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An Overview on Thermal Safety Issues of Lithium-ion Batteries for Electric Vehicle Application

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ABSTRACT Lithium-ion batteries (LIBs) are being intensively studied and universally used as power sources for electric vehicle applications. Despite the staggering growth in sales of LIBs worldwide, thermal safety issues still turn out to be the most intolerable pain point, and remain the focus of research for technological improvements. This paper presents a comprehensive overview on thermal safety issues of LIBs, in terms of thermal behavior and thermal runaway modeling and tests for battery cells, and safety management strategies for battery packs. Considering heat generation mechanism and thermal characteristics of LIBs, heat generation, dissipation and accumulation inside a cell are elaborated. The triggering factors leading to thermal runaway are also summarized. Finally, thermal runaway detection and prevention strategies for both cell- and pack-levels are introduced. Different engineering approaches from material refinement and additive adoption to thermal, electrical, and mechanical design are presented for thermal runaway prevention.

INDEX TERMS Electric vehicles, batteries, modeling, calorimetry, safety, thermal runaway, thermal management.

I. INTRODUCTION

LIBs have been extensively used as major power sources for electric vehicles (EVs) due to their intrinsic advantages such as high energy density, no memory effect, long lifespan and design flexibility. A LIB cell consists of a positive electrode and a negative electrode with a separator in between. Lithium ions shuttle between two electrodes during charging/discharging while electrons are forced through external circuitry for power sourcing and sinking as sketched in Fig. 1. The high volumetric and gravimetric energy densities are simultaneously achieved due to the realization of high cell voltage and capacity, resulting from refined material selection and delicate battery design.

Despite the impressive growth in sales of EVs worldwide, thermal safety issues of battery systems are often criticized as the main cause for fatal fire accidents in recent years. On one hand, a battery pack in an EV typically consists of hundreds of thousands of cells connected in parallel and/or series in order to meet the requirements of power and energy. This significantly increases the stored energy inside battery systems, which means enlarged damaging effects when a severe safety issue occurs. On the other hand, LIBs in a vehicle always face harsh working conditions such as vibration and shock, and may encounter overcharge, overheat, short circuit, collision or nail penetration under extreme abuse conditions. This raises the probability of thermal runaway occurrence for onboard battery systems. Overall, thermal safety issues have become a bottleneck for the mass adoption and market penetration of EVs.

Generally, thermal runaway can be triggered by mechanical, electrical or heat abuses. Mechanical abuse is often in the form of penetration or collision, which may incur internal short circuit in a battery cell or cause bus-bar short circuit. Electrical abuse includes overcharge that may cause internal short circuit due to lithium plating and arouse chain secondary exothermic side reactions. Heat abuse is usually

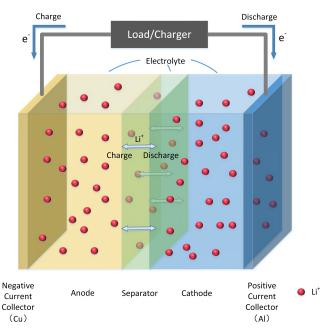


FIGURE 1. Schematic of the Lithium-ion battery.

induced by high environmental temperature and/or ineffective thermal management. Appropriate elevation of LIBs working temperature, can raise battery performance while reducing heat generation. However, extremely high temperature may cause exothermic decomposition of electrodes and electrolyte materials and separator shrinkage that can induce internal short circuit. In actual situations, rather than coming individually, abuse behavior always occurs one after another. Finally, the mentioned abuse behaviors evolve into a fierce and fast heat generation process, resulting in smoke, fire and even explosion.

In this paper, a thorough review of the major thermal safety issues of LIBs is presented, with emphases on thermal behavior and thermal runaway modeling. Safety management strategies to prevent thermal runaway occurrence beforehand and limit the damaging consequence afterwards are also reviewed.

II. HEAT GENERATION MECHANISM AND THERMAL MODELING

LIBs are a typical electrochemical system that works under the principles of electrochemical reactions, mass and charge transfer and energy balance. Heat generation in a battery cell represent a complicated process, where electrochemical reaction rates vary with time, temperature and current distributions [1]. From the viewpoint of safety, thermal runaway accidents mean wrecking the dynamic balance between heat generation and accumulation inside battery cells and heat dissipation with their surroundings. Hence, it is meaningful to cast light on heat generation mechanism and present latest studies on modeling and testing involving the complex thermal process. In this section, heat generation and thermal modeling approaches for LIBs will be briefed, followed by a summary of commonly used experimental methods for battery thermal behavior acquisition.

A. THERMAL MODELING

The heat effect with simultaneous electrochemical reactions was first addressed by Sherfey and Brenner in 1958 [2]. An equation for heat generation rate determination in terms of current fraction, entropy change, and over-potential for each reaction was proposed. Sherfey's work was then recognized and followed by continuous investigations on this frontier by other endeavors described in [3] and [4], until J. Newman and his group made remarkable contributions regarding thermal modeling of LIBs. The seminal work was conducted by Bernardi et al. [5] to predict the heat generation of LIBs via a thermodynamic energy balance equation. In the energy balance equation, the battery temperature was assumed to be uniform and only determined by internal reactions, heat capacity, phase changes, mixing and heat transfer. Rao and Newman [6] presented a heat generation calculation method for an insertion battery system using a general energy-balance equation and a local heat-generation method. Thomas and Newman [7] investigated the influence of the entropy, irreversible resistance and heat of mixing on heat generation rate. Doyle et al. [8] simulated the galvanostatic charge and discharge of LIBs using the concentrated solution theory to describe the transport properties in the electrolyte. Based on the model proposed by Doyle et al. [8], Botte et al. [9] further developed a thermal model that accounted for the decomposition reaction of the carbon anode in the energy balance. Kumaresan et al. [10] presented a thermal model that could predict discharge performance at different operating temperatures, which parameters were identified based on the experimental profiles at different working temperatures and discharge rates. Cai and White [11] utilized a commercial simulation software COMSOL Multiphysics to study the thermal behavior of LIBs during a galvanostatic discharge process with or without additional pulses.

