employing the heat pipe in the axis and spreader disc in the top and bottom of battery. This approach could make the heat pipe directly contacts the top and bottom portions of the cell material. The results showed that the internal cooling strategy could reduce the cell thermal resistance by up to $67.8 \pm 1.4\%$. However, this approach led to a decrease in energy density and an increase in cell mass. Based on Worwood et al.'s results, the maximum energy density decreases and cell mass increase of 18,650 and 32,113 cells could be 6.0% and 11.7%, respectively.

To avoid the potential energy density decrease, some researchers focused on the outside BTM by heat pipes. Because the smaller system leads to smaller work load, many works focused on a battery unit with a few batteries [220]. Rao et al. [221] and Liu et al. [222,223] have used flat shape sintered heat pipes for rectangular BTM, respectively. Both experimental results showed that the system could maintain the maximum temperature and temperature difference below 40 °C and 5 °C. Liu et al. have also developed a segmented thermal resistance model to accurately analyze the dynamic thermal characteristics of UMHP pack, and the simulation results showed good consistency with experiment results. Liang et al. [224] have also developed a similar heat pipe BTM with changing shape heat pipes that the part between the batteries was flat and outside circular under different cooling strategies. The results show that maximum temperature and the temperature difference of battery can be controlled within desired range using intermittent cooling, which was helpful for power consumption. Zhao et al. [225] have also studied the cooling strategies for heat pipe BTM. Five cooling methods including heat pipes in ambient, heat pipes cooled by horizontal fan, heat pipes cooled by vertical fan, heat pipe cooled by thermostat bath and cooled through wet cooling were used during the process. The results showed that heat pipes with wet cooling had the best performance which could control the maximum temperature lower than 21.5 °C and maximum temperature difference under 0.5 °C under 3C rate.

However, thermal management of battery unit with a few batteries

could not show the real situation faced in electrical vehicles application [226]. Thermal management for the battery pack level need to be studied. Tran et al. [227] employed heat pipes with grooves as capillary structure for battery cooling system under 10C and 12C rate. The heat pipes were placed at the surface of the battery module. The results showed that the HPTMS system could maintain the cells temperature within the optimal range under low ventilation rates and minimized power consumption and noise level because the air velocity did little on system performance enhancement. Then, they developed a battery module thermal management system with flat heat pipes between the wall of the battery module and the heat sink [228]. The results showed that adding heat pipe reduced the thermal resistance of a common heat sink of 30% under natural convection and 20% under low air velocity cooling. Consequently, the cell temperature was kept below 50 °C, which cannot be achieved using heat sink. Smith et al. [229] used sintered heat pipes to build a BTM system to achieve a long distance (300 mm) cooling. The maximum temperature of cells could be limited under 55 °C and the maximum temperature difference could be maintained with in ± 5 °C.

To further reduce the temperature difference, PCM was employed in HPBTMS and obviously reduced the temperature difference [230]. Zhao et al. [205] have designed a PCM/HP-based BTM module and experimentally tested its performance. The heat pipes were vertically placed (axial direction of battery) and cooled by the air flow in their experiments. The results show that the maximum battery temperature and temperature difference can be controlled below 50 °C and 5 °C, respectively. Huang et al. [231] have also designed a PCM/HP-based BTM system with heat pipes horizontally placed (radial direction of battery). The results show that the maximum battery temperature and temperature difference can be controlled below 44 °C and 3 °C, respectively. Faced to thermal runaway situation with big heat generation, Yamada et al. [232] proposed a PCM/HP BTMS and studied the performance experimentally and numerically. In the prototype system, when both the PCM and heat pipes were used, the temperature of the mimic battery was approximately 30 °C lower than when only heat pipes were used from 480 s to 3600 s, after the start time of the experiment.

The research shown above mainly employed conventional heat pipes (sintered heat pipe, grooved heat pipe et al.) as heat transfer devices. With the development of heat pipe technology, some relative new kinds of heat pipes such as micro heat pipe array [206,207], loop heat pipe [208,233], oscillating heat pipe [209,234].

