Detecting undesired lithium plating on anodes for lithium-ion batteries – A review on the in-situ methods

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HIGHLIGHTS

- Literature on in-situ detection methods of lithium plating is reviewed.
- The development of lithium plating is divided into 3 stages based on its reversibility.
- Principles, deficiencies and features of the existing methods are discussed.
- Prospects of future detection for on-board application are presented.

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ABSTRACT

Lithium plating on anodes, which can happen during fast charging and low-temperature charging, and/or after long-term cycling, plays a crucial role in the aging of lithium-ion batteries (LIBs) and leads to irreversible capacity fade and severe safety hazards. This study systematically reviews the recent progress in developing methods for in-situ detecting lithium plating in order to provide guidelines regarding selecting proper methods for on-board applications. In general, lithium plating can be divided into three stages according to the damage level. There are two categories of methods, electrochemical methods and physical methods, which can be used to detect lithium plating. Their principles, features, and limitations have been thoroughly analyzed. Trends for the prospective development of novel technologies are also discussed.

1. Introduction

Lithium-ion batteries (LIBs) are the most widely used power source for electric vehicles (EVs) [1], due to the advantages of the high energy density, environmental friendliness, long cycle life, low rate of self-discharge, and low cost [2]. The loss of lithium inventory (LLI) is one of the main mechanisms about the performance decay and aging of LIBs [3], usually as a result of the decomposition of electrolyte, formation of solid electrolyte interphase (SEI) [4], and lithium plating on the anode [5]. Among these factors, lithium plating can cause irreversible capacity fade and lead to safety hazards, and therefore plays a dominant role in the degradation of LIBs. Lithium plating usually occurs in fast charging [6], low-temperature charging [7], and/or long-term cycling, and it occurs locally where the negative electrode potential is below 0 V vs. Li/Li$^+$ [8].

The plated lithium could not only react with electrolyte [9], but also become electrically isolated from anodes (known as "dead Li") [10], both of which lead to the loss of active lithium and capacity irreversibly. In addition, Li dendrites formed during plating could penetrate the separator [11], causing internal short circuits [12]; and the active plated lithium could reduce the onset temperature of thermal runaway [13], leading to safety issues.

Considering the negative impacts of lithium plating, a reliable, in-situ, and real-time detection method is critical for developing fast and safe charging processes. For example, accurate detection can also be used to identify the maximum charging rate that does not cause lithium plating under different conditions. Moreover, in-situ and quantitative detection enables the real-time estimation of the aging state of LIBs by analyzing the amount of plated lithium, and real-time detection can improve charging safety of LIBs by adjusting the charging strategy

according to the occurrence of lithium plating [14]. Whereas, the detection of lithium plating is very challenging because a lot of factors could affect plating, including temperatures, charging rates, states-of-charge (SOC), cell designs [5], electrolyte composition [15,16], anode capacity [17], areal capacity ratio of negative to positive electrodes (N/P ratio) [18], cell defects [19], etc. Many efforts have been dedicated to the development of detection methods, however, some methods can only be used in the laboratory, in which experimental conditions can be controlled and various instruments are available [20]. For on-board applications, batteries are working under unpredictable conditions [21], and it is more difficult to detect lithium plating due to fewer available instruments and measured parameters. Therefore, it is of great importance to assess the feasibility and robustness of the detection methods that can be used on vehicles.

Even though there have been a few of reviews about lithium plating, as summarized in Table 1, some knowledge gaps can still be clearly seen. This paper is the first review work that studies different lithium plating detection methods for on-board applications in order to meet the urgent need of the EV industry for fast and safe charging, especially in low-temperature environments. In general, in-situ lithium plating detection methods can be divided into two categories, electrochemical methods and physical methods. Features related to on-board applications, which have not been mentioned in the previous reviews, are systemically analyzed. In addition, lithium plating is a cumulative process of damages, so its impact on battery lifetime and safety is changing

Table 1
A summary of reviews about lithium plating.

Ref	Scope	Main findings	Knowledge gaps
Li, et al., 2014 [22]	Solutions to suppressing lithium plating in Li-ion batteries and Li-metal batteries were reviewed.	The solid electrolyte interphase (SEI) plays an important role in plating for Li-metal batteries, and self-heating methods for Li-ion batteries may suppress lithium plating.	The detection methods for lithium plating are not included.
Liu at al., 2016 [5]	Factors influencing lithium plating in Li-ion batteries and some detection methods were reviewed.	Electrolyte compositions, anode materials, cell designs, and charging protocols should be optimized to restrain lithium plating.	The applicability, detection accuracy, merits, and limitations of each detection method are not discussed.
Waldmann et al., 2018 [23]	Morphologies, detection methods, and influences on battery aging and safety of lithium plating in Li-ion batteries were reviewed.	Lithium plating is affected by a lot of factors from the microscopic level to the macroscopic level, and operating conditions have significant influences on lithium plating.	The applicability, detection accuracy, merits, and limitations of each detection method are not mentioned.
Foroozan et al., 2020 [24]	In-situ imaging techniques for studying lithium plating in Li-metal batteries were reviewed.	Among different in-situ imaging methods, electron microscopy has the highest resolution, while optical imaging and resonance-based imaging techniques have the lowest.	No methods suitable for on-board applications were included.
Janakiraman et al., 2020 [25]	Common detection methods and models of lithium plating were reviewed.	Electrochemical detection methods are more preferable for real applications.	Clear classification and comprehensive comparison of different methods are lacking.

with the development of plating. This work also proposes a method that divides the plating into 3 stages according to the progress. More importantly, suitable detection methods that can be used in each stage have been identified and their pros and cons are compared.

The rest of this review is organized as follows: the process of lithium plating is introduced in Section 2; principles and pros and cons of electrochemical methods and physical methods are reviewed in Section 3, and Section 4 respectively; Section 5 compares different methods and prospects for future development trends are presented; conclusions are drawn in Section 6.

2. Lithium plating in LIBs

When a LIB is charged under normal conditions, active Li^+ ions are de-intercalated from the cathode and intercalated into the graphite anode directly, as shown in Fig. 1a. The corresponding reaction is:

$$6C + xLi^+ + xe^- \rightarrow Li_xC_6 \tag{1}$$

When a LIB is charged under harsh conditions (i.e. fast charging, low-temperature charging, etc.) and/or after long-term cycling, lithium plating can occur on the surface of anodes in LIBs, which experiences three steps, as illustrated in Fig. 1b, c, and d.

Step 1: When a LIB is charged under plating-induced conditions, the polarization increases and thus the anode potential decreases, and lithium plating occurs until the anode potential is below 0 V vs. Li/Li$^+$. It is worth mentioning that lithium plating and intercalation coexist during charging [26], as shown in Fig. 1b. Most of the plated lithium in this step is reversible, according to the following reaction:

$$Li^+ + e^- \rightarrow Li_{rev}^0 \tag{2}$$

where Li_{rev}^0 is the reversible plated lithium.

Step 2: Lithium stripping and chemical intercalation are two relevant reactions associated with the reversible plated lithium, as shown in Fig. 1c. In the following discharge phase after plating, the reversible plated lithium will convert into Li^+ and enter electrolyte, which is called lithium stripping. In the tapering charging or the following rest phase after Step 1, the reversible plated lithium can also dissolve and be chemically intercalated into the adjacent anode, which is called chemical intercalation. Both reactions lead to an anode potential plateau and have the same equation:

$$Li_{rev}^0 \rightarrow Li^+ + e^- \tag{3}$$

At the beginning of the lifecycle, all plated lithium is reversible. As the lifecycle progresses, only part of the plated lithium remains reversible.

Stage 3: The amount of irreversible plated lithium grows with the use of LIBs. Lithium stripping usually occurs from the surface of the anodes, which makes the top of plated lithium easy to become electrical isolated from anodes and be inactive, which is called "dead Li", as shown in Fig. 1d. "Dead Li" will not participate in the charge and discharge process, resulting in irreversible loss of active lithium, and the reaction is:

$$Li_{rev}^0 \rightarrow Li_{irr}^0 \tag{4}$$

where Li_{rev}^0 is the irreversible plated lithium.