Aiming at vehicular applications, C. Wang and his group developed multiple electrochemical-thermal-coupled models based on the multi-dimensional modeling approach. Gu and Wang [12] put forward a thermal-electrochemicalcoupled model using the volume-averaging method, which coupled the energy balance equation with a multi-phase electrochemical model via the heat generation and temperaturedependent physicochemical properties. The established model was multi-dimensional and capable of predicting the average temperature as well as the temperature distribution inside a cell. Considering reversible, irreversible and ohmic heat in solid and liquid phases, Srinivasan and Wang [13] further incorporated the temperature dependence of transport, kinetics and mass-transfer parameters in terms of Arrhenius expressions into the previous built electrochemical-thermalcoupled model. Fang et al. [14] validated the fidelity of this model under the constant-current and pulsing loads that emulated practical working conditions of hybrid electric vehicles (HEVs). Ferguson and Bazant [15] applied the porous electrode theory to non-ideal active materials, and developed a complete battery model on the basis of non-equilibrium thermodynamics.

For large-format battery cells, Kim *et al.* [16]–[18] studied the influence of electrode configuration including the aspect ratio of electrodes and the placing of current connecting tabs on thermal behavior of LIBs, and proposed a two-dimensional model to predict temperature distributions of LIBs based on the modeling of the potential and current density distribution on electrodes. Lee *et al.* [19] developed a numerical model for a 20-Ah large-format cylindrical cell using the Multi-Scale Multi-Domain (MSMD) model framework, where the wound potential-pair continuum (WPPC) model was presented as a sub-model to depict the heat and electron transfer across the longitudinal axis of the cell domain.

Zou *et al.* [20]–[22] proposed a framework for simplification of PDE-based LIB models that simultaneously described the coupled electrical, thermal and aging dynamics. Singular perturbations, averaging and order reduction techniques were utilized for model simplification purpose. A multi-time scale estimation algorithm adopting the extended Kalman filter (EKF) was further developed and validated by simulation results.

The thermal modeling of LIBs has been introduced and studied for decades. Electrochemical-thermal-coupled and multi-dimensional modeling methods have emerged to provide more details for the underlying electrochemical reactions inside battery cells. Control-oriented modeling gradually becomes the research hotspot, with the potential to be used in enabling battery management systems (BMSs).

B. EXPERIMENTS FOR THERMAL BEHAVIOR INVESTIGATION

Despite physical-based thermal modeling can give insights and theoretical explanations about heat generation process inside a LIB cell, experimental approaches are also necessarily used for model parameterization and validation. Calorimetry techniques are a basic temperature or heat flux measurement method under a certain thermal boundary condition. Heat generation of the measured object can be acquired through the heat balance equation. In this section, two widely-used calorimetry techniques, i.e., Accelerating Rate Calorimetry (ARC) and Isothermal Heat Conduction Calorimetry (IHCC), are elaborated.

1) ARC-BASED EXPERIMENTS

ARC is a thermal analyzing instrument/method for heat generation determination. It can measure the generated heat of an object by establishing an adiabatic environment. Traditional ARCs are proper and convenient for a small dose of chemicals. However, in order to test LIBs, especially large-format ones, large-scale ARCs are being developed and employed [23]. Heat generation rate can be calculated by the following energy balance equation as

$$q = MC_p \frac{dT}{dt} + hA(T_{surf} - T_{well})$$
(1)

The left-hand term is the amount of heat generated by a battery cell or module. The first term on the right-hand is the heat stored in the measured object, and the last term is the heat convection between the surface of the measured object and the calorimeter well.

Hong et al. [24] established an accelerated rate calorimeter-cycler test bench to analyze the thermal behavior of a commercial 18650 lithium-ion cell, and collected experimental data of heat dissipation during discharge and charge at various rates. Al Hallaj et al. [25], [26] presented an electrochemical calorimetric test method (ARC-Arbin) to conduct experiments on several LIBs with different chemistries. They acquired instantaneous heat generation rate and defined its correlation with cell impedance and discharge rate. Zhang et al. [27] utilized the ARC to measure the specific heat capacity of a battery cell and validated various heat generation estimation methods for large-format pouch cells based on the ARC result of battery heat generation rate [28]. Schuster et al. [29] tested 40Ah commercial NCM pouch cells under the adiabatic environment created with an ARC at various charging/discharging rates. The adiabatic tests were designed to simulate extreme heat generation conditions without cooling in order to maximize the self-heating effects.

2) IHCC BASED EXPERIMENTS

IHCC provides an isothermal test environment to ensure a constant temperature of the tested battery. The isothermal boundary is always realized by a temperature-fixed sink that directly contacts with battery cells or by a one-point temperature-controlled chamber that allows heat convection between the chamber well and battery cells. There are two types of IHCCs that are widely used in experimentation. One is the micro-calorimetry that places two battery cells in two separate heat sinks, i.e. a sample cell and a reference cell. The other is a one-heat-sink design, which uses Peltier phenomenon or power compensation method to control the sink temperature and measure the heat flux.

Bang *et al.* [30] employed an isothermal micro-calorimetry to investigate thermal profiles of LIBs with LiMn₂O₄ and LiAl_{0.17}Mn_{1.83}O_{3.97}S_{0.03} cathodes, where the heat flow rate obtained from the IHCC was used as a benchmark for heat calculation. Kobayashi *et al.* [31] adopted a joint electrochemical and calorimetric measurement composed of a one-sink micro-calorimeter and an X-ray diffraction facility to investigate the irreversible thermal behavior of LiMn₂O₄/C under cycling conditions and elevated temperatures. Researchers realized precise electrochemical calorimetry of LiCoO₂/C battery based on a dual-sink isothermal calorimeter and half-cells of each electrode. Results showed that thermal peaks that indicated phase transition could be easily measured by IHCC [32]. Thomas and Newman [7] used a dual-sink micro-calorimeter with an accuracy of $\pm 2\mu$ W to reveal the dependence of heat generation on time. Lu and Parakash [33] set up an *in-situ* measurement using a dual-sink isothermal micro-calorimeter to test heat generation rate under room temperature. Furthermore, Lu *et al.* [34] conducted experiments on LiNi_{0.8}Co_{0.2}O₂/Li and meso-carbon micro-bead (MCMB)/Li half cells and LiNi_{0.8}Co_{0.2}O₂/MCMB full cells with electrochemicalmicro-calorimeter to examine the modelling accuracy at low and moderate current levels. Downie *et al.* [35] took advantage of the isothermal micro-calorimetry technique to determine the effects of electrolyte additives, voltage-dependent parasitic heat flow [36] and time-dependent parasitic heat flow in LIBs [37].

In short, experimental-based studies on the thermal behavior of LIBs mainly involve the application of calorimetry. Calorimeters can be specially designed and configured for testing large-format commercial automotive batteries. It is worth noting that ARC has also become a popular tool for battery thermal runaway tests due to the working principle of self-heating and its availability under extreme test occasions.

