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Can Electrochemical Impedance Spectroscopy be Replaced by Direct Current Techniques in Battery Diagnosis?

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Electrochemical impedance spectroscopy (EIS), a conventional and alternating-current-(AC)-based technique for impedance measurement, is commonly used in battery diagnosis. However, it requires expensive equipment and demanding operating conditions and is complex and model-dependent in data analysis. Recently, novel direct current (DC) analytics have emerged as an alternative to EIS. They are simple yet powerful, being capable of revealing impedance information that traditionally could only be obtained through EIS and determining Li-ion diffusion coefficient. Besides, a complete EIS spectrum can be predicted based on constant current charging curves in the support of machine learning methods. This work highlights the similarities and discrepancies between DC techniques and EIS in the electrochemical analysis of Li-ion batteries. Looking ahead, DC techniques may be a promising substitute for EIS in future battery diagnosis, requiring simplified equipment while offering a deep understanding of battery impedance and its underlying electrochemical processes.

1. Introduction

With the ever-escalating demand for high-performance batteries for sustainable mobility and energy storage, it is imperative to gain a comprehensive and in-depth understanding of their electrochemical behaviors to accelerate their development and optimal use.[1] Battery impedance is a pivotal parameter that determines battery output voltage,[2] heat generation,[3] and overall health status, and therefore demands significant attention in the design and operation of the battery system.[4]

Battery impedance measurement methods can be broadly categorized into direct current (DC) and alternating current (AC) methods based on the approach used to apply the current, as shown in Figure 1.[5] The DC methods comprise DC resistance (DCR)[6] and hybrid pulse power characterization (HPPC).[7] The DCR and HPPC methods differ primarily in the duration of their charge and discharge currents, with the former generally having shorter durations than the latter, which typically is 18 seconds for DCR. Besides, for the impedance of Li-ion diffusion measurements, galvanostatic intermittent titration technique (GITT)[8] and intermittent current interruption (ICI)[9] can be employed. On the other hand, AC testing primarily pertains to EIS, which can rapidly measure all the aforementioned impedances. Here, we are using Li-ion batteries (LIBs) as an example to discuss the functions of DC methods vs. EIS, emphasizing that this discussion is not limited to LIBs.

LIBs are nowadays the dominant battery technology on the market.[10] They have been extensively studied since the 1980s and the principal electrochemical processes during battery charging and discharging have been identified.[11] Nevertheless, specific mechanisms, such as the impact of operating conditions on the aging of battery components, have not yet been

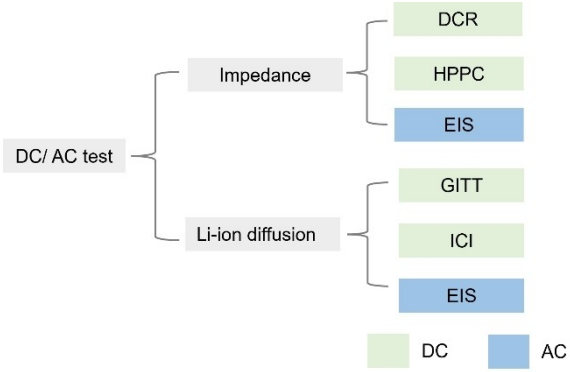

Figure 1. Common DC and AC methods for battery impedance and Li-ion diffusion measurements.

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fully understood.[12] EIS is a powerful technique capable of providing insights and resolving the unanswered questions related to battery research.[13] The main function of AC-based EIS in batteries is to determine the electronic and/or ionic conductivity of electrodes, electrolytes, interphases and interfaces, and provide information on the reactance (e.g., capacitance).[14] Besides, EIS proves to be exceptionally fast for measurement of the mass transfer (e.g., Li-ion diffusion) coefficient in solid electrodes of batteries.[15] Furthermore, to evaluate Li-ion diffusion resistance, the traditional GITT of DC method is much more time-consuming than EIS.[16] However, because EIS is highly sensitive and easily influenced by external factors, it requires precise equipment and can be challenging to use in various conditions.[17] This sensitivity can lead to significant errors in real-world situations. Although dynamic or operando EIS testing does not require the battery to be in equilibrium state, EIS testing under dynamic conditions is technically demanding in terms of control and the accuracy of the results is limited, compared with the conventional EIS.[18–20] Despite the research efforts in the past years, dynamic EIS is currently limited to the laboratory use and still far from real-life application for battery diagnosis. In contrast, the DC impedance measurement technique is easier to implement in practical applications, which do not need long time for stable.[21] Although it may not provide the same level of precision as EIS testing, it can quickly conduct many times in a short period, resulting in consistent and reasonably stable results. This is because the DC charging and discharging processes can be executed during battery operation, and thus allows for the realization of the DC impedance measurement while the battery is in use, making *in-situ* or *operando* impedance measurement feasible. Obviously, obtaining DC impedance test data is easier