Moreover, reversible plated lithium can react with electrolyte, and new SEI film is formed and both electrolyte and active lithium are consumed by this side reaction, leading to capacity fade of LIBs [27]. The new SEI film is formed on the surface of the plated lithium that grows through the original SEI. It can result in an increase in the total thickness of the SEI film, which in turn leads to an increase in the internal resistance of LIBs. The reaction is:

$$Electrolyte + Li_{rev}^0 \rightarrow SEI \tag{5}$$

The reversibility of lithium plating, which is defined as the ratio of
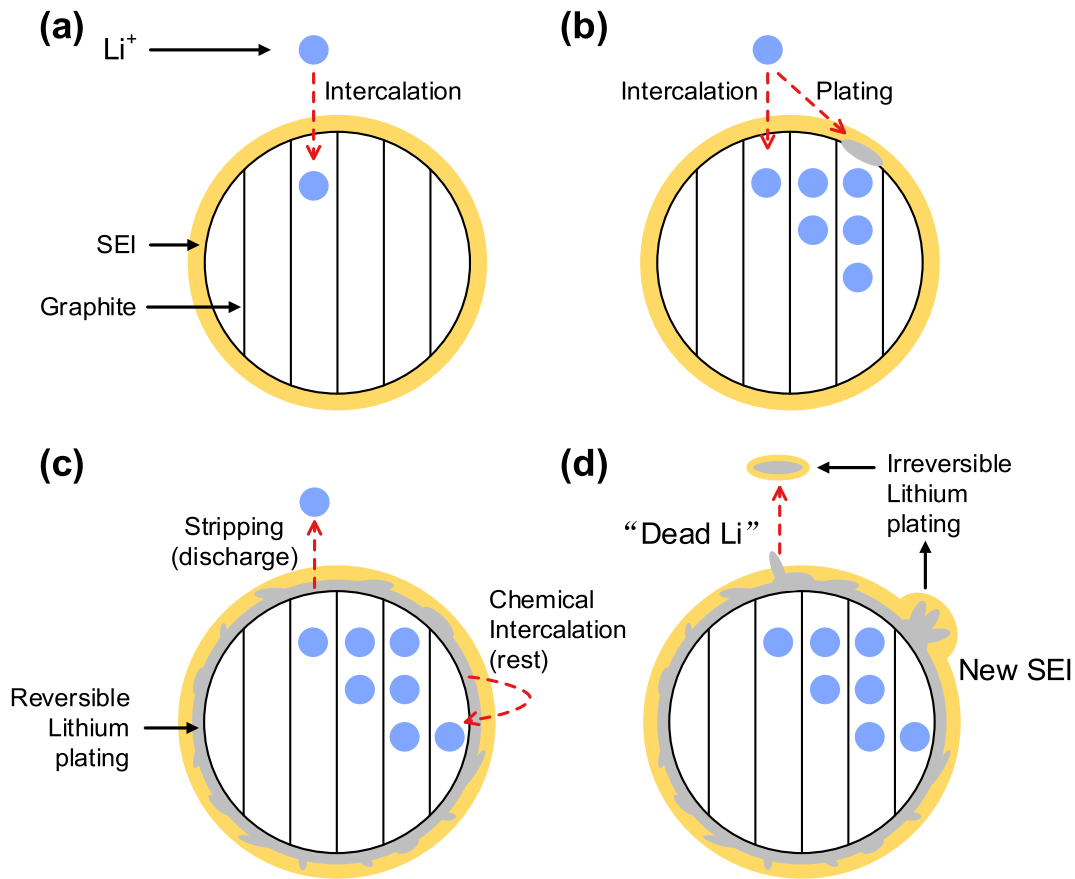
Fig. 1. Steps of lithium plating on the surface of anodes in LIBs. (a) Normal step: charge under normal conditions; (b) Step 1: the occurrence of lithium plating; (c) Step 2: reactions associated with the reversible plated lithium; (d) Step 3: reactions associated with the irreversible plated lithium.

the amount of the reversible part to the total amount, reflects the capacity fade of LIBs. The total quantity of plated lithium includes both the reversible and irreversible . According to the negative impacts, the development of lithium plating can be divided into 3 stages based on its reversibility: initial stage, moderate stage, and serious stage, as shown in Fig. 2. In the initial stage, $R_{initial}$, the reversibility of lithium plating, is 100%. For the boundary between the moderate stage and serious stage, $R_{serious}$, 90% is chosen according to [28]. In the initial and moderate stages, the performance of LIBs will not be significantly affected. But in the case of the serious stage, the amount of irreversible plated lithium increases rapidly, and the capacity and performance of LIBs will decline obviously.

3. Electrochemical methods for lithium plating detection

Lithium plating has a direct impact on the electrochemical process, thereby affecting electrochemical parameters, such as voltage, capacity,
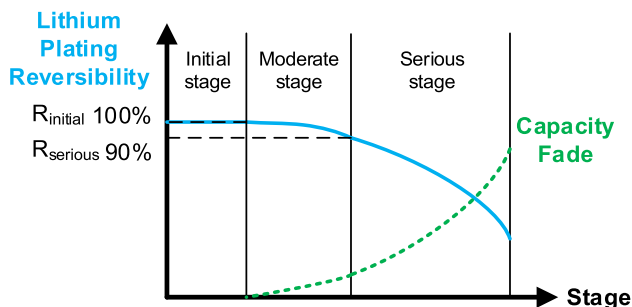

Fig. 2. Three stages of lithium plating.

impedance, etc., of LIBs during operation and relaxation. Due to the easy accessibility and good measurability of the electrochemical parameters, electrochemical methods are promising for on-board applications, which can be implemented in BMS of EVs to detect lithium plating in real-time and under actual driving conditions.

Electrochemical methods encompass voltage-based methods, capacity-based methods, impedance-based methods, and methods based on other electrochemical characteristics. The principles and features of these methods are discussed below.

3.1. Voltage-based methods

Methods based on voltage signals can be adopted in situ for on-board applications. Five voltage-based methods have been proven feasible, including methods based on anode potential measurement, differential voltage analysis (DVA), Voltage relaxation profile (VRP), differential charging voltage (DCV), and average voltage inspection. A brief overview of these voltage-based methods is given in Table 2.

3.1.1. Anode potential measurement

Lithium plating is caused by an anode potential below 0 V vs. Li/Li$^+$; therefore, the anode potential vs. Li/Li$^+$ could be applied to detect it. Nevertheless, for normal cells, only the potential difference between cathode and anode, also known as cell voltage, can be measured. To solve this, a reference electrode (RE), which contains lithium metal and represents the reference potential of Li/Li$^+$, can be inserted into the battery to form a 3-electrode cell. A schematic diagram of the 3-electrode cell is shown in Fig. 3a. During charging, the potential difference between the anode and RE is measured to obtain the anode potential against Li/Li$^+$; and consequently, lithium plating can be

Table 2
An overview of voltage-based methods.

