A review of the estimation and heating methods for lithium-ion batteries pack at the cold environment

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Abstract
The application of lithium-ion batteries especially for electric vehicles has been limited by the factors of safety, lifetime, charging time, and cost. One of the principal limitations is that the performance of Li-ion batteries drops intensely in a cold environment. Cold environment dramatically reduces the available capacity of the batteries and increases its internal impedance at the same time. Therefore, the estimation of state-of-health is of great importance in battery performance evaluation and lifetime prediction. Furthermore, the heating methods need to be developed to ensure that batteries work in abnormal temperature conditions. This paper conducts a comprehensive review specifically on the poor performance of lithium-ion cells under severe conditions. The content contains three sections. First, a comprehensive study on the aging mechanisms of lithium-ion batteries at cold temperatures is undertaken. Second, the estimation methods of the health state of the batteries are conducted, which is vital to understand the fundamentals and quantify the performance and aging effects for lithium-ion batteries. Third, the heating methods are classified and studied in detail to reduce the degradation mechanism and promote the performance of lithium-ion batteries under sub-zero conditions.

KEYWORDS
battery pack, cold temperatures, estimation, heating methods, lithium-ion batteries

1 | INTRODUCTION

Within the last two decades, rechargeable cells especially Li-ion cells have received a relatively wide application for large-scale electric storage, mostly in EVs (electric vehicles) and digital products such as mobile phones for its terrific superiority of high energy density, long lifetime, high voltage, and low self-discharge ratio. For instance, during the year 2000, the worldwide production of lithium-ion batteries reached about 500 million cells. Moreover, from 2000 to 2010, it increased with an annual growth rate of 800%.

On the other hand, there is a massive contradiction between the increasingly scarce oil resources and the increasing energy demand. Taking the automobile industry as an example, some countries have banned fuel vehicles on the agenda due to the lack of oil reserves, and will gradually ban the production and sale of fuel vehicles after 2030. It can be predicted that electric vehicles will be a major trend in the future development of automobiles. Battery packs, which are composed of hundreds of batteries and can provide enough energy and power for the regular work of electric vehicles, are particularly important as the energy supply of electric vehicles. Among many kinds of batteries, lithium-ion batteries have been widely used in the electric vehicle industry due to their high energy density, high power density, long cycle life, and low self-discharge rate.
However, there are still some problems in the safety, cost, and recycling of lithium-ion batteries. Also, some problems arise in low-temperature operation, such as lower charge and discharge capacity, lower rated voltage, shorter cycle life and so on.\textsuperscript{9,10} Such a situation can easily occur in winter-driven cars, especially in cold areas such as Russia, Canada, northern parts of China and USA.

To alleviate the demand for petroleum energy, lithium batteries are an indispensable alternative, and it is particularly important to reduce the negative impact of lithium batteries in a low-temperature environment.\textsuperscript{11} Therefore, it is essential to understand the changes in battery performance at low temperature and the principle of the changes. Fundamentally, lowering temperature will slow down the rate of chemical reaction in lithium batteries, thus affecting the overall performance of batteries. Lithium-ion batteries also experience lithium plating at low temperatures, which can reduce the energy and power capacity of lithium-ion batteries and lead to serious degradation of batteries.\textsuperscript{12} In the low-temperature working environment, the charging and discharging voltage and capacity of lithium batteries change significantly, and these indicators are closely related to the health of the batteries. To improve the working performance, cycle life and keep high health status of batteries, batteries are usually heated internally or externally.

There are several studies considering these problems: First, the degradation mechanism, previous studies have proved that the effect of aging degrades the operating performance of lithium-ion batteries. Researchers have also drawn some conclusions that the exceedingly reduced the power and capabilities of lithium-ion batteries are related to the severely cold climate, which accelerates the aging mechanism. Second, there are several heating methods proposed for lithium-ion batteries at cold temperatures, such as internal heating, air heating, and liquid heating. Air heating is typically used in electric vehicles at present.\textsuperscript{13}

Although some relative topics, such as the effect of cold temperature in Li-ion batteries, modeling a Lithium-ion battery, and heating strategies, have been reviewed, some technologies emerging in recent years, such as the role of PCMs in a low-temperature environment, high energy conversion spontaneous heating system, may have some potential role in promoting the development of low-temperature lithium batteries. With the motivation to promote battery performance under cold environment, this article is organized as follows: First, the detailed study concerning the mechanisms of lithium-ion cells in the subzero environment is conducted, which is crucial to understand the fundamentals. Second, some estimation methods for lithium-ion batteries in a low-temperature environment are presented, it is necessary to quantify the performance of lithium-ion batteries under these severe environments. Third, to reduce the aging or degradation of the batteries, the heating methods for Li-ion batteries are classified and compared in detail. Finally, conclusions are given.

2 | THE EFFECT OF LOW TEMPERATURES ON LITHIUM-ION CELLS

As is known to all, the ambient temperature considerably affects the performance of lithium-ion batteries. In general, no matter the ambient temperature is too high or too low, there will be performance losses for the batteries. Moreover, when the lithium-ion batteries are under subzero-temperatures, these performance losses such as decreased power capacity and degradation due to lithium plating will be much more severe. In this part, the effects of cold temperatures
on lithium-ion batteries will be thoroughly presented from principles.