III. THERMAL RUNAWAY MODELING AND SAFETY TESTS

Due to the harsh working conditions in EV applications, the issues of thermal, electrical and/or mechanical abuses may lead to thermal runaway events and thus significantly compromise the safety of battery systems and their host vehicles. Thermal runaway may happen inside a battery cell when elevated temperature incurs chain exothermic reactions, which further raises temperature and triggers more deleterious reactions. Such elevated temperature can come from gradually accumulated heat that fails to be timely dissipated, or externally-induced drastic reactions in a short period of time.

In this section, the modeling of thermal runaway under various triggering conditions will be reviewed, with a discussion on experimental approaches for thermal, electrical and mechanical abuses.

A. THERMAL RUNAWAY MODELING

Spotnitz and Franklin [38] systematically explored possible reactions inside a battery cell when experiencing thermal runaway. Common abuse conditions were discussed and grouped into five categories:

- (a) Oven test: exposing the battery cell to a step-elevated high temperature.
- (b) Short circuit: low resistance connection across the terminals of the battery cell, or internal short circuit between layers that possess different potentials.
- (c) Overcharge test: charging current is forced through the battery cell up to a certain voltage limit.
- (d) Nail: a nail penetration through the battery cell at a certain rate.
- (e) Crush test: a bar pressing towards the battery cell until incurring internal short circuit.

It is worth mentioning that the definition of "short circuit" in (b) is no longer constrained to refer to the external

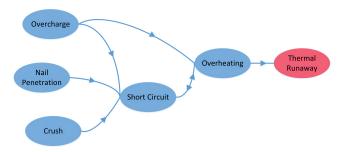


FIGURE 2. Relationship of different abuse conditions.

short circuit. In fact, tremendous endeavors have been made to study the internal short circuit (ISC) that is regarded as a major cause for self-induced thermal runaway of LIBs. Besides, the above-mentioned abuse conditions are categorized by their own characteristics; they may happen sequentially or even simultaneously. Actually, their mutual relationship is complex and heavily intertwined as shown in Fig. 2. Mechanical abuse including nail penetration and crush may cause the physical contact of electrodes and even result in short circuit between the high and low potential parts due to the invading conductive substance. Electrical abuse often characterized by overcharge can not only lead to ISC induced by lithium dendrites, but also intensify the side reactions that may give rise to overheating. Short circuit can originate from internal or external heating of battery cells since high temperatures would melt down the separator. Therefore, short circuit is a key factor causing overheating of LIBs.

Spotnitz and Franklin built a 1-D model to present the underlying exothermic reactions during the process of thermal runaway, which can be summarized as follows:

- (a) Solid electrolyte interface (SEI) layer decomposition: at 90-120°C.
- (b) Reaction of intercalated lithium ions with electrolyte: >120°C.
- (c) Reaction of intercalated lithium with fluorinated binder.
- (d) Electrolyte decomposition: $>200^{\circ}C$.
- (e) Positive active material decomposition.
- *(f)* Upon overcharging, metallic lithium is formed which can react with the electrolyte.
- (g) Lithium metal can react with the binder.
- (h) Discharge of the battery releases heat due to entropy changes, over-potentials, and ohmic resistances.

In this section, according to the categories of abuse conditions, the state-of-the-art models for thermal runaway will be introduced.

1) OVERHEATING

As depicted in Fig. 2, overheating can be triggered by high ambient temperature or other abuse behaviors and represent a typical embodiment of thermal runaway propagation mode.

Based on reaction kinetics and thermal properties of battery cells from literature, Hatchard *et al.* [39] proposed a predictive model for simulating the response of new cell sizes and electrode materials in oven exposure testing. Aiming at describing the thermal abuse behavior of large-format LIBs for automotive applications, Kim *et al.* [40] developed a three-dimensional model considering the shape and dimension of cell components and the spatial distribution of materials and temperature. Guo *et al.* [41] developed a 3-D thermal model for oven exposure testing, considering battery geometry, heat generation and multiple heat transfer processes such as heat conduction, convection and heat dissipation.

2) INTERNAL SHORT CIRCUIT

External short circuit can be easily executed and applied to battery safety tests. However, considered a primary cause for self-induced thermal runaway, ISC may have different causal factors. As a result, the existing studies have focused on ISC-related thermal runaway modeling.

Maleki and Howard [42] proposed a model by employing measured electrical, electrochemical, and chemical heat generation profiles to simulate ISC, which indicated that the thermal runaway was dependent on:

- (a) localized heating energy of the ISC spot and its duration.
- (b) separator shrinkage, melting point and propagation.
- (c) overall cell temperature rise.

Santhanagopalan et al. [43] built an electrochemicalthermal model to account for their experimental observations under several short circuit scenarios. This model considered the influence of model parameter variations including SOC and initial temperature. Fang et al. [44] developed a 3-D electrochemical-thermal model to describe the ISC events in a 1 Ah lithium-ion battery, and pointed out that heat generation in the reaction of aluminum current collector and anode was most remarkable. Zhao et al. [45] raised a 3-D electrochemical-thermal coupled model to scrutinize the ISC process in a stacked-electrode large-format Li-ion cell. Short circuit resistance and the number of shorted electrode layers were shown to play the most significant role. Yamauchi et al. [46] also proposed an ISC model for a lithium secondary battery with the emphasis on analyzing the structure-dependent properties from the perspective of balancing energy density and safety. Wang et al. [47] developed an electrochemical-thermal-coupled model to simulate the entire process from self-heating to thermal runaway. Through selecting separators with different meltingdown temperatures, the experimental results indicated that the ISC-induced thermal runaway can be mainly ascribed to separator melt-down when low-melting-point separators used and to electrodes decomposition when high-meltingpoint separators employed.

3) OVERCHARGE

Spotniz and Franklin [38] reckoned that overcharge would be a dangerous abuse due to the influx of excess energy exceeding the amount that the battery can safely accept. Arora *et al.* [48] revealed that overcharge of the negative electrode that causes lithium deposition would greatly decrease the cycle life of LIBs and pose serious safety hazard, and accordingly proposed a model to predict lithium deposition on the negative electrode during charge/overcharge. Thomas-Alyea et al. [49] developed a continuum-scale electrochemical model to investigate the effectiveness of electroactive polymers for protecting LIBs under overcharge condition. Wu et al. [50] built a complicated electrochemicalthermal-coupled model for valve-regulated lead-acid batteries (VRLAs), and studied the key parameters during overcharge. Despite VRLA chemicals are different from that in LIBs, modeling and abuse behavior simulation methods can provide valuable insights for LIBs-based investigations. Perkins et al. [51] presented a control-oriented reducedorder model to capture lithium deposition, resistance rise and capacity loss during overcharge, and found a way to use a physical-based model for practical control implementation. Chen et al. [52] looked into the performance of three redox shuttle electrolyte additives for protecting a Li-ion cell from overcharge abuse and established an Eb(ER) computational model for analyzing the stability of redox shuttles in cells.