6.2.3. Other approaches

Some other approaches have also been put forward and tried to solve the heat accumulation problem faced by power battery system accounting for the limitations of the above conventional methods. As is known, the heat transfer mechanism of the heat pipe based method is the boiling and condensation in confined space. If the boiling process is confined to the inner part of metal cavity, other issues like contact problem and additional thermal resistance occur. Since a novel way emerged that allowing the working medium contact the power battery directly and boil on the surface. Gils et al. [235] attempted to reveal the ability of homogenization and cooling of boiling process on a battery for the first time. In the experiment, a battery monomer was designed to immerse in non-conducting working fluid and its temperature is controlled by the convection heat transfer process, especially, the boiling process. The result presented that the battery can be thermally homogenized and the process of temperature control can be regulated through changing the vessel pressure. Then, Dincer group [236] further numerically studied the temperature management performance of battery pack through partial and direct-contact boiling. The effect of height of liquid propane relative to the battery was analyzed emphatically. Overall, the direct contact boiling method has been preliminarily proved to have ideal effects of thermal control and temperature uniformity. However, the present research is located in the stage of preliminary exploration. Comprehensive experiment research on the battery pack based on this method is still blank. Also, the efficient system structure considering the

boiling-condensation cycle process is to be designed.

Thermoelectric cooling, also known as semiconductor cooling, is mainly based on the peltier effect of an electrical refrigeration method, with small size, no noise, no vibration and many other advantages. Hence the thermoelectric cooling based BTM method started to be discussed and tested based on the above advantages. Zhang et al. [237] comparatively studied the performance of thermoelectric effect based BTM and forced liquid cooling based BTM through simulation method. Better performance can be obtained through using thermoelectric cooling structure combined with water cooling. Alaoui et al. [238] designed and built a thermoelectric module based thermal management system and compared it with a compatible forced air cooling system. It can be found through the experiment that the proposed thermal management system shows feasibility consuming very little parasitic power when ambient temperature falls within the range of 0–40 °C. Song et al. [239] designed a semiconductor thermoelectric device/phase change materials (PCMs) coupled BTM system and a battery pack model based on it. The factors including semiconductor thermoelectric device arrangement, temperature range of thermal management and cooling/heating power were focused. Some useful simulation results can be gained to guide the BTM system design. Though thermoelectric cooling has the advantage of high cooling performance, it has a serious limitation of low COP which constraint the development.

Evaporation is an efficient way of phase-transformation heat transfer. Convention air-based cooling method is limited by the relative low specific heat capacity and high energy consumption. The combination of evaporation and forced convection heat transfer has the potential to overcome the drawback of low specific heat capacity and decrease the energy consumption. Accordingly, another kind of novel BTM method, that is mist cooling, was proposed and researched by Saw et al. [240]. Comparative studies exhibited that mist cooling can ensure lower and more uniform temperature distribution compared to the dry air cooling, which prove that mist cooling can be a potential solution for the thermal management of power battery pack. However, the effect of mist generator on the system complexity and the cost needs to be assessed. In addition, more influential factors like pack configuration, properties of mist air and flow condition should be taken into considered.

In order to improve the adaptability of BTM system, the controllability of thermal management process is required. Smart functional material can be introduced to the design of BTM to enhance the intelligence of the controllability. Taking the BTM system with cooling function and heating function as instance, the cooling function and the heating function have the opposite requirements. In the cooling process, the battery

should be connected to the outside. In contrast, the battery should be isolated from the outside. On the basis of such requirement, an efficient BTM method containing a passive interfacial thermal regulator based on a shape memory alloy was proposed by Hao et al. [210]. Such novel method can reach excellent effect of process control in extreme environments and expands the usability of power battery. In the future, more new material technologies could be used to improve the performance of thermal management systems.

6.3. BTM for thermal safety at high temperature

6.3.1. Criticality analysis of thermal runaway of lithium ion battery

Criticality analysis is a novel and useful method to evaluate and understand the thermal runaway process from thermal perspective. Huang and Wang et al. [241] employed the Semenov and Thomas model to analyze the criticality of thermal runaway in uniform and non-uniform temperature distribution situations (Fig. 12a). The combination of Semenov and Thomas model can be used to analyze the influence of heat transfer coefficient, battery size and shape to criticality of thermal runaway in battery. The criticality of thermal runaway will increase with the increasing heat transfer coefficient, but decrease with the increasing battery size. Their work demonstrates that as the heat dissipation curve (q_L) is tangent to exothermal curve (q_G), the point E is regarded as criticality point, and when heat release rate is higher than that in point E, the thermal runaway can be triggered. While as the size of batteries becomes smaller and the heat transfer coefficient becomes higher, the criticality will move to point E_1 , increasing thermal safety of lithium ion battery. Meanwhile, the critical temperature based on the thermal balance among temperature-dependent heat generation inside battery, thermal conduction and heat dissipation on the surface of battery will be a useful parameter to guarantee the thermal safety of battery and battery pack. The trade-offs between thermal conduction inside battery and heat transfer outside the battery will have a dramatically effect on the criticality of battery (Fig. 12b) [242].