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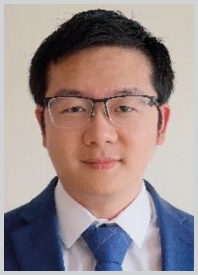

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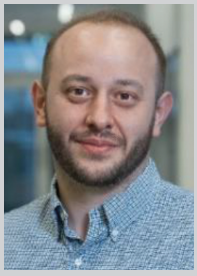

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compared to acquiring EIS data. However, DC impedance measurement cannot directly deconvolute electrochemical processes, hindering mechanistic understanding of the detailed electrochemical processes inside batteries. Achieving a wide range of electrochemical analysis results from DC test data poses a challenge for battery diagnosis.

With the development of theoretical computations, DC impedance measurement techniques have been continuously advanced, and can achieve some functions of EIS measurement. For example, the results of DC impedance measurement also have the potential to differentiate electrochemical processes inside batteries.[14] Through data training, the EIS impedance spectrum (typically obtained with AC measurement methods) can be simulated from the constant current charging curves with the help of machine learning.[22,23] Furthermore, conventional GITT method for measurement the impedance in Li-ion diffusion is highly time-consuming, while the novel ICI technology saves at least 75% of the measurement time.[1] As DC measurement continues to advance, it is anticipated that it may eventually be able to achieve all functions of EIS.

In this concept paper, we discuss the potential of DC impedance measurement for obtaining similar battery electrochemical understanding as we get from EIS measurement. Additionally, we explore the potential advancements and prospects of impedance measurement technology from an application perspective.

2. Origins of Impedance in Battery

Battery impedance is not only influenced by battery state of charge (SOCs) but also by the state of health (SOH) and operating temperature.[21] As discussed, battery impedance is rooted in multiple processes, such as the electrode surface film formation and growth, charge transfer, Li-ion diffusion inside electrode materials, and Ohmic impedance. These specific processes contributing to battery impedance and the battery aging mechanisms are shown in Figure 2.[24,25] As the battery impedance can be divided into ohmic impedance (related to conductivity loss, electrolyte consumption, and electrolyte decomposition),[21] surface film impedance (related to loss of Li, involving CEI on the cathode surface, i.e., R_{cei}, and SEI on the anode surface, i.e., R_{sei}),[26] charge transfer impedance (R_{ct}, related to charge storage in double layer, and charge transfer reaction),[13,27] and Li-ion diffusion impedance (R_{mt}, also related to loss of active materials, e.g., particles fracture and materials structure change)[28] during the measurement, the aging modes of the battery can be easily identified. This will allow for accurate and effective battery diagnosis. Therefore, EIS is often used to identify the battery aging mechanism, and to unravel the cause of battery aging. For instance, the loss of electrode material and loss of Li can be identified from the R_{ct} and R_{sei} features of EIS spectra.

EIS measurements are typically conducted at a specific state of charge (SOC) because the distribution of Li-ions in electrode materials can affect Li-ions diffusion and charge transfer resistance. In addition, as the SOC increases, the volume of the negative electrode (Graphite) also increases, resulting in a reduction in battery impedance.[29] Furthermore, temperature will also have an impact on battery impedance, such as at low temperature, the viscosity of the electrolyte is high, making it difficult for Li ions to diffuse, which will also increase the battery impedance.[30] Meanwhile, the impedance is related to and can be used to estimate battery SOH.