| Method	Procedure	Applicability	Output
Anode potential measurement	Plating can be detected during charging by measuring the anode potential.	It can be used to detect the initial stage.	Qualitative results
DVA	Plating can be detected in the following discharging by measuring voltage and current.	It can detect the moderate stage.	Quantitative results
VRP	Plating can be detected in the following relaxation by measuring voltage and time.	It can detect the moderate stage.	Quantitative results
DCV	Plating can be detected during charging by measuring voltage and current.	It can detect the moderate stage.	Qualitative results
Average voltage inspection	Plating can be detected after long-term cycles by measuring voltage.	It can detect the serious stage.	Qualitative results
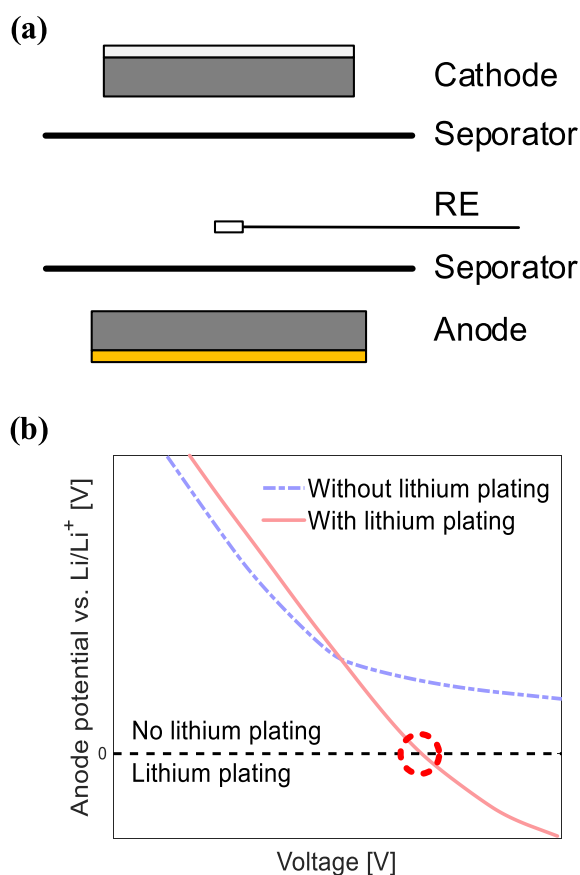
Fig. 3. (a) Schematic diagram of the 3-electrode cell. (b) Detection principle of the anode potential measurement [29] (the red circle highlights the occurrence of lithium plating).

detected in situ and in real-time. As shown in Fig. 3b, during charging, the anode potential decreases with the increase of voltage and SOC. If lithium plating occurs, the anode potential will be below 0 V vs. Li/Li$^+$, as highlighted in Fig. 3b, which can be used to detect lithium plating.

It is worth to point out that when using the anode potential measurement method, an improper location of RE will result in response artifacts [30]. Dess et al. [31] found an internal RE is preferred which is sandwiched between two electrodes with two separator layers to avoid

errors. In addition, RE is usually located near the anode to minimize the Ohmic drop. Besides, it could be positioned near the tab of the anode which has the highest current density thus the lowest local potential, as lithium plating is the most serious near the tab [32].

In addition, to minimize the measuring errors, a good RE is expected to meet the following requirements: non-polarizable, reproducible, having low impedance, keeping stable and definite potential even when the temperature and current are changed [33,34]. RE is usually a wire, which should be covered with polyurethane to prevent mixed potentials that exist along the wire and lead to erroneous data [35]. Lithium is the most commonly used material for RE [18,29,36]. However, lithium has two problems: i) it can only be prepared in an inert atmosphere [35], and ii) when measuring the reference potential, lithium may lead to a measurement error due to the formation of the SEI film and solvated electrons on the lithium RE, which causes a shift of the potential of lithium with time [33]. Compared with lithium, lithium alloys and compounds show some advantages when used as RE, such as providing more stable potential during long cycling, and being less likely to occur self-delithiation surface reactions [30]. Lithium alloys that have been verified include lithium-tin (Li-Sn) alloys [35-40], lithium-gold (Li-Au) alloys [30], lithium-bismuth (Li-Bi) alloys [34], lithium-copper (Li-Cu) alloys [41], etc.; and lithium compounds include Li$_4$Ti$_5$O$_{12}$ [33,42] and LiFePO$_4$ [33]. It should be noted that Li-Sn RE exists a gradual drift in its potential during continuous detections under long cycling, a second lithium RE is recommended to monitor anode potentials reliably [40]. When two REs are used, lithium RE is usually used to determine electrode potentials and Li-Sn RE can be used for impedance measurement [38].

Although using RE can detect lithium plating in situ and in real-time, there are still some disadvantages, which are summarized as follows:

(1) Application: i) it needs cell modification to add RE; ii) it can only detect if lithium plating appears or not, but cannot provide any quantitative results, such as how much lithium is plated; iii) the detection repeatability is poor at low temperatures and high currents; and iv) this method is recommended for brand new cells or cells at the beginning life and not suitable for aging cells [43,44].

(2) Accuracy: i) electrochemical processes in cells are affected by the introduction of the RE [31,45]; and the shape, dimension, and assembly process of RE will all lead to anode potential artifacts; and ii) there is always a difference between the potential of RE and anode due to the Ohmic drop, which is influenced by the distance between RE and anode and the applied current [41]. Such a difference leads to an extra unwanted potential in the measured anode potential and therefore introduces an extra error.

3.1.2. Differential voltage analysis (DVA)

DVA is an approach that can in situ detect lithium plating quantitatively by calculating the derivative of the voltage V to the capacity Q during discharging at a constant low rate [46]. If lithium plating occurs during charging, a high voltage plateau will appear in the following discharge step, which indicates the complete oxidation or stripping of reversible plated lithium [28]. This high voltage plateau always appears prior to the first normal de-intercalation plateau of graphite anode [28], because the stripping potential is around 80 mV lower than the potential of graphite de-intercalation stage 1 [47]. But the voltage plateau is not easy to obtain, so DVA is proposed to transform the plateau to identifiable differential voltage (DV) peaks, and lithium plating can also be easily detected by the peak.

The peaks on the DV curves reflect the lithium intercalation/de-intercalation processes [48], and the peak shift and changes of corresponding capacity are used to analyze the contributions of different aging mechanisms [49], including lithium plating. As illustrated in Fig. 4, the first additional peak on the beginning of DV curve indicates
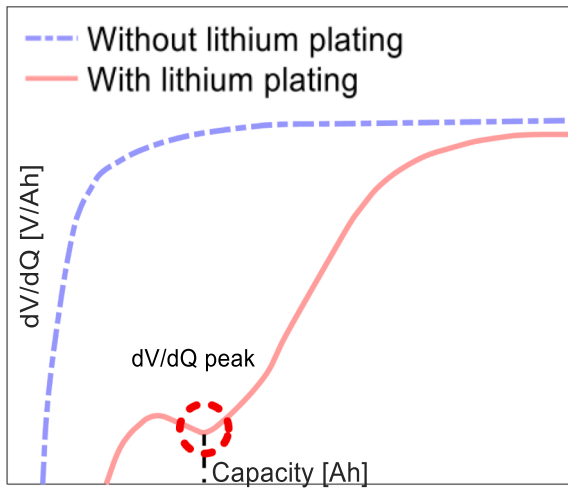
Fig. 4. Detection principle of DVA method [50] (the red circle highlights the occurrence of lithium plating).

the presence of lithium plating prior to charging, and the quantity of plated lithium is proportonal to the corresponding capacity of this additional peak. It is noteworthy that DVA can only detect the reversible lithium plating quantitatively because this additional peak used for detection is caused by the stripping process. For cells without lithium plating, no additional peak occurs at the beginning of the DV curve, indicating the absence of lithium plating. Although DVA is unable to detect irreversible lithium plating, it can be used to calculate the reversibility of lithium plating to identify stages and to qualitatively confirm the existence of the irreversible lithium plating.

DVA is effective for different cell chemistries (such as lithium iron phosphate (LFP) [28,51] and nickel manganese cobalt oxide (NMC) cathodes [50,52]), and different cell geometries (such as cylindrical cells [28,51] and pouch cells [50]), under different conditions (such as low-temperature charging conditions [28] and long-term aging conditions [51]).

To detect lithium plating accurately by DVA, it is important to discharge the cell immediately after charging to avoid the influence of chemical intercalation on the detection of DVA. And the discharging shall be controlled at a sufficiently small rate to hold a quasi-equilibrium state. Moreover, the measured voltage data contain noise that makes it difficult to identify plateau and *dV/dQ* peaks directly. Therefore, the measured data should be smoothed or fitted by a high-order polynomial [53].

The advantage of DVA is that it only requires voltage signals that could be collected by the Battery Management System (BMS), making DVA a promising method for EV application. However, there are still some disadvantages:

(1) Application: i) the contributions to the DV curves of cathode should be as small as possible when using DVA to detect lithium plating which occur at the anode, so it's more suitable for cells with stable cathode [49] or using half-cell to study the DV curves of the cathode; and ii) it takes a long time , which can be up to several hours, to detect lithium plating because the discharging rate should be kept very low.