2.1 | Low-temperature performance

For the correlation between the ambient temperature and the performance of lithium-ion batteries, it is vital to understand why its performance decreases dramatically from low-temperature effects. For instance, it has been measured when the current is the same, the available capacity of a lithium-ion cell at −20°C is only 60% of the average value at room temperature. Some experiments were conducted to understand the behavior of lithium-ion batteries in, the charge voltage profiles of lithium-ion batteries at a temperature range (from −15°C to 25°C) and the constant charging current of 1 A are shown in Figure 1A,B demonstrates the discharging characteristics at the constant current of 0.25 A at the temperature range. It is evident that cell delivers significantly less capacity at subzero temperatures compared with that at ambient temperatures of 25°C.

There are some reasons for poor performance of lithium-ion cells, such as the motion of lithium ions in the electrolyte solution (electrolyte conductivity), design of the structure, thickness of electrode, separator porosity, and separator wetting characteristics. First, it has been concluded that the electrolyte of the cells affects its performance. As is shown in Figure 2, low-temperature differential scanning calorimetry (DSC) curves of various electrolytes are plotted in Foss et al. With the decrease in temperature, the first response is an endothermic peak for all electrolytes, which represents the liquidus temperature. All liquidus temperatures are summarized in Table 1, where $T_{\text{liq}}$ is determined from the onset of the endothermic peak. As expected, the addition of PC to the electrolytes is very efficient regarding reducing the liquidus temperatures. The liquidus temperatures for the 1:1 EC:DEC, 1:2:2 EC:DMC:EMC and 4:3:3 EC:DMC:EMC solvents have been reported at approximately +23.5, −5 and +7°C, respectively. The addition of 0.9 mol/L LiPF$_6$ reduces the liquidus temperature for these solvents; as the addition of salt, the binary system becomes a ternary system, a ternary becomes a quarternary, etc. (analogous to freezing point depression). Reduction of the liquidus temperature and crystallization temperatures has previously also been reported for the addition of LiPF$_6$ in EC: EMC solutions. The reductions due to addition of salt amount to 16.5, 28 and 13°C for 1:1 EC:DEC, 1:2:2 EC: DMC: EMC and 4:3:3 EC: DMC: EMC, respectively. As pointed out in the works by Ouyang, ternary and quaternary carbonate solvents typically exhibit a wider liquid range compared to binary mixtures, as is also observed here. For electrolytes containing PC, the liquidus temperature drops to below −40°C. Notably, by replacing 20% EC with PC in the 4:3:3 EC: DMC: EMC electrolyte, the liquidus temperature was decreased by almost 40°C (as seen by comparing to 2:2:3:3 EC:PC:DMC: EMC in Figure 2). The liquidus temperature of 1:1:3 EC:PC:EMC is close to the one reported for 1:1:3 EC:PC:EMC with 1 mol/L LiPF$_6$ and 0.05 mol/L CsPF$_6$, −64°C.

However, previous studies mainly concern the design of lithium-ion electrolytes with lower freezing points and higher ionic conductivities for cold temperatures. In Shiao et al., the authors investigated several kinds of electrolytes in actual experiments under cold temperatures, such as MCMB anodes and LiNiCoO$_2$ cathodes. Discharge characterization tests were conducted under different discharge rates from C/100 to 3C and over an extensive

![Figure 2](image-url) Differential scanning calorimetry response of pure electrolytes. Endothermic peaks are upwards.

| Electrolytes               | $T_{\text{liq}}$ from DCS measurements [°C] |
|----------------------------|---------------------------------------------|
| 1:1 EC:DEC+1% VC, 0.9 mol/L LiPF$_6$ | 7                                           |
| 1:2:2 EC:DMC:EMC, 0.9 mol/L LiPF$_6$ | −32                                         |
| 4:3:3 EC:DMC:EMC, 0.9 mol/L LiPF$_6$ | −6                                          |
| 1:1:3 EC:PC:EMC, 0.9 mol/L LiPF$_6$ | −61                                         |
| 2:2:3:3 EC:PC:DMC:EMC, 0.9 mol/L LiPF$_6$ | −45                                        |
| 1:1:3 EC:EA:EMC, 0.9 mol/L LiPF$_6$ | −33                                         |
temperature range from −70°C to +30°C. The results of this study showed that if carbonate-based electrolytes were adopted, there would be adequate conductivity for the improvement of performance.

In Zhang et al.,23 the poor performance of lithium-ion cells is associated with high charge-transfer resistance. The author concluded that when a battery with a LiBF₄-based electrolyte, during the charge-transfer process, the resistance dramatically increased as the ambient temperature drops below −20°C. The results of the experiments demonstrated that comparing with the electrolyte based on LiPF₆, the electrolyte based on LiBF₄ will have a lower charge-transfer resistance so that the performance will be improved.

Moreover, some research was carried out for the impedance spectra of symmetric structural batteries,24 they concluded that the poor performance of the lithium-ion batteries under cold temperatures is associated with the increase in charge-transfer resistance for the graphite and cathode. Also, SOC (state of charge) affects the charge-transfer resistance. In cold environments, charging a discharged lithium-ion cell will cause more problem that for the electrodes than discharging a charged one. Huang et al.23,25 presented the idea that what limits the performance at cold environments is the graphite anode rather than the cathode. It has been confirmed that the reason for the poor performance is the Li⁺ diffusion in the carbon anode instead of the electrolyte.