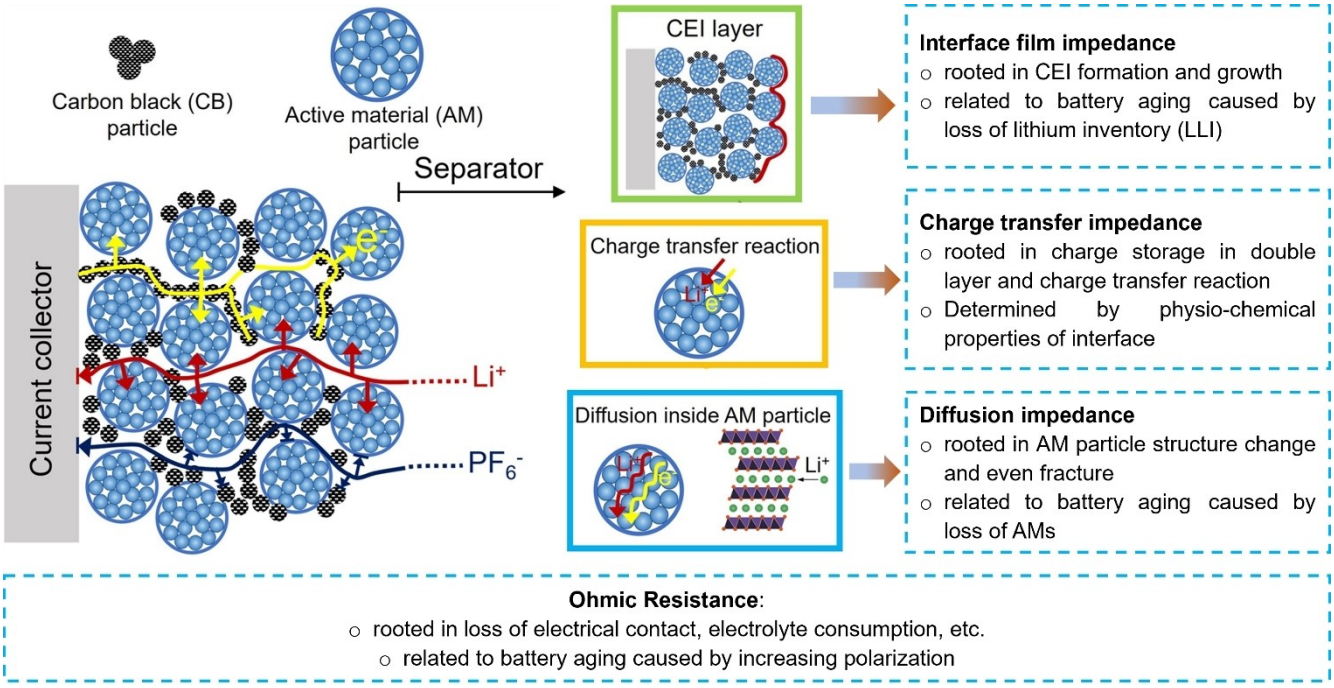

Figure 2. The electrochemical processes inside LIBs, taking cathode as an example, and the impedance contributions during battery operation and aging.[24,25]

Currently, the diffusion coefficient of Li-ions in solid electrodes can be measured using GITT, an intermittent constant current technique, next to EIS measurement. However, in the analysis of impedance contributions other than Li-ion diffusion that are related to electrochemical processes, EIS remains the preferred method that allows for the deconvolution and detailed analysis of various electrochemical processes that contribute to battery impedance.