4) NAIL PENETRATION

Nail penetration is a typical mechanical abuse and constitutes an integral part of compulsory battery test standards. It serves to simulate the possible impact of batteries in accidental events. For instance, nail penetration can simultaneously lead to mechanical destruction and ISC of LIBs. In this section, methodologies for nail penetration testing and modeling will be briefly introduced.

Zhao *et al.* [53] developed a 3-D multi-scale electrochemical-thermal-coupled model for a large-format LIB in order to investigate the nail penetration process. A parametric analysis was carried out and the result showed a strong coupling relationship between thermal response and electrochemical reactions. In addition, the thermal behavior was also strongly influenced by short circuit resistance, nail diameter, nail thermal conductivity and cell capacity.

M. Ouyang and his group carried out a series of studies on thermal runaway of large-format LIBs. The ISC was considered the main contributor that directly induces thermal runaway of LIBs, and accordingly ISC fault detection methods were intensively researched. For nail penetrationinduced ISC, Feng *et al.* [54] developed a lumped thermal model that was able to predict thermal runaway of a 25Ah NCM battery pack. Again, Feng *et al.* [55] built a 3-D thermal runaway model for a 6-cell large-format LIB module to study the thermal runaway propagation behavior inside a battery pack, caused by nail penetration. Empirical equations and equivalent thermal resistance layer were included for model simplification.

Chiu *et al.* [56] combined an electrochemical model and thermal runaway equations to represent mass and charge transport behaviors and delineate temperature distribution characteristics during nail penetration process. Yamauchi *et al.* [46] found that thermal runaway of LIBs caused by nail penetration was primarily determined by the total Joule heat generated by the ISC current.

5) CRUSH

Crush is one of the most common abuse modes for LIBs in car collision or rollover accidents. Similar to nail penetration, crush can easily trigger ISC. Sahraei *et al.* [57] built a finite element (FE) model composed of shell elements for casing and solid elements for active materials in the software of LS-DYNA. Simulation results showed a good agreement with experiments. Then, the established FE model was utilized to determine the onset of thermal runaway on the basis of stress values.

In order to depict the interaction between the mechanical and electrical behaviors during crushing, Zhang *et al.* [58] developed a coupled mechanical-electrical-thermal model to simulate the mechanical abuse by coupling mechanical stress with displacement/deformation. This model calculated the mechanical deformation in LS-DYNA using the FE method, and then the deformed mesh was employed to refining the electrical and thermal equations for better accuracy.

B. BATTERY ABUSE EXPERIMENTAL TESTS

Abuse tests are designed to reproduce the impacts of car accidental events on battery systems. Many codes and standards have been released by international organizations, laboratories and governments. Among these are the hazardous materials transport regulations developed by the United Nations (UN) (UN 38.3.4), Code of Federal Regulations (CFR) (49 CFR), the consumer electronics safety standards developed by UL (UL 1642 and UL 2054) and more recently by the Institute of Electrical and Electronics Engineers (IEEE) (IEEE 1725 and IEEE 1625), the International Electrotechnical Commission (IEC) (CEI/IEC 62133 and IEC 62281), and the GB (National Standard of China) (GB/T 31467.3 and GB/T 31485), etc.

Existing battery abuse tests can be divided into three categories [38]:

- (a) Thermal abuse tests: simulated fuel fires, thermal stability, overheat, etc.
- (b) Electrical abuse tests: short circuit, overcharge, overdischarge, etc.
- (c) Mechanical abuse tests: mechanical shock, vibration, drop, nail penetration, immersion, crush, etc.

1) THERMAL ABUSE TESTS

Thermal abuse test is often performed by oven exposure testing. While, calorimetry is also widely employed for quantitative analysis.

Chen *et al.* [59] recorded an explosion event by conducting a 150°C oven exposure testing for a LiCoO₂/C battery. The analysis results manifested that the SEI layer decomposition started at about 70°C and the anode/electrolyte reaction began at about 120°C. It was pointed out that the anode/electrolyte reaction churn out enough heat to further trigger the electrolyte and cathode decomposition reactions which gave off amounts of heat and gas and even caused an explosion. Feng *et al.* [23] tested a 25Ah large-format prismatic NCM LIB cell in an adiabatic environment using extended volume accelerating rate calorimetry (EV-ARC). Temperature, voltage and internal resistance profiles before and during thermal runaway process were recorded and analyzed. Significant temperature difference between the surface and the core, voltage drop and internal resistance sequence of the battery cell were observed. Feng *et al.* [60] modified the EV-ARC to heat the tested batteries to a specific temperature well before thermal runaway to observe the phenomena of separator melting, capacity fade and recovery.

Q. Wang and his group also investigated thermal behavior and reaction dynamics of LIBs under various abuse conditions from the perspective safety engineering [61], [62], [69], [119]. Wang *et al.* [61] conducted battery cycling experiments in an adiabatic environment and found out that higher current rates and increased initial temperatures would markedly increase thermal hazards. Regarding the explosion in a thermal runaway event as a catastrophic phenomenon, they utilized the energy conservation equations to describe the heat generation process during discharging [62]. Taking advantage of the dimensionless method, a swallowtail catastrophe potential function was presented to help control synthesis for thermal runaway prevention.

2) ELECTRICAL ABUSE TESTS

Electrical abuse tests including external short circuit, overcharge and overdischarge can be readily conducted using professional battery testing facilities. Therein, the overdischarge is actually less likely to incur thermal runaway. However, side reactions like Cu dendrites growth during the overdischarge process would degrade the safety of LIBs and increase the possibility of thermal runaway occurrence.