(2) Accuracy: i) the position of *dV/dQ* peak for plating detection can change under different discharging rates even after the same charge procedure and for the same amount of plated lithium, leading to high detection uncertainties [52]; and ii) DVA cannot detect lithium plating when the amount of plated lithium is small or below a certain threshold [54], resulting in an underestimation, for example, plated lithium of about 65mAh in a 7.5 Ah LIB cannot be detected by DVA [50].

3.1.3. Voltage relaxation profile (VRP)

VRP is another approach that can in situ detect lithium plating quantitatively by using the derivative of the voltage *V* to the time *t* during relaxation. After plating-induced charging, the plated lithium will chemically intercalate into direct contact graphite during the following relaxation [55], and the anode potential will remain at 0 V vs. Li/Li^+ during the chemical intercalation [56]. Similar to the stripping plateau used by DVA, VRP calculates *dV/dt* to resolve the chemical intercalation process in the relaxation step. The peak of *dV/dt* indicates the end of dissolution of plated lithium and the amount of reversible plated lithium in the previous charge step is dependent on the peak position. Fig. 5 shows the detection principle of VRP. The *dV/dt* peak indicates the presence of lithium plating in the previous charging, and the peak position *τ* is used to quantify the amount of reversible plated lithium. The larger *τ*, the more plated lithium in the previous charge step.

VRP has additional benefits over DVA. Because there is no net current in case of relaxation and thus no overpotential is introduced, the detection results are not affected by the selection of discharging rate [57]. However, disadvantages still exist:

(1) Application: the detection time of VRP is long. Schindler et al. [57] found that it took even longer time to get the peak of *dV/dt* in relaxation than to get the peak of *dV/dQ* in stripping discharge with 0.025C used for DVA.

(2) Accuracy: it is found that the peak position *τ* can be impacted by many factors, including the charging rate prior to the appearance of plating [58], the lithium stripping of plated lithium in the anode during relaxation [52], and the aging degree of cells [56].

3.1.4. Differential charging voltage (DCV)

DCV detects lithium plating based on the first and second derivative of the voltage *V* to the capacity *Q* during charging. An additional peak in the curve of the first derivative *dV/dQ* and an intersection between the curve of the second derivative d^2V/dQ^2 and the zero line indicate the onset of lithium plating [59].

Adam et al. [59] proposed DCV to detect lithium plating in fast charging at 25 ℃. Compared with DVA and VRP, DCV is a time-resolved method that can detect lithium plating during charging directly, and the detection time is shorter since there is no restriction of low rate or long-term relaxation. Some limitations are shown below:
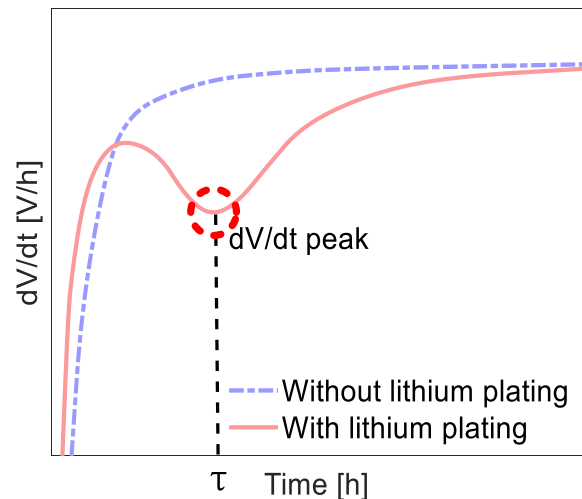

Fig. 5. Detection principle of VRP method [56] (the red circle highlights the occurrence of lithium plating).

(1) Application: theoretical explanations are still missing regarding the correlation between the first and second derivative curves and lithium plating.

(2) Accuracy: i) detection at low temperatures has not been studied yet; and ii) applicability for different cell chemistries and geometries remains unknown.

3.1.5. Average voltage inspection

It is known that the average voltage of a LIB is affected by two main factors: internal resistance and lithium inventory. The internal resistance increase will lead to an increase in the average voltage in both charge and discharge, while LLI will result in a decrease [60]. LLI is mainly caused by SEI formation and lithium plating; hence, the change of average voltages can be used to detect lithium plating.

Harlow et al. [60] proposed a new method to detect lithium plating in commercial cells under the aging process by using average charge voltages ($V_{av,c}$) and discharge voltages ($V_{ac,d}$). The changes in average voltage attributed to resistance increase) and LLI were investigated respectively. It was found that the sharp increase of ($V_{av,c} - V_{ac,d})/2$, was a precise sign of lithium plating, as shown in Fig. 6.

The inspection of average voltages is a simple method, which only needs common instruments to measure the voltage. However, as a new approach, the mechanism and application of this method have not been systematically studied:

(1) Application: it is only suitable for the lithium plating detection during cycle aging, and the applicability in a single cycle is still unknown;

(2) Accuracy: there has been no available result about the performance when using the average voltage method for different cell chemistries.

3.2. Capacity-based methods

Same as voltage signals, capacity signals can be obtained directly from BMS, so capacity-based methods can also be potentially used for on-board application. Three feasible methods are developed, including methods based on incremental capacity analysis (ICA), coulombic efficiency (CE), and capacity difference analysis (CDA). A brief overview of these voltage-based methods is given in Table 3. More details are discussed below.

3.2.1. Incremental capacity analysis (ICA)

Similar to DVA, ICA uses the peaks of incremental capacity (IC), which differentiate the capacity as a function of cell voltage [61,62], to
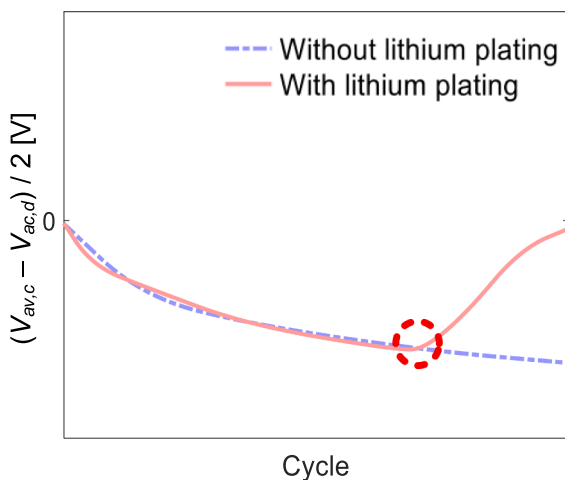
Table 3
An overview of capacity-based methods.

Method	Procedure	Applicability	Output
ICA	Plating can be detected in the following discharging by measuring capacity and voltage.	It can detect the moderate and serious stages.	Quantitative results
CE	Plating can be detected in a charge/discharge cycle by measuring charge and discharge capacity.	It can detect the moderate and serious stages.	Quantitative results
CDA	Plating can be detected after long-term cycles by measuring capacity.	It can detect the serious stage.	Qualitative results

detect the occurrence of lithium plating. The features of peaks on the IC curves due to reversible and irreversible are different, as shown in Fig. 7. For reversible lithium plating, a new peak will appear at high voltage in the IC curve; while for irreversible lithium plating, the intensity of peak 1 will continuously decrease. Therefore, a strong reduction of peak 1 can be observed and a new peak 0 appears due to lithium plating. In ICA, the reduction of peak 1 is derived from LLI [63], usually originating from the SEI formation and "dead Li", both of which are caused by lithium plating.

ICA can be used to analyze the electrochemical reactions in cells by extracting the magnitude, position, or area of the peaks on the IC curves [65,66], and the amount of plated lithium can be reflected by the area of the corresponding IC peak [28]. Similar to DVA, only the quantity of the reversible lithium plating can be detected. In addition, ICA is applicable for detecting plating in cells with different geometries, different sizes, and different electrode materials [67,68]. Furthermore, compared with DVA, ICA can calculate the stripping voltage of lithium plating and is more suitable for detecting lithium plating at the serious stage in long-term aging cycles [69] and aging due to fast charge [64].