3. EIS and DC Impedance Show Similar Results in Impedance Identification

Both EIS and DC impedance measurements can be used to identify different battery electrochemical processes, such as R_{ohm}, R_{sei}, R_{ct}, and R_{mt} (Figure 3).[14] The EIS results are obtained based on the changing of current frequency. With the frequency changing, the transfer (Li-ions move and diffusion) process can be identified from the EIS spectrum, as shown in Figure 3**a**. In the high-frequency region, the primary occurrence is the resistance (R_{ohm} and R_{sei}) of electron conductive and Li-ions as they traverse the interface film. In the medium frequency region, the impedance is from the charge transfer process. In the low frequency region, the impedance is mainly from the Li^+ diffusion in electrode. Similarly, DC constant current also can be used to measure battery impedance. Depending on the duration of current applied, the battery impedance originating from different electrochemical processes can be calculated, as shown in Figure 3**b**. In the first few to tens of milliseconds, the electron conductive occurs, followed Li-ions go through the SEI film. The charge transfer process can be finished within the following few seconds. In the next ten seconds, the Li-ions diffusion process is finished. This means that both EIS and DC techniques can obtain battery impedance results, which are dependent on the rate of the chemical reaction process. Stolz et al.[31] summarized the mechanisms of EIS impedance and DC impedance, respectively. They highlighted the advantages of DC measurement techniques. Consistent with Stroe's study, the advantage of the DC pulse technique is that one can extract the impedance/resistance directly from the battery's real-life operation.[32]

Both EIS and DC measurements rely on discerning the reaction rates of diverse electrochemical processes to deconvolute these processes. In theory, DC testing can serve as a replacement for EIS. However, some technical barriers need to be addressed.

First, how to distinguish the timescales for R_{mt}, R_{ct} and R_{sei} in DC impedance measurement is still a challenge. This is expected to be solved with the developments in computational methods, such as employing advanced data fitting or prediction methods to identify the timescales of individual reaction processes. Besides, the battery's SOC and electrochemical status also change constantly during charging process. As shown in Figure 3**c**, the polarization of the battery is related to the battery's SOC. The SOC changes during the charging or discharging process, leading to inaccurate test results. Therefore, in DC impedance testing, DCR and HPPC testing, the test time needs to be limited. For the identification of electrochemical processes by DC curve, it is necessary to consider the gradual changes in this stoichiometry. Furthermore, DC impedance results are additionally influenced by the magnitude of the applied current, as shown in Figure 3**d**. The non-linear fluctuations in voltage and current within the battery can lead