Wu et al. [63] set up a three-electrode system to observe the change of open-circuit potential in LiCoO₂ batteries and pinpointed the decreased lithium content as the major reason for degraded thermal stability with cycling. Belov and Yang [64] utilized a "soft" overcharge technique to make LIBs reach different stages of overcharge and then executed differential scanning calorimetry (DSC) and scanning electron microscopy (SEM) on the recovered anode, cathode and separator, respectively. The heat rate (related to current), cell construction, and design were considered as the main factors of LIBs failure at overcharge. Ohsaki et al. [65] studied the overcharge using a 650mAh prismatic hermetically sealed Li-ion cell and found that the thermal runaway during overcharge was mainly caused by the violent reaction of overcharged anode and electrolyte solvent at high temperatures. Leising et al. [66] conducted an overcharge test to find the failure point on a 1.5 Ah prismatic lithium-ion cell and confirmed that the charge rate plays a significant role in triggering thermal runaway. Moreover, there existed significant temperature difference between the internal and surface of the cell, which implied a necessity

of thermocouples deployment for accuracy measurement of internal temperature [67]. Ouyang *et al.* [68] tested a large-format prismatic NCM LIB cell under overdischarge condition to investigate capacity degradation, internal resistance increase and internal short circuit phenomenon. Guo *et al.* studied the overdischarge mechanism for large-format LIBs by discharging a battery cell to -100% SOC, where ISC was observed by SEM and X-ray diffraction (XRD). The Cu collector dissolution at a special voltage platform was considered to be the main reason for internal short circuit. Ye *et al.* [69] employed an ARC to construct an adiabatic environment, in which an overcharge process was realized thorough a connected cycler to investigate heat accumulation and failure mechanisms of commercial LIBs.

3) MECHANICAL ABUSE TESTS

Typical mechanical abuse tests are nail penetration and crush tests based on respective testing facilities. However, most testing facilities can merely work as open systems and lack the ability of heat generation measurement. Fortunately, the latest ARCs have been equipped with a nail penetration unit, which can provide effective thermal boundary control and heat generation rate measurement during a nail penetration test.

Feng *et al.* [70] probed the propagation mechanism of nail penetration-induced thermal runaway by testing a large-format LIB pack composed of six battery cells and equipped with thermocouples and voltage measurement wires. The experiment result showed that: (a) Compared with an ARC-based oven-heat-induced thermal runaway, a penetration-induced test offered a lower onset temperature; (b) Temperature difference within an individual cell was as high as 791.8°C; (c) Heat transfer via electrode connectors was only 1/10 of that via the shell; (d) Fire could damage the accessories of the battery pack but imposed limited influence on thermal runaway propagation. Ren *et al.* [71] conducted a specially-designed pinch-torsion test and discovered that ISC would be incurred at lower axial load.

C. ADVANCED EXPERIMENTAL APPROACH APPLIED TO THERMAL RUNAWAY INVESTIGATION

In addition to the well-adopted thermal analysis approaches mentioned above, more advanced and interdisciplinary test methods and facilities are being gradually introduced to gain more insights on thermal runaway research. These methods include Nuclear Magnetic Resonance (NMR) [72], Industry Computed Tomography (CT) [73], Electrical Resistance Tomography (ERT) [74] and X-ray Diffraction [75], etc.

IV. SAFETY MANAGEMENT STRATEGY

In order to deal with the severe challenge of thermal runaway, researchers have made strenuous efforts in developing efficient methods for precaution, prevention and protection of battery systems. In this section, mainstream safety management strategies will be elaborated in terms of thermal runaway detection, prediction, and protection. A diagram

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that sketches out the existing safety management strategies is illustrated in Fig. 3 for demonstration.

A. BATTERY THERMAL MANAGEMENT SYSTEM

Battery thermal manage systems (BTMSs) are responsible for controlling LIBs work within an appropriate temperature range through functionalities of cooling or heating. In this section, the cooling functionality of BTMSs will be emphatically explored since most BTMSs are also required to have the ability to curb temperature rise in the early stage of thermal runaway events. Generally, BMTSs can be classified into four groups according to the heat transfer medium [76]:

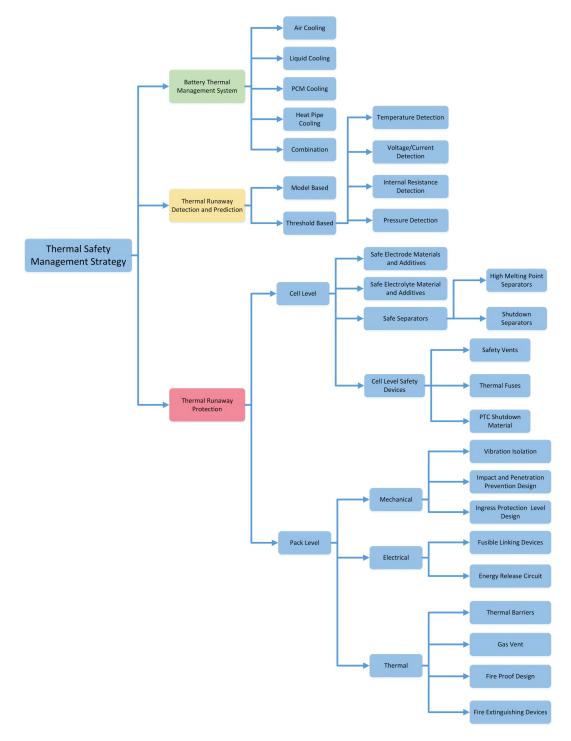
- (a) air cooling.
- (b) liquid cooling.
- (c) phase change materials (PCM).
- (d) heat pipe for cooling.
- (e) combinations of (a)-(d).

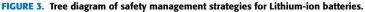
1) AIR COOLING

Air cooling has the advantages of simplicity and easy implementation along with low cost. Kelly et al. [77] and Zolot et al. [78], [79] conducted a series of experiments to investigate the thermal characteristics of battery systems in commercialized Honda Insight 2000 and Toyota Prius 2001. Battery systems in these two vehicle models utilized numerous NiMH battery cells and adopted air cooling designs. Chen and Evans [80] found that battery temperature may see a significant increase under high discharge rates if the thickness of the battery module exceeds a certain value. Besides, the air cooling was insufficient to cool down the core of batteries due to the poor thermal conductivity of polymer. Pesaran et al. [81] concluded that the air cool might be only suitable for the use in parallel HEVs while EVs and series HEVs required a better heat transfer medium. In order to mitigate temperature inconsistency within a battery pack, Mahamud and Park [82] introduced a reciprocating air flow thermal management method that was experimentally proved capable of reducing the temperature difference by 4°C (72%) in 120s.