The shortcomings of ICA are:

(1) Application: i) ICA requites cycling with low current to reduce resistive and diffusion effects, which presents challenges in EV application [70]; and ii) the implementation of ICA needs high computational complexity and efficiency [71];

(2) Accuracy: IC curves usually contain noise, but the current filtering methods are complex, time-consuming, and not suitable for different charging strategy when the filter is optimized for one particular task [66,71].
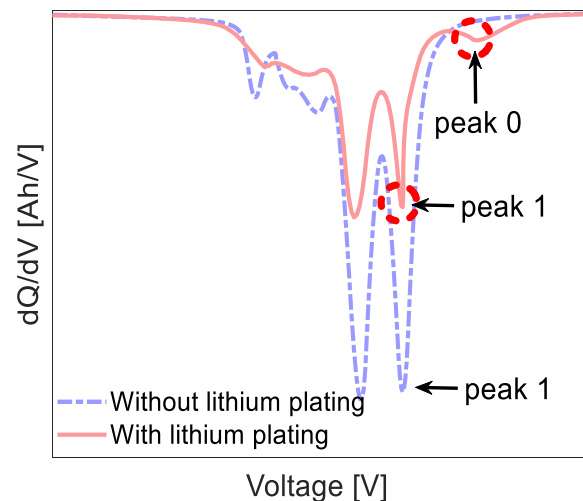

Fig. 6. Detection principle of average voltage inspection method [60] (the red circle highlights the occurrence of lithium plating).

Fig. 7. Detection principle of ICA method [64] (the red dotted circles highlight the occurrence of lithium plating).

3.2.2. Coulombic efficiency (CE)

CE, a crucial parameter which is used to characterize the degradation of cell capacity, is defined as:

$$CE = \frac{Q_{dc}}{Q_{ch}} \qquad (6)$$

where Q_{dc} is the discharge capacity at a cycle, and Q_{ch} is the charge capacity at the same cycle. The turning point of CE represents the onset of lithium plating, because CE is highly influenced by LLI [72], and LLI is deeply affected by the irreversible lithium plating. The quantity of the irreversible lithium plating is proportional to Q_{ch} - Q_{dc}. As shown in Fig. 8, the evolutions of CE are plotted with respect to the charging rates, and a turning point occurs at the high charging rate that indicates the appearance of plated lithium.

CE is more effective than DVA and VRP for the detection of irreversible plated lithium, which can detect lithium plating at moderate and serious stages. Moreover, it is reasonable to ignore other aging mechanisms only in several cycles in a stable environment, so only lithium plating is detected. However, there are some drawbacks as well:

(1) Application: i) the cell needs to be fully charged and discharged to calculate capacities, which is impracticable for on-board applications; and ii) CE is affected by many parasitic reactions, so it's not suitable for cells after long-term cycling or in the unstable environment, which could lead to other side reactions.

(2) Accuracy: i) the change of CE due to lithium plating in a single cycle is so small and sensitive to many factors [74]; therefore, accurate set currents, precise voltage measurements and strictly controlled temperature are required [75,76]; and ii) CE of commercial cells is too high for most equipment and BMS to measure accurately [77], the strict requirements further lead to the need of high precision instruments, which are costly.

3.2.3. Capacity difference analysis (CDA)

CDA is a new electrochemical method to characterize the occurrence of a deposition layer on the surface of anode for cyclic and calendric aged cells. The cells are first discharged at a low rate (e.g. 0.25C) and then at a high rate (e.g. 1C), and the capacity difference (CD) is determined by $Q_{low} - Q_{high}$. It is found that the increase of CD indicates the presence of a dense deposition layer [78], which is likely attributed to the formation of lithium plating. This method is simple and easy to implement but has not been well studied. Further efforts about the repeatability and accuracy are needed. The authors [78] suggest that CDA is best used for lithium plating detection during fast charging at low temperatures. In this case, lithium plating plays a dominant role in the
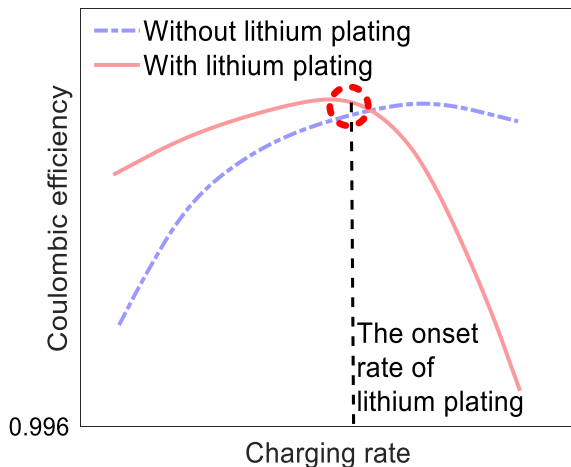
aging of LIB, avoiding the interference of other aging factors on the detection. Some disadvantages are listed as follow:

(1) Application: it is difficult to distinguish the contributions of SEI and lithium plating to the detected deposition layer on the surface of anode, so CDA is only suitable for harsh charging conditions, under which lithium plating is dominant.

(2) Accuracy: it can only detect lithium plating at the serious stage after long-term cycling, and only qualitative information can be obtained.

3.3. Impedance-based methods

The occurrence of plated lithium will affect the impedance of LIBs through the formation of surface films and electrolyte degradations [79]. Therefore, tracking the impedance of cells is also feasible to detect lithium plating. There exist two impedance-based detection methods in the literature, both of which can detect lithium plating qualitatively. It should be noted that once lithium plating can be detected by impedance-based methods, it implys that plating has developed to the serious stage.

3.3.1. Electrochemical impedance spectroscopy (EIS)

Electrochemical impedance spectroscopy (EIS) is an effective and accurate method to evaluate the internal electrochemical process in a LIB. By imposing a small sinusoidal current or voltage signal, the resulting output is measured and the phase and magnitude of impedance signal can be obtained at the given frequency. Usually, EIS is performed at a wide range of frequencies, from mHz to kHz [80]. The results are presented and decomposed through a Nyquist plot, which allows the quantitative separation of contributions of different elementary electrochemical processes [81], including lithium plating, as shown in Fig. 9.

EIS has been used to detect lithium plating in cells during relaxation and cycle aging. As for cells in the relaxation phase, lithium plating can be identified by two characteristics: a reduction of R_{ohm} and a shrinkage of the arc corresponding to the charge transfer process [57]. As for cyclic aged cells, especially those under fast charging or low-temperature charging conditions, there are also two features used to detect lithium plating: increases in R_{ohm} [82] and R_{SEI} [83]. An increase in R_{ohm} with increasing cycle numbers at plating conditions can be applied as an indicator of lithium plating, because the ohm resistance rise highly depends on the decreasing ionic conductivity of the electrolyte, which is caused by the formation of passive films on the surface of plated lithium. Additionally, the formation of passive films on the surface of plated lithium also leads to an increase in R_{SEI}, which is highly related to LLI [83].

EIS can qualitatively detect lithium plating in situ and without destruction. Furthermore, LIB degradations due to lithium plating can also be detected simultaneously. However, there are still some weak points of EIS:
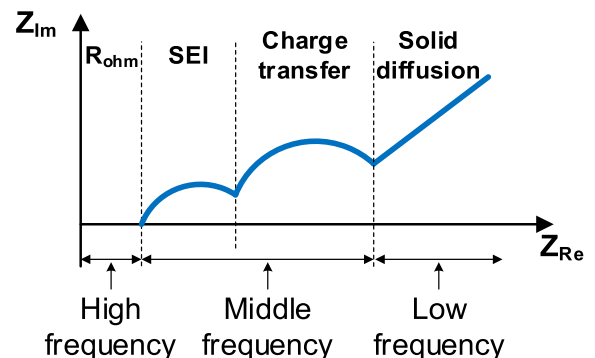
Fig. 8. Detection principle of CE method [73] (the red circle highlights the occurrence of lithium plating).

Fig. 9. Relationship between EIS and electrochemical processes.

(1) Application: i) EIS is mainly used for offline testing, which is time-consuming and requires large computation [84]; and ii) the need for auxiliary equipment hinders the on-board implementation of EIS.

(2) Accuracy: the impedance spectrum is clearly affected by the cell chemistry [85], thus the detection results of different chemistries of LIBs are very different.