6.3.2. BTM for preventing propagation and deterioration of thermal runaway

Heat generation in thermal runaway process is quite high because of the fast self-heating rate. As the temperature is higher enough, the combustion and explosion will happen. Hence, a BTM for thermal runaway process to prevent the propagation and deterioration of thermal hazard in battery pack is necessary. The self-heating rate in thermal runaway process is quite high. Hence, it is difficult to control the

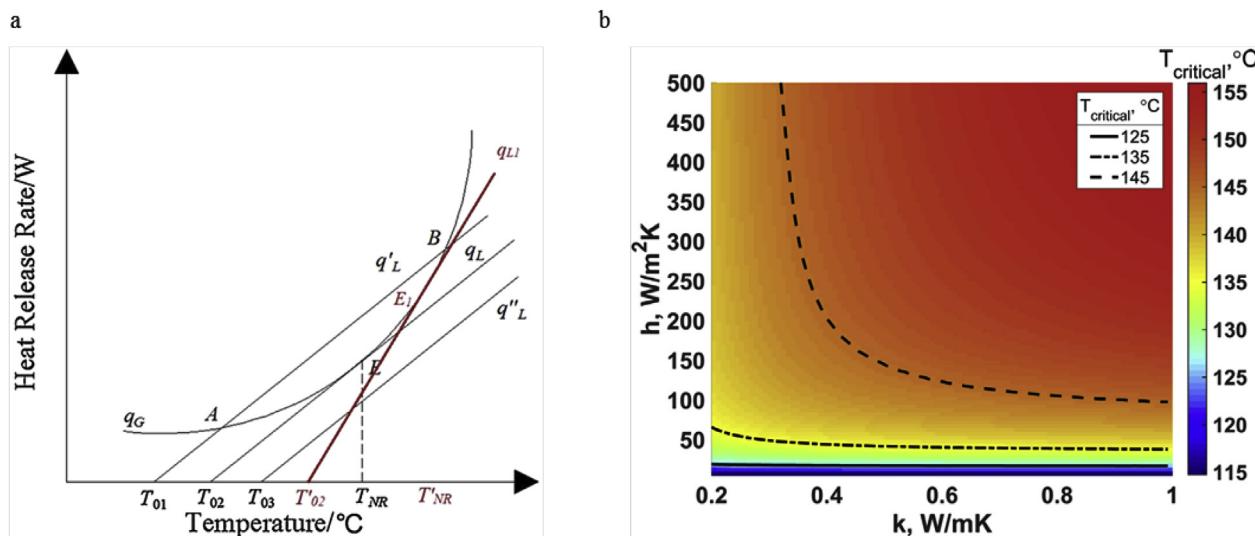


Fig. 12. Thermal analysis in thermal runaway process. **a**, Thermal graph of heat generation and dissipation. **b**, Critical temperature ($T_{critical}$) in heat transfer coefficient (h)-thermal conductivity (k) parameter space. Panels reproduced with permission from: a, ref [241] (Elsevier); b, ref [242] (Elsevier).