2) LIQUID COOLING

Pesaran [83] and Bandhauer *et al.* [1] systematically compared air cooling and liquid cooling in terms of performance, functionality, volume, mass, cost, maintenance and safety. It was pointed out that liquid cooling could be achieved by various designs, i.e., discrete tubing around the module, setting a jacket around the module, submerging battery modules in a dielectric fluid for direct contacts, or placing battery modules on a liquid-cooled plate as heat sink. Compared to the air, direct-contact liquids have much higher heat transfer rate and can achieve 1.5 to 3 times higher cooling performance than the air cooling. Indirect-contact liquids, usually water or water/glycol solutions, is able to achieve more than 3 times better performance than the air cooling only at the expense of efficiency droop.





Liquid cooling systems have been successfully employed in some passenger vehicles. For example, Mercedes S400 Blue HYBRID adopted a liquid cooling design with the refrigerant used as the cooling medium [84]. For a HEV, the installed battery system is relatively small in size but have to bear heavy current and large heat generation. BEVs such as VOLT and TESLA [85] usually prefer active cooling/heating methods, achieving high thermal management efficiency and low temperature inconsistency. However, this comes off with more complicated system design, additive weight and space occupancy.

Aiming to solve the above mentioned problems, tremendous efforts have been made to optimize the cooling systems. Based on computational fluid dynamics (CFD) simulation and numerical optimization techniques, Jerrett and Kim [86] realized design optimization for a serpentine-channel cooling plate through optimized channel geometry, and improved the cooling performance in terms of pressure drop of channel, average temperature and temperature uniformity of the battery pack.

In order to deal with the problem of limited convective heat transfer inherent with conventional straight-channel cooling plates, Jin *et al.* [87] designed a set of oblique cuts across the straight fins to form an oblique-fin array, which was demonstrated effective even under heavy loading conditions for BEVs.

3) PCM COOLING

Rather than the use of air or liquid for cooling, some researchers have tried phase change materials (PCMs) by exploiting their large latent heat during the liquid/solid phase change process. Hallaj and Selman [88] introduced a PCM-based BTMS design and tested its performance using FEM. Simulation results manifested that the PCM-based BTMS was able to effectively contain the temperature rise of the battery module as well as to improve the temperature consistency. This would be more appropriate for heat-sensitive batteries such as Li-ion and Li-polymer batteries with a remarkable reversible heat effect. Khateeb et al. [89] tested the LIB module in an electric scooter, which selected the paraffin with a melting temperature of 41-44°C as PCM. However, the poor thermal conductivity of PCM may form a near-adiabatic environment after melting. Sabbah et al. [90] built numerical models to simulate a PCM-graphite composite matrix and an active forced-air cooling system mounted to a same LIB pack for HEV applications. The simulation results showed that the PCM-based cooling yielded better performance in both normal and stressed conditions without additional fan power.

4) HEAT PIPE COOLING

The heat pipe is a heat-transfer device that takes advantage of thermal conductivity and phase transition and provides a highly-efficient heat transfer between two solid interfaces [91]. Heat pipes have been extensively used in electronic devices but rarely seen in commercially-available battery packs.

Wu *et al.* [92] presented a combined metallic aluminum fin and heat pipe design to regulate temperature rise during discharge. Through experimental analysis, it was found that the connection between the heat pipe and the test batteries hold the key for heat transfer rate and correspondingly the heat pipe was inserted into an aluminum fin for effective temperature and uniformity control. Swanepoel [93] employed pulsating heat pipes (PHPs) to manage heat dissipation in a HEV battery pack. A series of parameters including heat pipe geometry, fluid type, pipe material, power inputs, filling ratios and inclination angles had considerable impact on the cooling performance. Swanepoel [94] proposed an ammonia axial grooved heat pipes (AGHPs) for a solar battery used

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in the International Space Station. Great attention was paid to its reliability and lifetime based on an accelerated test. Tran *et al.* [95] investigated a flat heat pipe for battery cooling system used in a HEV. Under a variety of cooling conditions and several inclined positions of the heat pipes, the addition of flat heat pipes would greatly reduce the thermal resistance for both natural and forced convection compared to a conventional heat sink.

5) COMBINATIONS

Since all the existing cooling methods have their inherent limitations, researchers tried a combinative use of more than two methods for better overall performance. Typical examples include optimized PCM with air/liquid cooling and optimized heat pipe system with air/liquid cooling.

PCM has great potential for absorption of heat but exhibits poor thermal conductivity. Thus, it is natural to combine PCM with other cooling methods to enhance heat conduction. Kizilel *et al.* [96] tested an 18650 cell module (4S5P) using a PCM-graphite matrix cooling setup under a thermal runaway condition. It was seen that the thermal runaway propagation from the faulty cell to surrounding cells could be effectually suppressed.

It is easily discernable that the heat pipe must work together with air or liquid cooling because the heat pipe needs to be cooled through timely and efficient heat transfer. Rao *et al.* [97] conducted experiments on a heat pipe BTMS for a large-format prismatic LiFePO₄ battery pack, which used a water sink $(25\pm0.05^{\circ}C)$ to cool down the condensation section. Burban *et al.* [98] performed a test on an open-looped PHP for an HEV electronic thermal management application, where various factors such as working fluid, air temperature, air velocity and inclinations were considered. Furthermore, Weng *et al.* [99] and Rao [100] combined the heat pipe with PCM to enhance heat flux rate and thermal uniformity, while reducing the power consumption of air cooling fans.

B. PREDICTION AND DETECTION OF THERMAL RUNAWAY PROCESS

Although routine BTMSs are expected to tackle heat generation and accumulation problems even under extreme high environmental temperatures, they may be unable to respond timely and effectively to prevent thermal runaway events. Many prediction and detection methods/techniques have been developed to work with BTMS in order to enhance the capability of early warning and protection before thermal runaway. In this section, we will review studies that assess the safety condition of LIBs and predict and detect thermal runaway events from the perspectives of methodologies and device designs.