Figure 3 shows the voltage–capacity curves of the discharging process for two cells using LiPF₆ and LiBF₄, respectively, at various temperatures. At 20°C where the value of “relative capacity” is defined as 1.0, discharging capacities of these two cells are 1.13 mAh/cm² (LiPF₆) and 1.05 mAh/cm² (LiBF₄), respectively. Relatively lower capacity of the LiBF₄ cell results from its higher irreversible capacity in the first cycle, which consumes parts of the limited lithium ions in the cathode. As shown in Figure 3, with the decrease in temperature, these cells not only suffer a lower operating voltage (power) but also the loss of capacity (energy). The decreased operating voltage at the low temperatures can be attributed to the polarization mechanisms originated from the increase in the electric cell resistance (including the electrolyte, electrodes, and SEI) and by a slowdown of the cell electrochemical reaction. When the temperature is above −10°C, the relative discharging capacities of these cells are close to unity. In these cases, the decrease in ambient temperature is mainly reflected by an increase in the cell polarization with a negligible change in the discharging capacity.23

There were some Electrochemical models proposed to analyze the physical and chemical degradation phenomena of lithium-ion batteries in the real application. Electrochemical model is regarded as a fundamental or particle-based distribution model, which is proposed to offer a deeper perception of the occurrence of physical and chemical phenomena during battery application. Such models are applied at the nanoscale using partial differential equations.26,27 The development of these models provide a profound understanding of non-measurable battery parameters, such as the work of using the anode potential as an indicator of lithium plating by Newman’s group,28,29 they proposed some approaches for the estimation of battery performance on the basis of Butler–Volmer equation and porous electrode theory.30 Feng et al.31 proposed a model about the temperature prediction of lithium-ion cells. This model was studied from electrical theory, which modified correction factors to improve precision. Some factors such as current and temperature dependencies were considered along with the increase in internal resistance at the cold environment to enhance the precision.

In Foss et al.,15 the performance of graphite electrodes in various electrolytes containing ethylene carbonate (EC) and mixtures of EC and propylene carbonate (PC) was studied at the temperature range between 0 and 40°C, the effect for the addition of ethyl acetate (EA) was also introduced in this research. Differential scanning calorimetry (DSC) was adopted to investigate phase transitions at cold environment (down to −80°C) and decomposition at elevated temperatures. Capacity loss was compared for graphite electrodes cycled at the temperature range from 0 to 40°C for these electrolytes. It was validated that suitable electrolytes can work at a wide temperature range. The addition of EA improved the low temperature properties of the electrolyte and the graphite.

**FIGURE 3** Voltage–capacity plots of lithium-ion cells at various temperatures. A, LiPF₆ and B, LiBF₄ (Relative capacity is treated as a ratio of the capacity at a specified temperature to the one obtained at 20°C).
electrode, but the electrodes failed while cycling at +40°C. For the addition of PC to a multi-component system, which accounted for 40% of the total amount of cyclic carbonates (ie, 20% EC and 20% PC), expanded the liquid temperature range of the electrolyte. However, the addition of PC also led to considerably high initial irreversible capacity loss of the graphite electrode, and reduced the capacity significantly at 0°C, most likely related to the higher resistance of solid electrolyte interphase. Therefore, mixture of EC and linear carbonates such as dimethyl carbonate (DMC) and ethyl methyl carbonate (EMC) was found to have best performance in this temperature range.

As denoted in Table 1, graphite electrodes were cycled together with the electrolytes. Compared with the electrolytes without PC, electrochemical performance of electrolytes containing PC exhibited a relatively severe initial capacity loss as seen in Table 2. This phenomenon is expected, and normally attributed to exfoliation during the initial charge due to lithium species solvated by PC, co-intercalating into the graphite structure before a protective SEI is formed by solvent reduction. A significant increment in the charge consumed above the first intercalation potential is observed for the PC containing electrolyte, the voltage profiles during the first cycle is shown in Figure 4. The irregular profile observed for the PC containing electrolytes is typical for electrodes experiencing some exfoliation during the first cycle, as was previously verified for graphite anodes cycled in PC-containing electrolyte.26 However, after a protective SEI is formed, the capacity stabilizes.

The de-lithiation capacities at different temperatures for all the investigated electrolytes are plotted in Figure 5. The red line and the right y-axis show the temperature at which the cells were cycled. The best 0°C capacities are found with 4:3:3 EC:DMC:EMC (172 mAh/g) and 1:2:2 EC:DMC:EMC (169 mAh/g) as electrolyte, respectively. The expected disadvantages with EC at lower temperatures (solidification) do not exhibit at 0°C, and the temperature should be even lower to observe these effects. Both electrolytes with 40 and 20% EC therefore seem acceptable for lithium-ion cells’ built for practical applications operating in the temperature range from 0 to 40°C.

To sum up, lithium-ion batteries undergo a drop in energy and power for higher resistance and lower diffusion of Li+ ions at the anode/electrolyte interface. Also, as the result of significant aging mechanisms, capacity will decrease dramatically when the lithium-ion cell is operated in a cold environment.

### 2.2 Lithium plating and aging effects

Aging occurs at the interfaces between the electrolyte and the electrodes under the chemical composition of the electrolyte.32 Aging mechanisms can be classified as either mechanical or chemical because of the electrode composition. And there are two principal effects for lithium-ion batteries: impedance rise and capacity fade.33 According to chemical-based mechanisms, performance loss is related to electrode materials. The consequences of aging effects are listed below:

1. There will be a passivation layer on the surface of the anode, which is called solid electrolyte interphase (SEI), and it results in an impedance rise for the anode.34 Generally, this phenomenon mainly occurs from the beginning of aging, but its growth lasts throughout the whole process.

2. For both electrodes, the cyclable lithium will decrease at the same time, at the carbon anode, SEI will grow for electrolyte oxidation or changes in the oxide surface structure.35

3. Contact losses will gradually increase in the composite anode with the formation and growth of SEI, which results in the increase in the impedance for batteries.34

4. The loss of active electrode materials will occur, such as material dissolution, structural degradation, particle isolation, and electrode decomposition.
5. Capacity fades for the loss of active materials and cyclability, and power fades for the increase in resistance.