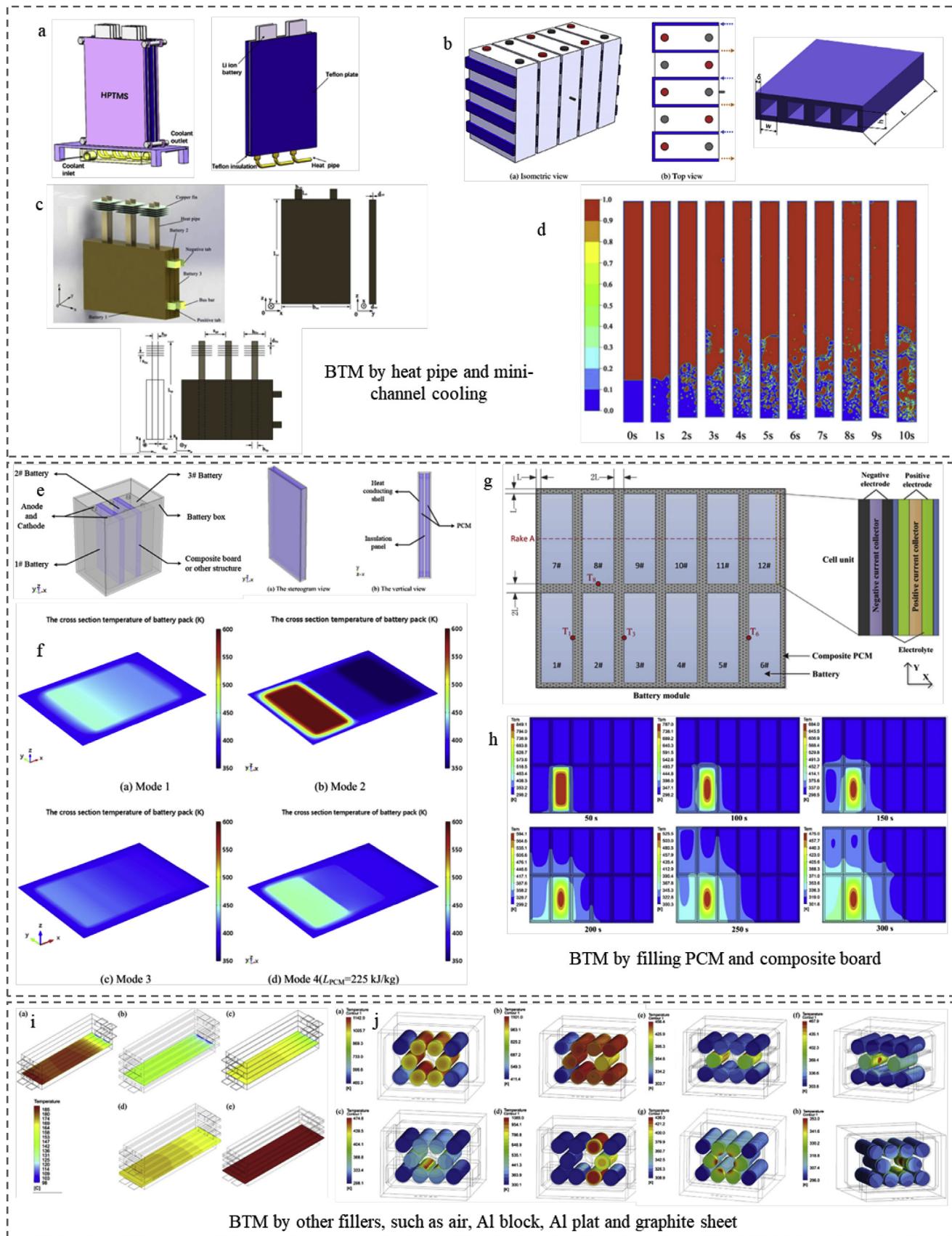


Fig. 13. BTM strategies for thermal runaway. **a-d**, liquid based BTM for thermal runaway process, heat pipe cooling (**a, c** and **d**) and mini-channel cooling (**b**). **e-h**, phase change materials (PCM) and the composites based BTM, PCM based composite board (**e** and **f**), PCM and the composite (**g** and **h**). **i-j**, Filler based BTM, metal plat (**i**) and Al block, Al plat and graphite sheet (**j**) Panels reproduced with permission from: **a**, ref [244] (Elsevier); **b**, ref [245] (Elsevier); **c, d**, ref [243] (Elsevier); **e, f**, ref [246] (Elsevier); **g, h**, ref [247] (Elsevier); **i**, ref [248] (Elsevier); **j**, ref. [249] (Elsevier).