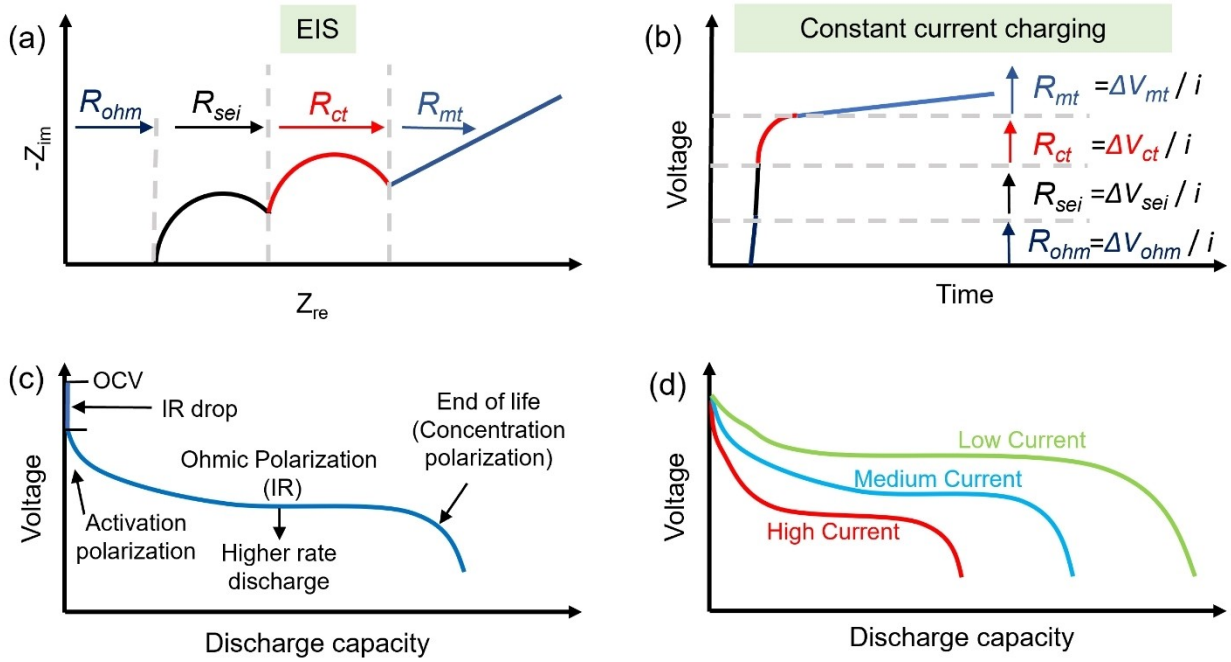

Figure 3. (a) EIS and (b) DC techniques, *e.g.* constant current charging, for identifying and quantifying battery impedance (including R_{ohm}, R_{sei}, R_{ct}, and R_{mt}, modified based on Ref. [14]). (c) Battery polarization in battery discharging process[33]; (d) Discharge voltage curves at different current rates.[34]

to corresponding variations in the impedance results. Usually, DC impedance testing requires a lower current of 0.5 C or 1 C, or even lower, to reduce the polarization effect caused by the current magnitude. These challenges represent significant hurdles in the advancement of DC impedance technology. If these issues are resolved, there is no doubt that DC impedance testing technology has the potential to supplement EIS technology.

4. Determination of Li Diffusion Impedance in Solid Electrodes with EIS and DC Techniques

As shown in Figure 4, both EIS and DC techniques can be used to measure the Li diffusion coefficient.

EIS is the most convenient method for measuring Li-ion diffusion as it takes as short as 5–30 min to measure the EIS of LIBs, which can be used to determine Li-ion diffusion coefficient in the electrodes. The low frequency range of the EIS spectrum and the Nyquist plot can be used to calculate the Li-ion diffusion coefficient.[35]

GITT is a conventional and primary method for determining Li^+ diffusion coefficients for insertion electrode materials. GITT involves two consecutive steps: firstly, applying a constant current for a duration where the assumption of semi-infinite diffusion is valid, and secondly, interrupting the current until the voltage stabilizes to indicate equilibrium.[36] By analyzing the electrode potential during the current pulse and the subsequent change in equilibrium potential, GITT allows for the determination of the chemical diffusion coefficient of the charge-carrying ions.[1] The disadvantage is that GITT is time-consuming, typically between hours to days, as it requires a significant time interval (over 60 min) between current pulses.[37]

Recently, a novel ICI method has been proposed as an alternative to the GITT measurement.[1] By conducting linear regressions of the potential change against the square root of step time during the current pauses, it becomes possible to extract both the time-independent and time-dependent components of the resistance. These components are referred to as the internal resistance and the diffusion coefficient, respectively. The ICI method shortens the relaxation interval to 1–10 seconds, significantly reducing the measurement time. The innovative ICI method can quickly determine the diffusion