The phenomenon of lithium plating plays a dominant role in aging effects for high polarization of the anode. The author of\textsuperscript{36} revealed the phenomenon of negative polarization of lithium ion by measuring pouch cells with reference electrodes. At low temperatures, metallic lithium was deposited on the surface of the anode. Moreover, there was grey metallic lithium plated on the surface of anodes, after the experiments about pouch cells. The phenomenon of lithium plating results in capacity loss through hindering the intercalation of lithium between the anode and electrolyte. Lithium plating also leads to electrolyte decomposition, the loss of lithium-ion will accelerate capacity loss and decrease the lifespan in turn.\textsuperscript{37} In Mühlbauer et al,\textsuperscript{38} for a “fatigue” cell, the distribution of the lithium concentration inside the graphite anode under fully charging state was detected by spatially resolved neutron diffraction. Each cell was tested under 1000 CCCV cycles (charge/discharge under 1 and 25°C). The result obtained from these tests was compared with the previously published lithium distribution in a “fresh” cell. The comparison revealed the fundamentally lower level of lithiation in a “fatigue” cell, the comparison is well associated with capacity loss, and the results can be explained by the decrease in moving lithium for the electrode slippage during the extensive cycling. Moreover, an evident difference from average (plateau-like) lithium distribution can also be detected after these tests.

When a single cell is charged at the cold environment, there will be the dendritic growth of metallic lithium on the surface of anodes.\textsuperscript{39} Metallic lithium with the shape of dendrites will penetrate the separator to reach the positive electrode, which leads to some dangerous results such as internal soft shorting. Lithium plating results in distinctive effect when cell approximately reaches its lifespan. Thus, for metallic lithium, lithium plating and dendrite growth should be regarded as the parasitic side reactions during the charging process. Charging lithium-ion cells in a cold environment is one of the most significant tasks for broader applications of lithium-ion batteries. In Remmlinger et al,\textsuperscript{40} an electrochemical model was introduced to predict aging effects and
understand lithium deposition reaction. Parameters of the model were determined by the data from the frequency domain through the method of EIS (electrochemical impedance spectroscopy). During the process, batteries were charged under a wide temperature range (from 25 to 40°C) and current range (from 0.1 C to 6 C). The results were compared with simulated anode potential, which showed that capacity faded after a significant number of cycles. As expected, high current and cold environment both resulted in the acceleration of aging.

Currently, the dominant method adopted to solve the problem of lithium plating is using the anodes that operate at high voltage values, such as using lithium titanate, which will be harmful to energy density. The battery chemistry was changed by different coating amounts of carbon coating (1,3-propane sultone) to the surface of natural graphite to lessen the phenomenon of lithium plating under low temperatures. The modification was finished by uniformly applying carbon to the surface of graphite through the method of CVD (chemical vapor deposition). During the process, the entire graphite surface was covered by carbon coating; unwanted active points will be eliminated, which will be helpful for the optimized SEI layer to form. The results proved that the carbon coating on the surface is helpful for suppressing the impact of lithium plating and ensuring the safety of lithium batteries.

2.3 | Methods to detect lithium plating

Since the phenomenon of lithium plating plays a dominant role in aging effects for high polarization of the anode, it is necessary to study the methods to detect lithium plating. And there are manners of detection methods.

2.3.1 | Imaging methods by the optical microscopy, SEM and AFM

Osaka evaluated the electrodeposited Li on nickel substrate using optical microscopy in two types of liquid solutions to investigate the correlation between the surface morphology and the resistance distribution, and studied the lithium plating process in liquid, gel, and solid electrolyte. Brissot conducted comprehensive studies about lithium plating in poly- ethyleneoxide/Li-salt electrolytes using a charge-coupled device (CCD) camera fit on an optical microscope in refs. From the 1990s, scanning electron microscope (SEM) and atomic force microscope (AFM) have been extensively used for situ and ex situ studies of the process of lithium plating in many electrolyte systems and achieved much better resolutions than optical imaging methods. To avoid the possible contamination of the sample or the secondary reaction with atmospheric components, special attachments have been developed in to provide a vacuum environment in the sample transfer process.

In Petzl and Danzer, an investigation was conducted using pulse-relaxation experiments, scanning electron microscopy (SEM) and optical in situ microscopy to improve the understanding of Lithium plating (Figure 6). In this study, current pulses up to 10 C were applied on graphite half-cells to induce plating on the anode's surface. The characteristics of the results such as cell voltage and changes in surface morphology were analyzed during pulses and subsequent relaxation. Some characteristic attributes can be detected as symbol of lithium plating: (i) a prominent kink of the voltage transient during charging, (ii) a distinctive plateau in the subsequent relaxation of the cell voltage, (iii) a gray deposit covering the anode surface which under high magnification shows, (iv) a net-like structure covering the carbon particles. The observed characteristics for lithium plating were

![](image-url)
2.3.2 Nuclear Magnetic Resonance (NMR)

As mentioned before, the optical microscopy, SEM and AFM can visually reveal the morphology of the lithium plating. However, imaging methods are not suitable for quantitative studies in dynamic conditions. In Bhattacharya et al., in situ NMR spectroscopy was used in the observations of lithium plating, which provided time-resolved quantitative information about the deposition on Li metal electrodes. The research reported in Schweikert et al. demonstrated that the NMR spectroscopy could reveal the “skin-depth” of the electrodes with a penetration depth of 14.7 mm to monitor the chemical changes in this range of depth. The presence of the “two Li resonance peaks” in the NMR spectra was attributed to different surface structures on the Li electrode during charging. The first resonance peak, approximately at the same position of the original resonance peak in the spectra of a fresh cell, was assigned to the remaining initial Li surface. The growth of new Li deposition explained the second resonance peak as dendrites on the surface. Chandrashekar et al. revealed the location of microstructural Li as a result of charging using the magnetic resonance imaging (MRI) technique with a Li metal symmetric cell. This method provides quantitative information of the compositions, as well as high spatial resolution, which could be readily applied to investigate the formation of dendrites under different electrochemical conditions (charging/discharging rates, different electrolyte salts, additives or solvents, etc.), the approach is also helpful for the design of batteries.