3.3.2. Cell impedance estimation

Koleti et al. [86] proposed a new method to detect lithium plating by interrupting the charging currents for 3 seconds for every 1% increase of SOC to estimate the impedance during charging and without auxiliary equipment. Compared with cells at low charging rates, the impedance profiles of cells at high charging rates shows a steep decline, which represents the occurrence of lithium plating.

When lithium plating occurs, the applied charging currents are divided into normal intercalating current and plating current, thus a new impedance is introduced in parallel to the charge transfer limitations (R_{CT}) at the anode [86], and R_{CT} will decrease [87]. In addition, as shown in Fig. 9, the impedance in the middle-frequency region (R_{mid}) is introduced by R_{CT} and the ion migration through SEI (R_{SEI}); and R_{mid} is dominant in the whole impedance. It is assumed that the change of R_{SEI} is ignorable at a single cycle because there is no significant change of SEI thickness at only one cycle [88]. Therefore, R_{CT} contributed largely to the R_{mid} variations, thus to the whole impedance changes. Based on this, it is possible to detect the onset of lithium plating in real-time by detecting the decrease of impedance during charging.

This approach has the potential to be implemented on board. But as a new method, the problems needed to be solved include:

(1) Application: this impedance estimation method prolongs the charging time.

(2) Accuracy: the performance of detection for aged cells and different cell chemistries is unknown.

3.4. Other methods

In addition to voltage, capacity, and impedance signals, other electrochemical signals affected by lithium plating can also be used to develop detection methods.

3.4.1. Nonlinear frequency response analysis (NFRA)

NFRA is a novel method to analyze nonlinear behaviors of cells [89]. To detect lithium plating by NFRA, a sinusoidal current is applied with a large amplitude at a given frequency. The voltage output is measured and transferred from time to frequency domain by a Fast Fourier Transformation (FFT), and the high harmonics responses from Y_2 to Y_n are analyzed [90]. Harting et al. [91] found that the intensity of high harmonic response Y_3 in the middle-frequency range would increase and overtake that of Y_2 if lithium plating occurred. Therefore, the increase of the intensity of Y_3 in the middle frequency range is an effective indicator of lithium plating.

NFRA has some unique advantages over EIS. In contrast to EIS, which is limited to the linear response of the system, NFRA can be used to identify and characterize electrochemical reactions, which are usually nonlinear processes, thus can investigate the nonlinear behavior and estimate the state of LIBs [92,93]. Except for the mentioned nonlinear response analysis, NFRA does not require an equilibrium state of the cell, which is inevitable for DVA, ICA, and EIS. Moreover, NFRA is fast. For example, one test takes only 15 min [91]. Thereby, NFRA is a promising method to detect lithium plating qualitatively. Nevertheless, the shortcomings of NFRA include:

(1) Application: i) only lithium plating at the serious stage can be detected; and ii) there is no theoretical explanation for the correlation between Y_3 and lithium plating.

(2) Accuracy: like EIS, the chemistry and aging degree of LIBs can strongly affect the detection performance of NFRA.

3.4.2. Electrochemical calorimetry

Electrochemical calorimetry is a technique to integrate the isothermal heat flow calorimetry of LIBs with electrochemical cycling. The thermal spectrum will show an identifiable characteristic once the lithium plating occurs, which allows qualitative detection of lithium plating by electrochemical calorimetry.

Downie et al. [94] used this method to in situ detect the onset of lithium plating in a full and half a cell with different types of graphite. They found that a rounded feature would appear in the thermal spectrum when lithium plating happened, and this feature was observed in both full and half cells with a G25 or MCMB graphite under plating conditions. And the detection results were verified by CE and the presence of plateaus in voltage profiles.

Although electrochemical calorimetry is simple, and wide-applicable, its limitations include:

(1) Application: i) only lithium plating at the serious stage can be detected; ii) this method is not suitable for implementation in BMS; and iii) the detection time is relatively long due to the need of low currents under quasi-adiabatic conditions.

(2) Accuracy: the detection accuracy is yet unknown.

4. Physical methods

Physical characteristics can also be used for lithium plating detection. Lithium plating and the corresponding formation of a plated layer on the anode lead to the change of morphology and microstructure of electrodes. In-situ physical methods can not only detect the growth of plated lithium but also obtain the distribution of plated lithium at different positions.

Physical methods include cell thickness measurement, acoustic detection, and H_2 gas detection, which usually require auxiliary instruments. The features of these physical methods are summarized in Table 4. Although they may not be suitable for on-board applications, physical methods are of importance to understand the principles of lithium plating and investigate the crucial charging rates without lithium plating for charging strategy optimization of the commercial cells.

4.1. Cell thickness measurement

Measuring the cell thickness is a common method to detect lithium plating in pouch LIBs. It is well known that the intercalation and de-intercalation of lithium ions are associated with the significant volume changes of electrodes [95], and consequently with the thickness changes of pouch cells. When cycling under moderate conditions, the change of cell thickness is reversible and will not increase permanently [95]. Furthermore, the thickness change is mainly contributed by the anode [96]. NMC expands about 1% [97] and LiCoO$_2$ (LCO) expands 1.8%

Table 4
An overview of physical methods.

Method	Procedure	Applicability	Output
Cell thickness measurement	Plating can be detected during charging by measuring thickness.	It can detect the moderate stage.	Qualitative results
Acoustic detection	Plating can be detected after long-term cycles by measuring acoustic signal.	It can detect the serious stage.	Quantitative results
H_2 gas detection	Plating can be detected during charging by detecting H_2 gas.	It can detect the serious stage.	Quantitative results

[98], compared with 10% expansion of Graphite [99] during cycling. Therefore, once lithium plating occurs, the metallic lithium film and the resulting SEI formation on the anode will lead to additional growth and irreversible change of cell thickness. Fig. 10 shows the change of the cell thickness for one charging process. Compared with the original thickness, the thickness of plating-induced cells will reach a peak and then decrease at the constant voltage phase due to chemical intercalation, but there is still a permanent increase caused by the irreversible lithium plating. Bitzer et al. [100] concluded that only lithium plating will cause the thickness change in a single cycle because the influence of gas evolution and thermal expansion on the cell thickness can be neglected.

Technologies for cell thickness measurement cover from single-point measurement to 3-D scanning measurement. Dial indicators [100,101], or displacement sensors [102] are usually applied in the single-point measurement. The reversible and irreversible lithium plating could be identified by comparing the change of thickness with and without plating [100]. To detect small quantities of lithium plating, a high-resolution sensor is necessary, which can be used to determine the maximum permissible charge rates without lithium plating in relation to SOC, temperature and charge quantity [101], and optimize charging strategies [102].

However, only one measured point on the cell is not enough, because the occurrence of lithium plating is usually random and the distribution is inhomogeneous. To avoid the influence of measuring position on detection, 3-D scanning method, which can monitor the thickness of the entire cell surface, is needed. With the help of laser sensors [103], a 3-D scanning method also can analyze the distribution of lithium plating. Rieger et al. [104] applied a 3-D scanning measurement to investigate the local lithium plating during fast charging.

Cell thickness measurement can detect lithium plating in situ and qualitatively, and has the potential to optimize charging protocols for EVs. But its limitations exist in:

(1) Application: i) it can only be used for pouch cells due to their pressure sensitivity; ii) this method is only suitable for laboratory applications due to its requirement of stable environments and high precision equipment.
(2) Accuracy: i) a long relaxation time is necessary to obtain a stable cell thickness and avoid the error caused by the thermal expansion or other factors; ii) there exists a detection threshold, because a small amount of plated lithium cannot cause a significant change of cell thickness; iii) the measuring position is very important because of the inhomogeneous distribution of plated lithium, for instance, plating tends to occur near the cell edge [103] or the tabs [104].