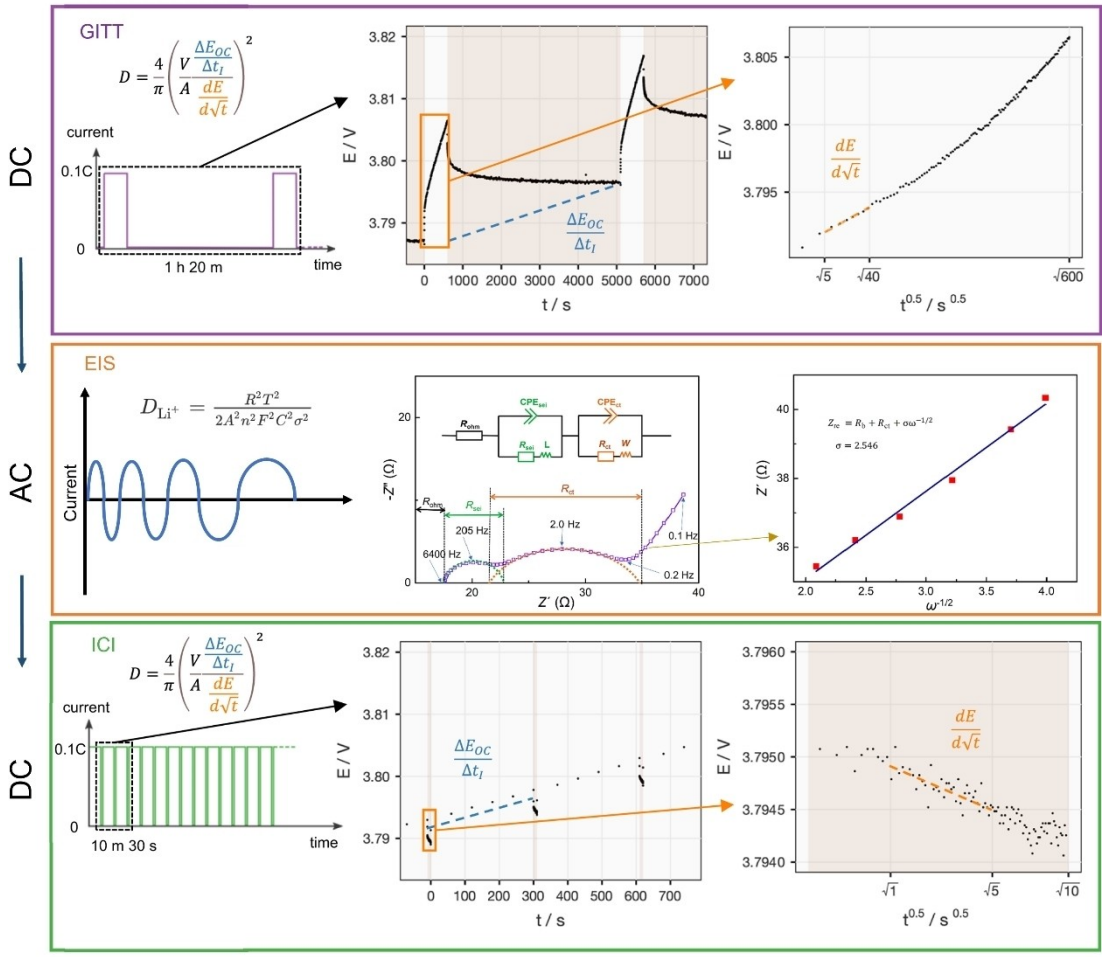

Figure 4. Methods for measuring Li^+ diffusion coefficient in insertion electrode materials. (a) EIS[35]: the slope of the low-frequency region can be used to calculate Li-ion diffusion coefficient; (b) ICI: the relaxation time is short (only a few seconds); and (c) GITT: the relaxation time is long (over 60 min).[1]

coefficient of Li-ions. It is also easier to apply in real-world application. With a deep understanding of Li-ion diffusion process in electrodes and advancement of data processing, a more simplified DC technique determining Li diffusion may be envisaged.

5. EIS Spectrum Prediction Based on Constant Current Charging Curves

As mentioned before, conducting EIS tests in real vehicles poses significant challenges. If the complete EIS can be approximated through DC testing, it would provide essential information required for subsequent battery management system.[38] This information encompasses parameters such as ohmic impedance, charge transfer impedance, interface film resistance, and lithium diffusion coefficient.[39] Using some of this data as input, models can be developed to accurately diagnose battery health,[40,41] even predict its future performance.[42] The first thing to note is the predictability of the constant current charging curve and EIS.

With the development of machine learning, it has been found that the constant current charging curve can be used to predict the complete EIS spectra with marginal error.[22,23] The prediction process is shown in Figure 5. First, the model undergoes training with an extensive dataset, which includes voltage-capacity curves collected at different SOH levels, together with the corresponding EIS data. For the dataset used by Guo et al.,[22] they trained the model using 300 EIS datasets. They then validated the model using 75 EIS datasets. Another example, Duan et al.[43] utilized 1200 pairs of EIS datasets and battery voltage curves for training the model. They reserved 300 pairs for model validation. Then, the established model is used to predict the complete impedance spectrum. Finally, the machine learning model establishes a mapping between the battery charging curve and the corresponding all EIS data points. Because EIS is tested at certain fixed frequency points from 6500 Hz to 0.01 Hz, the prediction results can correspond to the test frequency one-to-one, ensuring the valuable of the prediction results. In addition, the step time recording interval of constant current charging is 10 s, and changing the time interval has little impact on the prediction accuracy.