2.3.3 Electrochemical Impedance Spectroscopy (EIS)

Lithium plating has been observed by EIS in Schweikert et al. that in a Li/Li$_4$Ti$_5$O$_12$ battery in LiPF$_6$ and EC/DMC electrolyte, the higher-frequency resistance related to the SEI layer decreased with time rapidly, which was caused by the SEI damage due to the dendrite formation. In Yang et al., the cell impedance evolution was measured from two symmetric lithium cells with a polymer electrolyte, whereas in each symmetric cell the two electrodes were made of electrodeposited Li films and commercial Li foils, respectively. The EIS results showed that the interfacial impedance of both cells increased during cycle aging process. Furthermore, the interfacial impedance of the cell with electrodeposited Li films as the electrodes was relatively small and the cell reached equilibrium more quickly.

Besides, the characterization methods to detect the material composition, such as X-ray diffraction (XRD) or X-ray photoelectron spectroscopy (XPS), were used in the research of lithium plating. Although the composition of the surface has certain impacts on the morphology, the morphology characterization technologies such as scanning electron microscope (SEM) and Atomic Force Microscope (AFM) are more effective to observe the detrimental formation of dendrites directly. Voltage relaxation and impedance spectroscopy were introduced as in-operando approaches for detecting lithium plating in commercial Li-ion batteries with graphitic anodes. Voltage relaxation is monitored after defined charge steps of variable amplitudes, charge throughputs, termination criteria, and at different ambient temperatures yielding dependencies over a wide experimental parameter range. Adapted differential voltage analysis was also introduced to determine the characteristic mixed potential evolving in the case of plating. Impedance spectroscopy was used in parallel to the relaxation phase to trace a possible alteration of the cell’s impedance due to the concurrent depletion of reversibly deposited lithium. The introduced voltage differentials were shown to determine the mixed potential with restrictions only for little charge throughputs. The comparison of voltage relaxation and already established stripping discharge showed similarities of the underlying physicochemical processes and allowed an estimate of the amount of deposited lithium in case of relaxation. During the evolution process of the cell’s impedance, a reversible shrinkage of the high-frequency intersection resistance and the arc representing the anodic charge transfer process were identified as indicators towards lithium plating. The presented methods only rely on non-destructive measurement quantities and thus are completely suitable for the application in battery management systems.

2.3.4 Other methods

Voltage plateau during relaxation or discharge process after charging can be used as a distinct indicator associated with the stripping of deposited lithium metal and hence a feasible tool for online detection of lithium plating in lithium-ion cells. In Yang et al., a physics-based model was proposed with the incorporation of lithium plating and stripping to gain a fundamental understanding of the voltage plateau behavior. To be specific, the study was focused on the internal cell characteristics and key factors affecting the shape and duration of voltage plateau when voltage plateau occurs. Furthermore, the validity of using the duration of voltage plateau to estimate lithium plating amount is assessed. It has been proved that the duration of voltage plateau depends on the rate of lithium stripping, while the stripping rate is restricted by the capability of Li$^+$ intercalation into graphite. Parameters like intercalation kinetics, solid-state diffusivity of graphite and cell temperature can substantially influence...
the voltage curves even with the same amount of lithium plating. Besides, an interesting phenomenon has been found that during lithium stripping one part of anode near the separator has net oxidation current (local stripping rate > intercalation rate), providing Li\(^+\) ions and electrons to the other part of anode near the foil which has net reduction current.

As Figure 7 shows, metallic lithium at the anode consumes more space than intercalated into the graphite, the volume of the battery will increase in the case of lithium plating. In Bitzer and Gruhle,\(^55\) an experimental setup for precise measurement of the thickness for pouch cells with a resolution of 1 mm was built. The cell thickness as a function of its charge under standard cycling conditions was recorded as a reference. The additional increase in the thickness can be observed in the case of lithium plating, which can be treated as a signal for the non-destructive detection of lithium plating in pouch cells. Compared with the standard capacity-based methods, this procedure has higher sensitivity and the detection process will be much quicker.

In Aneán et al,\(^56\) a commercial graphite LiFePO\(_4\) cell was cycled under dynamic stress test (DST), and a comprehensive analysis for situ lithium plating was conducted. A framework was designed based on incremental capacity (IC) analysis and mechanistic model simulations to quantify degradation modes, relate their effects to lithium plating occurrence and assess cell degradation. The results showed that lithium plating was induced by loss of active material on the negative electrode that eventually led the electrode to over-lithiate. The framework revealed the symptoms of lithium plating formation, and it was proved to be a useful online strategy for early lithium plating detection.