temperature rise in battery pack. The strategies for thermal runaway process mainly include heat pipe, mini-channel, phase change materials and fillers based BTM (Fig. 13). The heat pipe has a much higher thermal conductivity, which can effectively accelerate heat dissipation in battery pack. However, the heat pipe can only prevent the thermal runaway propagation between two adjacent batteries, but cannot prevent the thermal runaway in single battery [243,244]. Meanwhile, the mini-channel can be also used to control thermal runaway in battery pack. While the mini-channel cannot stop the thermal runaway in single cell even as the flow rate is up to 10 L/min, but can prevent thermal runaway propagation between cells [245]. Besides, a composite board that is consisted of phase change materials, insulation panel and heat conductive shell can also prevent the thermal runaway propagation between cells. In order to achieve better thermal performance, the latent heat capacity of phase change materials should up to 1125 kJ/kg, a really high enthalpy [246]. Filling phase change materials into battery pack is a promising method to prevent thermal runway propagation between cells. If the thermal conductivity of phase change materials can be improved (adding materials with higher thermal conductivity), the temperature control ability will be further improved. And the space between cells will be further saved [247]. Air along cannot stop the propagation of thermal hazard, while the heat sink (made by aluminium), graphite composite sheet and Al plate can also be used to prevent thermal runaway propagation in battery pack, meanwhile, thermal resistance between cells is also important to stop the thermal propagation [248]. If the battery failed in the case of side rupture, Al plate will be useless in preventing thermal runaway propagation, while the graphite can stop the propagation with a side ruptured cell [249]. The sprays refrigerant can be used to cut off the heat and combustion once the thermal runaway is about to occur, which is a promising candidate for prevent thermal runaway in cell [250].

7. Outlook

Thermal safety is the crucial aspect for the further development of lithium ion battery. In this paper, the potential inducements with temperature sequence were summarized and the relevant solutions were also reviewed. We have considered the potential inducements at different temperatures, including low temperature (<0 °C), normal temperature (0–90 °C) and high temperature ($>\sim 90$ °C). At low temperature, Li plating and dendrite were considered, which may cause ISC in lithium ion battery. At normal temperature, the heat generation in lithium ion battery may induce thermal runaway. In general, reversible heat generation and irreversible heat generation (ohmic heat generation and polarized heat generation) are the main heat source. While at thermal runaway process, the exothermic reactions are the main factors that induce the heat accumulation and a series of chain exothermic reactions. In recent years, the frequent combustion and explosion in electric vehicles seem to indicate the necessity of more efficient way to prevent the heat generation, accumulation, propagation and to accelerate heat dissipation in battery pack. The material perspective is definitely an effective approach to improve battery safety. While the external electric management system and thermal management system are also necessary to be further developed. There are some potential strategies for battery thermal safety, which is as follows:

From material perspectives, it is necessary to improve the Li-ion diffusion, electronic conductivity, thermal stability, flame retardance of electrode materials, electrolyte, separator and SEI layer to decrease heat generation and deterioration in battery. Improving the decomposition temperature of SEI layer and separator can delay internal short circuit and thermal runaway. Besides, the high thermal conduction inside battery can be an effective way to improve the critical temperature and control the temperature rise. For fast heat dissipation in battery, the thermal properties (thermal conductivity and specific capacity) of battery materials also should be further developed. The critical temperature for thermal runaway is determined by the material properties and heat dissipation efficiency. The improving of critical temperature can also put

off thermal runaway.

In cell level, a more safety cell design is much important for the further development of lithium ion battery. The design should consider the electric performance, heat generation and heat dissipation comprehensively, considering the specific parameters such as thickness, size of battery, size and site of tab etc. Besides, for different temperature conditions, strategies for thermal safety is distinguished. Heating and heat preservation is important for lithium ion battery at low temperature to prevent Li plating and dendrite. Efficient cooling for normal temperature is an effective way to prevent the start of thermal runaway. BTM both in normal state and thermal runaway process is the last ditch for thermal hazard. The reported BTM strategies (air, liquid phase change materials, boiling and heat pipe based strategies) can effectively control the temperature rise at normal state and prevent thermal runaway propagation in battery pack. Considering the limitation of electric system when thermal runaway occurred, passive heat dissipation (phase change materials, boiling and heat pipe etc.) can be a promising candidate for further temperature control.

From the system level, considering the effective response for thermal runaway, the development of electric-thermal detection in battery pack is really necessary. The thermal behavior in battery pack is always induced by electric behavior, which means that electric detection in battery and battery pack can forewarn the possible thermal issue for battery. And the thermal (temperature) detection is a more direct way to monitor the thermal behavior in battery and battery pack. Besides, relevant researches show that pressure detection is also a useful way to forewarn the underlying thermal runaway threat. Hence, a comprehensive detection method is necessary to prevent thermal runaway in battery and battery pack.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgment

This work was supported by the National Natural Science Foundation of China (No. 51776218) and the Natural Science Foundation of Jiangsu Province (No.BK20180083).

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