In order to understand the likelihood of thermal runaway, Shah *et al.* [101] considered heat generation and heat removal and further presented a non-dimensional parameter, i.e., Thermal Runaway Number (TRN), to forecast thermal runaway events and thermal safety design space in a LIB pack. To be more comprehensive, Cabrera-Castillo *et al.* [102] raised a

parameter, i.e., State-of-Safety (SOS), to evaluate the safety of an energy storage system based on the concept that safety is inversely proportional to abuse. Kim et al. [103] presented a pseudo 2-D transient heat transfer model to predict timedependent temperature changes for thermal runaway prevention. A multi-layered LiCl-LiBr-LiF electrolyte battery cell was studied with a special attention on componentlevel behaviors. The built model was demonstrated effective in predicting the temperature characteristic of combustion flame propagation of the heat pellets in radial direction. Ouyang et al. [104] and Feng et al. [105] utilized both equivalent circuit and electrochemical-thermal-coupled models for online detection of ISC. Recursive least square algorithm was employed to estimate the indicator parameters. Jin [106] put forward a model-based method for battery thermal runaway prediction based on the energy conservation law and the Fourier law, which parameters were identified through ARC-based tests. Mu [107] presented a method to predict the maximum temperature on the battery surface using a back-propagation artificial neural network (BP-ANN) during charging process. The BP-ANN was trained by temperature profiles of a battery cell generated during charging in a thermal chamber. Ingalls et al. [108] presented a parameterthreshold-based method to forecast a thermal runaway event for a battery module used in an uninterruptible power supply unit. Voltage and temperature were monitored and maintained within their respective defined ranges by using a relay as control switch for charging. Wang [109] presented a method to predict thermal runaway propagation using voltage and temperature information collected from BMSs. The method is mainly composed of four steps: (a) monitor a battery cell to judge whether it is in a thermal runaway state by testing the temperature rise rate in different temperature ranges; (b) detect the battery voltage and calculate the voltage decline rate, and determine an irreversible thermal runaway event if it is lower than 0.2V/min; (c) detect the cells contiguous with the thermal runaway cell and perform (a) and (b) to assess whether a thermal runaway or irreversible failure happens; (d) if step (c) finds a thermal runaway cell, the BMS would issue a thermal runaway propagation alarm.

Benhan et al. [110] developed a device to detect thermal runaway of a Ni-Cd battery. The device included a current sensor and a converter to convert measured current to a pulse train, whose frequencies are proportional to current magnitude. The frequency changes of the pulse train was able to determine the slope of the charge current and used for thermal runaway detection. McShane et al. [111] invented an apparatus to detect thermal runaway events during battery charging based on the increase of internal resistance (impedance) and/or conductance (admittance). Zhang [112] designed a thermal runaway detection and mitigation system using flame, temperature, gas and smoke sensors and a fire extinguisher that would take effect once a thermal runaway event was detected. Tesla Motors patented a series of methods for battery thermal runaway detection by monitoring operating states such as battery pack pressure [113] and battery pack isolation resistance [114]. Specially designed sensors including optical fibers [115], thermal interruptible electrical conductor [116] and electrical conductors with thermally-fusible insulators were added to guarantee the functionality of their detection system [117].

Obviously, the most remarkable symbol for a thermal runaway process is the dramatic and significant temperature rise in additional to the explicit smoke or flame. Temperaturedetection-based methods would be reliable and effective for the initiation of passive safety devices. However, temperature is usually sampled on the cell surface, and there would be a significant discrepancy between the core and surface when thermal runaway events occur. This inevitably impairs the prediction ability for early-stage thermal runaway detection. Abnormities of operating parameters such as voltage and resistance can be used to prelude thermal runaway, which are more reliable for the early-stage prediction. However, the underlying mechanism still needs to be addressed before practical applications.

C. PROTECTION METHODS AGAINST THERMAL RUNAWAY Protection methods are employed to prevent the occurrence and mitigate the negative consequence of thermal runaway within battery systems at both cell- and system-levels.

1) CELL-LEVEL

Safety design on cell-level has been conducted by introducing safety vents, shutdown additives and current cut-off device and so forth. Balakrishnan et al. [118] and Wang et al. [119] investigated the safety mechanism of a battery cell from the aspects of separator, electrolyte, and active materials. Regarding safety chemicals/additives for batteries with different cathode materials, modification methods for coating and doping have been proposed. Research concerning the anode safety mainly focuses on the SEI layer formation and its stability. Meanwhile, the electrolyte is also being intensively studied to enhance battery safety, with the addition of non-flammable additives, redox shuttle additives, shutdown additives or employment of stable electrolyte salts and ionic liquid. Solid state electrolyte is a promising solution to the electrolyte safety problem, which possesses intrinsic thermal characteristics and can form a natural barrier for ISC prevention. Separator is the key element in LIBs to prevent direct contract of anode and cathode but provides viable corridors for lithium-ions transport. Thermal runaway events evolving from separator melting have been well documented. To address this issue, separators with high melting point and low shrinking rate are under development. To face up to overcharge or high temperature abuse, a shutdown separator could shut down the micro-pores in the film and cut off ionic transport between the electrodes, thus preventing short circuit current. Apart from these component design optimization, cell-level safety devices are also considered important for mitigating the propagation of thermal within a module or pack. For example, safety vents and thermal fuses are usually used to prevent overpressure and

layer between the cell connections to prevent a short circuit

incident. CATL Company [137] designed a sandwich struc-

ture in cells, which comprises of two heat conducting layers

and a heat resisting layer in between. This structure was able

to conduct heat to the outside of battery pack and mitigate

overcurrent during extreme conditions. Self-resetting devices such as ceramic PTC/conductive-polymer materials have been reportedly incorporated into cell design, which can cut off a battery cell in overcurrent and overheat situations and revert when the temperature reduces.

2) SYSTEM-LEVEL

Battery safety problems could be addressed from three aspects on system-level, i.e. isolating the battery system from potential external abuses, preventing thermal runaway propagation inside the battery system and discharging energy in time when thermal runaway happens.

Tesla Motors devised an array of thermal runaway propagation prevention strategies via adding thermal barriers at different levels such as intumescent materials on the cell surface [120], thermal barrier plates in battery pack [121], and multi-layer thermal barriers between battery pack and passenger cabin [122]. They also raised a rigid battery holder made from heat resisting material to hold cells apart so as to lower the thermal conductivity among cells [123]. Moreover, a strategy that combines thermal barriers and battery pack cooling approaches was proposed to limit the heat transfer between cells as well to transport the generated heat to the outside of battery pack [124]. Another method for thermal runaway propagation alleviation was invented using temperature measurement devices associated with switches [125]. Moreover, by using fusible link wires [126] and tunable frangible components [127], battery cells and modules could be disconnected under over-current, short circuit, mechanical crush and over-heat conditions. Exhaust vents [128], [129] on battery pack were also designed to realize quick gas release with directional ventilation and adaptive outlets determined by the amount of gas release.