Compared with the most commonly used method EIS, Nonlinear Frequency Response Analysis (NFRA) is not limited to the linear response of the system, as it uses higher sinusoidal excitation currents \(I_{AC}\) of approximately 1.5 C. Higher harmonic response signals \(Y_n\) with \(n_2\) were analyzed, thus additional dynamic information about the system is accessible. In Harting et al,\(^37\) the author detected the Lithium plating process by evaluating the potential of NFRA. NFRA results were analyzed and compared with EIS for cells aged at −10°C with lithium plating occurrence and for cells aged at 25°C without plating. The third harmonic, \(Y_3\), is significantly sensitive to plated lithium of the analyzed batteries at time constants characteristic for electrochemical reactions. As a result, lithium plating causes an increase in charge transfer resistance between the anode and the electrolyte, which affects the nonlinear behavior. Ex situ analysis of this study validated that lithium plating occurs only for the batteries aged at −10°C, but not for the batteries aged at 25°C. Furthermore, it has revealed that \(Y_1\), analyzed with EIS, is not sufficient for the diagnosis of this aging process.

For battery safety operation and to increase the lifespan of Li-ion cells, nondestructive analysis of short-term plating effects was proposed to detect, characterize, and quantify lithium plating in a commercial graphite/LiFePO\(_4\) battery, which was based on a high voltage plateau in the discharge profile after charging at plating conditions. This voltage plateau corresponds to the stripping of plated lithium from the graphite surface. The results showed that differential analysis of such voltage profiles provides a quantitative estimation of lithium plating. The correlation between lithium plating and stripping makes it necessary to distinguish reversible and irreversible plating. Several factors of real-world operations such as charge temperature, state-of-charge (SOC), and charge current, on the plating behavior were investigated to illustrate this degradation mechanism. Furthermore, this approach determines the reversibility of lithium plating.\(^38\)

Based on previous studies mentioned above, a reliable battery thermal management system is urgently needed to avoid low temperature, especially for the safety of electric vehicles. Operation limits, performance, and lifespan should be considered to ensure suitable operational environment. Furthermore, establishing models is essential for monitoring aging effects.

3 | STATE OF HEALTH ESTIMATION

The effects of cold temperatures on lithium-ion batteries appear as an acceleration of losing in capacity and increase in impedance, which is relevant to the SOH estimation of the battery. Generally, SOH estimation for the batteries is needed to estimate the aging or degradation level of the batteries. SOH estimation can help us to predict the time to replace the battery.

The determination of the SOH can be achieved by four methods: methods based on direct measuring, degradation-model-based methods, statistical methods, and adaptive estimation methods. Methods based on direct measuring rely on the collected cycling data. So, the SOH can be calculated very fast.

![Figure 7](image-url) Photo of a plated anode (right) and cathode (left)\(^55\)
The other methods are based on the current data and previous data sets. These methods require a good understanding of the interrelation between real operation and degradation, which consume more computational resource. Details of each methods are described in the section.

3.1 | Direct measuring methods

The simplest and efficient method to monitor the behavior of the cell is to measure some critical parameters (voltage, current, and temperature). Experiments can be beneficial to research and individual analysis. The internal resistance and capacity are direct standards to estimate the SOH of the battery. Remmlinger et al.40 considered that the internal resistance of lithium-ion batteries was a function of temperature and employed a method to measure the resistance. Tsang et al.58 analyzed the relationship between the internal direct current resistance and capacity of lithium-ion batteries. These methods are easy to be understood and implemented, but the internal resistance and capacity measurement require high accuracy which is difficult to meet in actual operation.

3.2 | Degradation-model-based methods

In Tatsuma et al.41 a dual-sliding-mode observer method, which includes a fast speed observer and a slow one, was designed to measure the internal resistance and capacity of lithium-ion batteries timely. The adaptive methods are commonly-used for its advantages of high accuracy, simple structure and this method is easy to realize in actual operation. But these methods are computation-intensive. Another kind of method is based on the modeling of the battery degradation mechanism. Ouyang et al.16 came up with a capacity degradation model based on mechanics which showed us the number of active material and lithium ion inside the lithium-ion batteries. The degradation model based methods have estimation results with high accuracy but are complex and require deep understanding of the electrochemical mechanism to understand it. The state-of-health (SOH) of the lithium-ion batteries can also be estimated by probabilistic and statistical methods, which are shown in Refs.16,59-61 etc. The neural network is an effective method to estimate SOH of lithium-ion batteries. Eddahech et al.63 proposed a battery model based on EIS, and came up with a method of SOH monitoring, which used recurrent neural network to predict the deterioration in performance of the lithium-ion batteries. You et al.64 proposed a datadriven method to estimate SOH of the lithium-ion batteries based on neural network in a practical environment. Wu et al.65 estimated SOH of the lithium-ion batteries through computing data to get the features of the charging curves, and then built a polynomial neural network.

3.4 | Adaptive estimation experimental methods

Although the neural network method showed high nonlinearity and good self-adaption, a huge amount of experimental data was still required to get ideal results. To avoid this problem, the SVM/support vector regression (SVR) method is adopted. The SVM/SVR can fit the regression problem with less experimental data samples compared with the neural network. The method can transfer the nonlinear problem in a low-dimensional space into the linear problem in a higher-dimensional space. Patil et al.66 proposed a novel multistage SVM-based method. The authors used SVM to classify the battery into four types according to the battery cycle times in the first stage and selected some features as SVM’s inputs. Then they used SVR to estimate battery remaining useful life (RUL) in the second stage. Klass et al.67 regarded battery capacity and internal resistance as the evaluation indicators of the SOH, and used SVM to estimate the terminal voltage, which showed high accuracy for estimation.