4.2. Acoustic detection

Acoustic detection, also known as ultrasound acoustic time-of-flight (TOF) detection, is a spatially and temporally resolved technique for identifying lithium plating. Acoustic detection analyzes the reflect
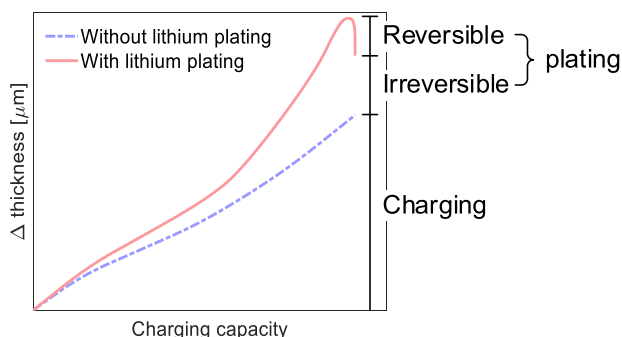

Fig. 10. The evolution of cell thickness at a single charge step [100].

signals emitted from a point source on the measured material interfaces [105]. Due to the regular and layered internal configuration of LIBs [106], this method can in-situ monitor the changes of anode surface affected by lithium plating during cycling.

Acoustic detection is a semi-quantitative method for lithium plating, as the more plated lithium, the more hysteresis of the acoustic TOF [107]. Considering the sensitivity of the acoustic signal to gassing and temperature, Bommier et al. [107] proposed the TOF endpoint difference to indicate the appearance of lithium plating. They found that, contrary to the non-plating conditions, the TOF endpoint maintained at a position higher than the initial position when plating appears, regardless of the resting time. However, two disadvantages exist:

(1) Application: acoustic detection can only be used for pouch cells.
(2) Accuracy: the acoustic signal is sensitive to gassing [108], which also occurs in harsh conditions; and therefore gassing can lead to a high uncertainty in the detection of lithium plating.

4.3. H_2 gas detection

Gas detection has been widely applied for the safety warning of LIBs. When lithium plating occurs on the surface of anodes, the plated Li metal can react with electrode polymer binders and generate H_2, which can be detected by H_2 gas sensors [109]. Therefore, detecting H_2 gas during charging can monitor lithium plating in situ and in real time.

Recently, Jin et al. [109] proposed an H_2 gas detection method for lithium plating detection. They concluded that this method was suitable for different electrode polymer binders, including commonly used as polyvinylidene difluoride (PVDF), styrene-butadiene rubber (SBR), and carboxymethylcellulose (CMC). Moreover, overcharge experiment results shown that this method was suitable for commercial LIB packs.

H_2 gas detection is an in-situ, non-intrusive, and real-time method for detecting lithium plating. However:

(1) Application: this method only works when H_2 leaks, so this method may not be suitable for cylindrical batteries, because the shell of cylindrical batteries is hard and the battery is difficult to rupture, making it difficult for H_2 to leak.
(2) Accuracy: detection accuracy depends heavily on the sensitivity of sensors.

5. Comparison and prospects

5.1. Comparison

Fig. 11 shows an overview of the advantages and disadvantages of each detection method, and six crucial features are included: BMS implementation, environmental adaptability, real-time detection, quantitative detection, fast detection, need of additional instruments, and application scope (i.e. applicable battery chemistries), all of which are important factors for on-board detection of lithium plating. Radar diagrams are used to visually compare the developed methods. The corresponding scores represent the degree how a feature is met: 1 – fully met, 0.5 - met under certain restricted conditions, and 0 - not met.

BMS implementation means if one detection method can be embedded into BMS. If feasible, lithium plating can be detected online during charging, especially during fast charging and low-temperature charging; therefore, the charging strategies can be regulated by BMS accordingly to suppress plating, and consequently, the charging safety can be improved. Most electrochemical methods are suitable for BMS implementation due to the simple input signals and uncomplicated signal processing algorithms.

Environmental applicability is the adaptability of one detection method to the actual driving environment (not only for laboratory uses). Most electrochemical methods do not require a stable operating condition. However, the electrochemical calorimetry method needs an
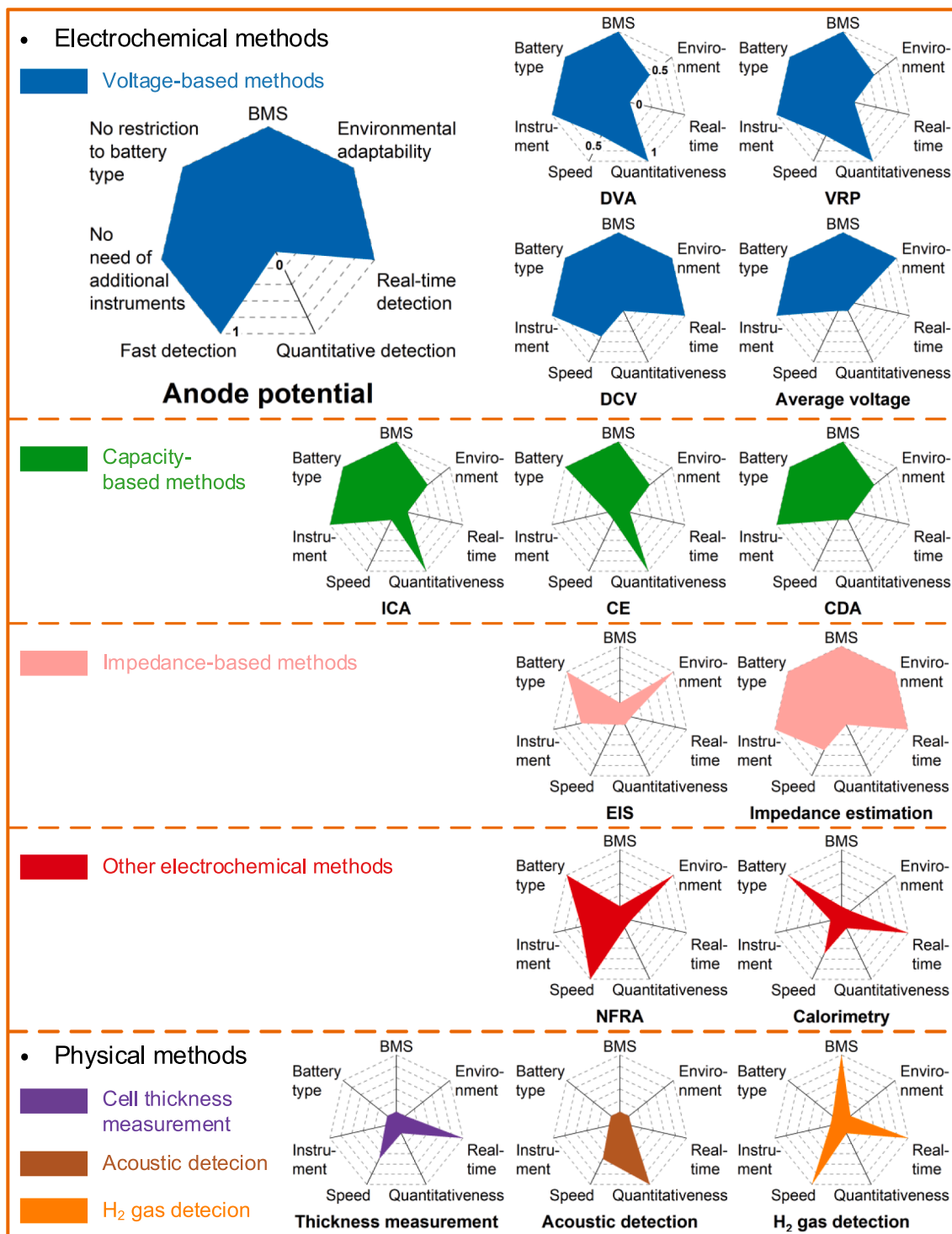
Fig. 11. Overview of the two types of lithium plating detection methods by radar diagrams. Each axis of the radar diagram indicates a feature of this method, including BMS implementation, environmental applicability, real-time detection, quantitative detection, fast detection, no need for additional instruments, and no restriction to battery type, as described in the first radar diagram. The corresponding scores represent the degree how a feature is met: 1 – fully met, 0.5 - met under certain restricted conditions, and 0 - not met. The following diagrams have the same meanings as the first one so only brief descriptions of each axis are added.

isothermal condition. In addition, all physical methods can only be applied under a stable environment without vibration and interference, which seriously limits their potential for on-board applications. DVA and capacity-based methods require EV to stand still and keep the charger connected when detecting lithium plating for the acquisition of stable current and smooth voltage curves. VRP requires the vehicle to rest for a while after charging.