Harm and Timm [130] attached a thermal sensor to the test battery cells, with which the charging current could be adjusted to prevent thermal runaway under certain conditions. The Boeing Company [131] invented an active thermal management and thermal runaway prevention system for LIB packs. This system included thermal sensors on every battery cell and a control system to determine a cooling operation in case of thermal runaway by pumping the cooling liquid to circulate among battery cells. Muniz [132] used a device that consists of thermal insulators and thermal conductors, where the thermal insulators were placed to prevent thermal runaway propagation and thermal conductors were used to draw heat from the faulty cells and distributes it to the surrounding cells properly. Hu et al. [133] presented a protection structure for preventing thermal runaway propagation with a battery module casing and a composite heat conduction plate design. Zhai [134] took advantage of high heat conduction efficiency inherent with heat pipe cooling for thermal runaway prevention. Tartaglia [135] invented a battery case made of sturdy materials and held the power interface terminal and electronics within the case wall. The protective layer can protect battery cells from external heat, out-gassing and explosion of other battery cells. Ma [136] invented a PTC shutdown

a thermal runaway propagation process. They also invented a fire extinguishing kit for battery pack [138], which comprised of a water-based fire extinguisher for putting out fire and reducing temperature and a water container for preventing electric shock accidents. BYD Company [139] invented a cell protection device connected to positive and negative cell connectors. The device included two separate alloy bars sealed in insulating containers, which were connected with the cell connector through wires. Under high temperature conditions, the alloy bars would melt and form an electric path, through which the battery cell could be quickly discharged. A battery protection device composed of a cover plate and a shape memory alloy (SMA) component was also presented [140], in which the cover plate was electrically connected to one cell connector while the SMA component was connected to the other. The SMA component possesses two different shapes under low and high temperatures. When the battery was under a low temperature, the SMA component would be separated from the cover plate; when the battery has high temperature or endures thermal runaway, the SMA component would change its shape and contact with the cover plate, thus forming a short circuit to release the stored energy. Mayer and Whitehead [141] developed a two-stage control mechanism to deal with an internal over-pressure of a battery cell. The first stage was to disconnect the cell from others by a contactor. If the pressure was still rising, the second stage would be activated and the cell would rupture to release internal pressure. Bandhauer and Farmer [142] invented a fire suppression system based on the liquid-vapor phase change heat removal principle. When a thermal runaway event occurs and is sensed by either active or passive sensors, inert high-pressure refrigerant will be ejected through a specially designed path within the battery pack. Similarly, Hou [143] utilized a closed-loop air cooling system to control the temperature of battery pack, which would eject inertia gas through a solenoid-valve-controlled inlet into the battery pack when a thermal runaway event was detected. Wilke et al. [144] used phase change composite (PCC) material to prevent thermal runaway propagation. PCC material was placed between cells in a LIB module and experimental studies showed that the maximum temperature experienced by the surrounding cells was decreased by 60°C, which indicated an effective hindrance of thermal runaway propagation within the pack. Commercial battery modules or packs always employ current interrupt devices (CIDs) to shut down the circuits under overcharge or overpressure occasions. However, Augeard et al. [145] raised the awareness of electrical arc issue during the current interrupting processes and found that a DC voltage of 19V was sufficient for 18650 battery cells to maintain an electrical arc, which was well lower than the upper limit of 48V permitted by the National

Electrical Code. Accordingly, a method for electrical arc detection based on its voltage characteristic was synthesized to reduce the arc occurrence risk. Arora *et al.* [146] and Wang *et al.* [147] investigated mechanical design and strategic placement to confront safety issues. Structural design for battery packs should be considered to realize protective function of (a) vibration isolation to avoid mechanical fatigue failure of electrical and mechanical components; (b) sufficient strength and stiffness to survive the rear, side and front impacts and even nail penetration; (c) sufficient ingress protection (IP) level to proof water, dust or other harsh environments a vehicle may face; and (d) fireproof and flame-retardant structural and material design.

It is well-known that a LIB cell is a close system. Once a cell is manufactured, its intrinsic safety would be determined on cell-level. Therefore, improvements of material and manufacturing technology would be the most effective way to enhance LIB safety. On the other hand, there are numerous safety designs and devices, aiming to further enhance the safety protection functionality on system-level. The basic idea mainly comes from ways for handling the released energy upon the advent of thermal runaway. The major routes for energy release include cutting off heat energy spread paths and discharging electrochemical energy via other paths. Up to now, it is still an open problem to integrate practical hardware/software with battery system design for better overall safety.

V. CONCLUSION

In this paper, technological progress relevant to thermal safety issues of LIBs is systematically and thoroughly reviewed. The detailed discussions mainly cover (a) thermal behavior, modeling and tests, (b) thermal runaway modeling and tests, and (c) safety management strategies for LIB packs.

Heat generation and temperature rise have been explicated from the perspective of the underlying electrochemical reactions. Thermal modeling often utilizes a coupled partial differential equations to describe the nonlinear dynamics of LIBs under different charge/discharge conditions. 1-D, 2-D and 3-D electrochemical models have been reported in literature to depict the battery thermal behaviors with varied precision. Among them, the pseudo-two-dimensional (P2D) model strikes a good balance between modeling performance and computational cost and is thus widely adopted. Moreover, electrochemical-thermal-coupled modeling methods have been presented to improve the modeling precision considering the influence of temperature-dependent factors. In addition, experimental approaches to probe thermal characteristics of LIBs are also briefly discussed.

Modeling methodologies for thermal runaway of LIBs are invariably complex due to the high nonlinearity of the thermal runaway process and its versatile triggering factors. Multiphysics fields including electrochemical, electrical, thermal and mechanical are needed to be considered. Modelling for thermal runaway propagation always contains multiple cells and has three dimensions. In such case, empirical formula and experimental data are always employed to lower the model complexity with acceptable precision. Furthermore, experimental methods are also elaborated from the aspects of mechanical, electrical, and thermal abuses. Testing approaches are crucial in the study of thermal runaway and need to be able to simulate real abuse occasions in EV applications. Advanced measurement methods are also mentioned by using NMR, CT, and ERT, etc.

For thermal runaway management and protection, both cell- and system-level strategies have been briefed. Cell-level studies mainly focus on materials refinement and inclusion of enabling additives for electrodes, separator and electrolyte. Cell-level safety devices such as safety vents, thermal fuses, and shutdown separators have been widely used to enhance cell safety. For practical consideration, more emphasis is laid on system-level management methods. BMTS is a routine system to prevent thermal runaway. Multiple choices such as air cooling, liquid cooling, PCM cooling, heat pipe cooling and their combinations can be used. For the most dangerous occasions, various algorithms and devices for thermal runaway prediction and detection can leave as much time as possible for passengers' exit and fire extinguisher activation. Once thermal runaway incidents happen, the protective methods may offer the last resort to postpone its propagation by shutting down the respective circuitry and discharging the stored energy.

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