Based on these SVM methods mentioned before, a novel least square support vector regression (LSSVR) based method is presented to estimate the SOH. Compared to the basic SVR method, LSSVR has faster solving speed and simpler solving process. In this paper, a novel state-of-health estimation method is presented for lithium-ion cells based on statistical knowledge. The preparatory work contains modeling and parameter acquisition. An improved battery model, which combines the advantages of open-circuit-voltage (OCV) modeling and the Thevenin equivalent circuit model, is presented to improve the model accuracy and study the relation between internal parameters and states of the cell. The joint extended Kalman filter-recursive least squares (EKFRLS) algorithm is employed to estimate state-of-charge (SOC) of the cells and identify the model parameters and open-circuit-voltage simultaneously. Then a particle swarm optimization-least square support vector regression (PSO-LSSVR) method is adopted to give a reliable SOH estimation result with high accuracy and good generalization ability. The PSO algorithm is employed to improve the algorithm ability of global optimization.68 The proposed method focuses on the dynamic performance of cells. With the purpose of verifying the accuracy of the
proposed approach, static and dynamic current profile tests are carried out on lithium iron phosphate batteries in different aging levels. And the estimation results are compared with the LSSVR method without optimized by PSO and a neural network method showing the superiority of the PSO-LSSVR approach. The experimental results indicate that the proposed approach can present suitability for SOH estimation with the estimated RMSE less than 2%. Additionally, dynamic tests are carried out to verify the robustness of the proposed approach.

4 | HEATING METHODS

Lithium ion battery suffers decreased power capacity and degradation under subzero-temperatures. Poor performance under low temperatures will significantly hinder the application of EVs. Thus, preheating the ambient temperature to normal range and improving the performance of lithium-ion batteries, will be necessary for a battery thermal management system (BTMS) of EVs at the cold environment. For different operating environments, considering battery life and performance, reliable thermal management systems should have the ability to maintain an appropriate environment. The thermal management system can be divided into passive (PCMs) or active (external or internal heating or cooling battery) thermal management.

4.1 | Passive methods

Passive methods do not consume energy to heat battery pack, one typical and efficient passive method is the phase change materials (PCMs) based thermal management, which is a substance capable of storing and releasing a vast amount of heat when it melts or freezes at the given condition. Seven hundred and forty kinds of PCMs classified by materials were reviewed in order to evaluate various available PCMs. Figure 8 compares various materials. And the solidification properties of PCM can be used to prevent temperature drop.

Thermal management performance of two composite phase change materials was investigated with cooling tests, single-discharge tests, and charge-discharge tests over 20 cycles. Figure 9 shows the cooling curves of three battery packs with different thermal management, the results show that the composite phase change materials can prevent the warm battery pack from cooling too fast. Composite PCM with high thermal conductivity can reduce the temperature change between these batteries, thus helping to reduce the voltage difference and improve the consistency of battery performance. It was concluded that the PCMs plays a more important role in balancing the temperature and voltage distribution between batteries and are less important in keeping batteries warm.
Battery modules with two different phase change materials (pure paraffin and Paraffin-Graphite composite) are partially discharged for some time, and then subjected to different cold periods, representing short and long stops in winter. Results showed that with high thermal conductivity, the 60 wt% RT44HC/EG composite PCM can narrow the temperature variation among cells in each pack. Hence, it will be helpful to reduce the voltage differences. The highly thermally conductive PCM plays an essential role in achieving a uniform temperature distribution to improve the consistency of the battery performance. In Ghadbeigi et al., parameters representing battery performance, such as capacity, power, temperature, and thermophysical performance of each module, are recorded, calculated and compared for different situations. The results show that the paraffin wax does not provide the advantages of regulating the temperature rise in the discharge process or keeping the module warm after a short (10 minutes) cold shutdown.

The effects of thermal conductivity, latent heat, and environmental temperature of PCMs are investigated using the lattice Boltzmann method, and the results show that lower thermal conductivity, greater latent heat, and higher environmental temperature will slow down the solidification process of PCM and keeps the temperature of the cell.

A semiconductor thermoelectric device/ PCM (phase change materials) coupled BTMS (battery thermal management system) was proposed in Song et al. to control the temperature of a base station spare battery pack. As shown in Figure 10, semiconductor thermoelectric devices can cool and heat batteries in a short time. PCM uses the latent heat of phase change to keep the temperature of batteries in the optimum range and obtain uniform temperature distribution during the insulation process. By combining the two components, the heat preservation performance is theoretically improved. However, the basic principle and characteristics of this technology are not clear, and it has not been confirmed in practical experiments and applications. Therefore, further research is needed.

In conclusion, the main contribution of PCM-based thermal management in cold weather is to balance the temperature and voltage distribution between batteries, and the contribution of keeping temperature is not significant.

4.2 | Active heating

Unlike passive heating, active heating consumes energy to heat the battery pack within a short period. Various internal heating strategies, including internal core heating through AC, internal resistance heating and mutual pulse heating, and external heating strategies, such as the use of air and liquid, have been evaluated.

4.2.1 | Internal heating

AC current heating and mutual pulse heating are usually used for internal heating, they have some advantages as follows: First, because the heating system has no convection heat transfer technology, it has high reliability. Second, without any moving parts, the heating system does not need much maintenance. Third, the heating effect is relatively uniform due to the internal heating of the battery.
Finally, the energy efficiency is high, and the heating rate is high. However, some significant shortcomings need to be noted. Inter pulse heating technology requires specially designed circuits and control systems, which significantly increases the cost. Also, when the battery is at a high SOC level, considering the effect of lithium plating, the technology should be used cautiously. Given this risk, a high-frequency pulse is proposed.