Real-time detection means that lithium plating can be detected immediately when it appears. It is crucial to control the charging process to restrain lithium plating in real time. However, most methods are not capable of doing this and can only detect lithium plating when the charge is complete. Only the methods based on anode potential measurement, cell impedance estimation, electrochemical calorimetry, cell thickness measurement, and H_2 gas detection enable real-time

detection.

Quantitative detection is beneficial to accurate control of lithium plating. It can make a trade-off between the charging speed and lithium plating, which can realize an increased charging speed by scarifying a small amount of lithium plating in the initial or moderate stage. The quantitative detection of plated lithium can also assist the estimate of the aging status of LIBs. Currently, only DVA, VRP, ICA, CE, and acoustic detection enable quantitative detection.

Fast detection refers to the detection time. A systematic comparison of the detection time is shown in Table 5. In this work, Score 1 means the detection time is less than 1 h, Score 0.5 means the detection time is several hours; and Score 0 means it takes long-term cycles. Most methods, such as DVA, VRP, ICA, CE, EIS, electrochemical calorimetry, and most physical methods, require an equilibrium state of cells, therefore a sufficiently small discharging rate or a sufficiently long rest time is needed. Methods based on average voltage inspection, CDA, and acoustic detection even need long-term cycling to detect lithium plating. By now, only the methods based on anode potential measurement, NFRA, and H_2 gas detection can achieve fast detection less than 1 h.

The application of some methods needs additional instruments. For example, CE needs a high-precision battery test system [73], and electrochemical calorimetry uses an isothermal heat flow calorimetry [94]. Moreover, the cell thickness measurement requires a high-resolution dial indicator to measure the subtle variation of cell thickness [101]; acoustic detection requires expensive auxiliary equipment including a pulser, a receiver, and several transducers to produce, transmit and receive acoustic signal; and H_2 gas detection needs H_2 gas sensors. EIS and NFRA require the additional functions of the charger for sinusoidal current or voltage signal input.

The application scope of detection methods may also be constrained to specific types of cells. In general, electrochemical methods show great flexibility to all battery chemistries with different geometries, while cell thickness measurement and acoustic detection are only suitable for pouch cells due to their high-pressure sensitivity and simple internal structure. H_2 gas detection is not suitable for cylindrical cells due to their hard shells.

Furthermore, methods that can be used to detect plating at initial and moderate stages are highly recommended. In general, detection at initial and moderate stages is more significant for early-warning of lithium plating and enables a timely inhibition to avoid the deterioration of battery capacity and safety caused by lithium plating. Most electrochemical methods can detect lithium plating at as early as the moderate stage and are also easy to implement.

Based on the above comparison, all electrochemical methods except electrochemical calorimetry are highly recommended for on-board applications, but the detection threshold and time of electrochemical methods should be considered. Here, the detection threshold means the minimum amount of plated lithium that can be detected. Physical methods can analyze lithium plating intuitively such as the distributions of lithium plating and are more recommended for the laboratory application to study the mechanism of lithium plating because a stable environment and additional instruments are needed.

5.2. Prospect of future technology development

Some detection methods require complex algorithms to perform accurate calculations, and a BMS with more computing power to handle these complex algorithms has already become the trend. To comply with the development trend of the BMS and EVs, suggestions for future works are presented in Table 6 on lithium plating.

For electrochemical methods, the mechanisms behind the relationship between the operating conditions and the electrochemical reactions in LIBs need to be revealed. Currently, the impacts of operating conditions such as charging/discharging rates, ambient temperature, and rest time on the electrochemical signals are not clear. Especially for methods that detect lithium plating after charging, even though the charging process is exactly the same (i.e. the situation of lithium plating is identical), different operating conditions after charging will lead to obvious differences in detection results. Hence, it is essential to quantify these impacts. Based on the quantified impacts, accurate results under different conditions can be obtained.

In addition, although advanced algorithms, like artificial neural networks, have already been employed by some electrochemical methods such as ICA for estimation of the state of health (SOH) and remaining useful lifetime (RUL) [110], they have not been used for lithium plating detection. Therefore, using artificial intelligence algorithms to analyze the evolution of lithium plating with changes of environments and charging strategies represents another promising research topic, which can be used for on-board applications [111] and has good applicability for different conditions [112,113]. Furthermore, in order to handle a large amount of calculations, cloud BMS is emerging as an innovative solution [115]. The powerful data processing

Table 5
An overview of the required detection time.

Level	Method and required time	Experimental	Ref.
less than 1 h	Anode potential measurementIn real time	15 ℃ ~ 45 ℃ Charge at 0.2 C ~ 3 C	[29]
	NFRA About 15 min	0 ℃ Charge at 1 C	[91]
	H_2 gas detection About 0.5 h ~ 1 h	Room temperature Overcharge at 0.5C	[109]
1 ~ several hours	DCV About 1 h ~ 2 h	25 ℃ Charge at 1 C ~ 2 C	[59]
	VRP About 1.5 h	−15 ℃ Charge at 1 C ~ 3 C Relaxation	[57]
	Cell impedance estimation About 2 h	20 ℃ ~ 30 ℃ Charge at 1 C ~ 3 C	[86]
	Cell thickness measurement About 5 h ~ 10 h	−5 ℃ Charge at 0.7 C	[100]
	DVA About 20 h	−20 ℃ ~ −26 ℃ Charge at 1 C Discharge at C/20	[28]
	Acoustic detection About 35 h	5 ℃ Charge at C/10, 1 C	[107]
	Electrochemical calorimetry About 40 h	40 ℃ Charge at C/10	[94]
long-term cycles	EIS About 120 cycles	−22 ℃ Charge at 1 C	[82]
	CE About 600 h	12 ℃ ~ 60 ℃ Charge at C/50 ~ 5 C	[73]
	ICA About 900 cycles	23 ℃ Dynamic stress test	[64]
	Average voltage inspection About 1200 cycles	23 ℃ Charge at C/3	[73]
	CDA About 150–300 days	40 ℃ Charge at 1 C ~ 8 C	[60]

Table 6
Potential improvements in the lithium plating detection methods.

Method	suggestions for future development
Electrochemical method	1. To consider the impact of rates and operating conditions on the electrochemical signals; 2. To quantify the impacts of operating conditions after charging on the detection results; 3. To introduce artificial intelligence algorithm for signal processing; 4. To utilize cloud BMS to achieve the fusion of different detection methods.
Physical method	1. To consider the impact of the aging degree on the change of physical signal; 2. To develop advanced scanning technologies that are simple and fast; 3. To be embedded in BMS for on-board applications.

capabilities of cloud BMS make it possible to process different electro-chemical signals and apply different electrochemical methods simultaneously to combine the advantages.

For physical methods, it is necessary to study the influence of cell aging on the detection of lithium plating, because aging has a great influence on the properties of battery materials, thus, affects the physical signals. In addition, physical methods highly depend on the selection of measuring positions. Inhomogeneous distributions of lithium plating lead to clear differences in the results at different positions. Developing simple and quick scanning technologies is essential to apply physical methods for on-board applications.

6. Conclusions

In this paper, the state-of-the-art about in-situ detection methods of lithium plating for on-board applications are reviewed systemically, which provides researchers and engineers guidelines about how to choose a method based on their requirements and application environments, as well as insights on developing innovative methods. The present methods are categorized into two categories, including electrochemical methods and physical methods. The development of lithium plating is divided into 3 stages based on its reversibility: initial stage, moderate stage, and serious stage. The pros and cons of each method have been analyzed and its application for different plating stagnates is also discussed. Through a comprehensive comparison of these methods, it is concluded that electrochemical methods are more suitable for on-board detection, and physical methods are more recommended for the laboratory applications. For future development of novel detection methods, the impacts of operating conditions and aging degree on the detection results are suggested to be studied.

Declaration of Competing Interest

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

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