According to the electro-thermal coupling (ETC) model, the optimum excitation current amplitude, the appropriate frequency and the optimum variable current amplitude are calculated accurately to achieve the maximum thermal temperature rise. The proposed internal self-heating strategy can heat the single battery from −20.3 to 10.02°C within 13.7 minutes, with the average temperature rise of 2.21°C/min, and even faster when heating a battery pack. As shown in Figure 11, no noticeable capacity is found after 40 cycles with the proposed short and efficient heating strategy.

Zhang et al. claimed that although the existing internal heating methods can heat the battery in a fast way, the conversion percentage of electrical energy into heat is still not high enough, and the structures are sometimes too complicated. They proposed an internal heat structure shown in Figure 12. A nickel foil is embedded inside a cell, and when the temperature is low, the switch will turn on, then the cell works at self-heating mode. The electric current passes through the nickel foil, and heat will be generated 100% from electrical energy. A 1-sheet design is proposed in Wang et al. and the heat generation is not uniform and self-heating process is not fast enough. A 2-sheet design is proposed in Zhang et al., which increased self-heating speed by 45%-56% and reduced energy consumption by 18%-24%.

Considering the structure of a single Ni foil at the middle of the cell, the increase in temperature of internal self-heating mainly depends on the ohmic heat generated in the nickel foil, which causes a sizeable internal temperature gradient from the nickel foil to the outer cell surface. The temperature gradient resulted in lower efficiency of energy utilization, and the current distribution across the cell thickness was found to be non-uniform. The automotive lithium-ion battery cells are becoming larger, and the large size brings a challenge in the aspect of heating. To overcome the problems, Yang et al. compared different heating strategies with the recently developed self-heating method for large-size battery cell. The schematic structure is shown in Figure 13. The technical challenge comes from the increasing cell thickness for traditional lithium-ion battery cell, since the self-heating power is limited to avoid over-heating inside the cell. In contrast, the proposed internal self-heating strategy provide a fast temperature rise of 1-2°C/s, and nearly 100% heat conversion.

Compared with other heating methods, the consumption of proposed self-heating battery in Yang et al. is enormous for the expense of changing the inner structure of the cells. Moreover, for the characteristics of this structure, the implementation needs to be precise, which also increases the cost of a real application.

4.2.2 Convective heating

Air heating method is commonly employed external heating method in EVs because the function can be efficiently designed by heating the pack with a flow of
hot air. The schematic of an air heating is shown in Figure 14. This convection heating method improves the internal and external temperature, which is achieved by both resistance heater and fan. The resist heater transfers electrical energy to heat, and then the fan generates convection for heat transfer from the heater to the air and then to the battery. This method requires a closed system, including flow channels, heaters, fans, batteries, while ensuring minimal heat loss.

Air is usually applied to the pack as transfer media for heating. The hot air flows along the axial direction of batteries, so it is reasonable to assume that the circular-tube annulus flow is between the wall of the pack and the side surface of cells. The flow regime can be designed as laminar or turbulent, which depends on the mass flow rate of the air and the size of the pack. For the airflow around the heater, because of the small wire diameter of the heater coil, the flow can be regarded as flowing over an infinite cylinder. Previous studies were conducted for the parameters of the air heating system considering some factors such as air mass flow rate and wall-cell distances, which concluded that for air heating system, the application of turbulent flow is better than laminar flow.

Comparing with air heating, liquid heating is more efficient, thermal conductivity and the convective heat transfer ratio is higher, so the temperature difference is smaller than air-based systems. A typical pack is shown in Figure 15, the liquid flows under the operation of bump increase the temperature of each cell to reach its ideal temperature range. A series of tests were conducted on the 20-Ah flat cell pack considering several factors, such as ambient temperature, Reynolds number, flow direction, and discharge rate. The results of the simulation indicated that the temperature distribution is more uniform
than the other heating methods, although the equipment is a little complicated. Additionally, the authors found there is no great influence of the Reynolds number for the liquid heating method.

4.3 | Conclusions on heating methods

Different kinds of heating methods have been evaluated by Jaguarmon in.13 By incorporating the research in recent years and the heating rate listed in Table 3, an updated analysis is summarized in Table 4.

5 | CONCLUSIONS

In this paper, a comprehensive analysis of the effects of low temperatures on lithium-ion cells, the mechanisms and detection methods of lithium plating, the estimation of performance parameters of lithium-ion batteries and heating methods for low temperatures is conducted. The review illustrates the two principal problems related to the operation at cold environment: performance loss and degradation. These phenomena make it difficult to expand of the electric vehicle market, particularly for some cold-weather countries and subzero temperatures in winter. Therefore, an efficient thermal management system, a reliable estimation approach and a stable method of heating are necessary to solve these harmful actualities.

Some conclusions from the current study is drawn as follows: (a) the internal resistance and capacity are direct standards to estimate the SOH (state of health) of the cells; (b) Electrochemical Impedance Spectroscopy has received relatively wide application for the detection of lithium plating; (c) adaptive sliding observer method is the most commonly used approach compared with the other approaches for estimation, with high accuracy results and of simple structure; and (d) Although liquid heating is usually chosen for low-temperature conditions, internal self-heating would be one good option, since it guarantees a high conversion percentage.

Nevertheless, some challenges still exist, because the estimation methods are either not accurate enough or cannot be used in real time; and the battery pack with external heating system consumes lots of space and energy in an electric vehicle, while the internal self-heating method also faces the challenge of high process level and maintenance cost.

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