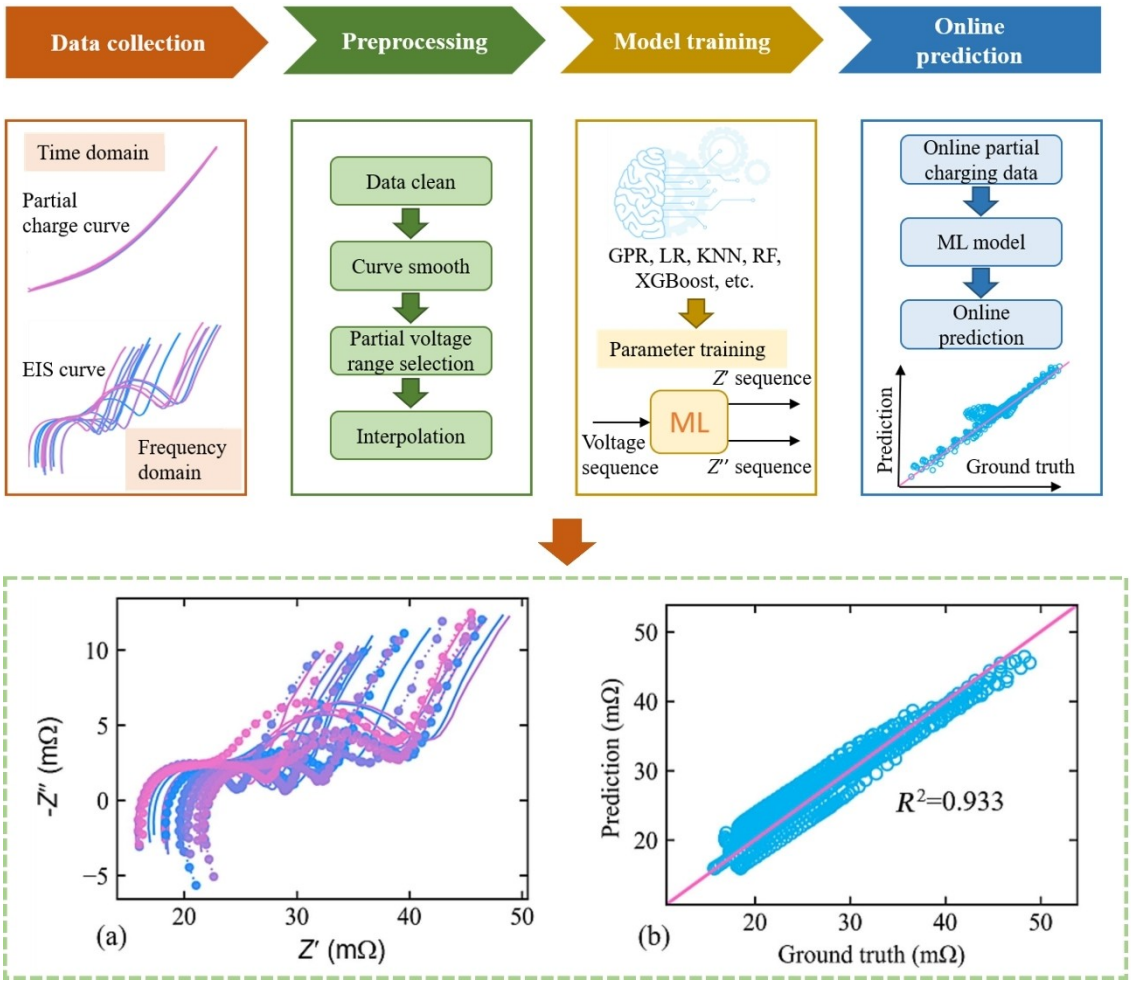

Figure 5. Prediction of EIS results based on battery constant current charging curve with machine learning methods. The algorithm training process was carried out using dataset collected at early stage.[22]

This demonstrates that there may exist a discernible correlation between DC charging curve and EIS data. However, the machine learning process is like a black box,[44] and the intricate relationship between battery charge curves and EIS remains elusive. It is worth noting that, as illustrated in Figure 3**c** previously, the charge and discharge curve of the battery inherently encompasses polarization and impedance data within the battery. These data points might exhibit some numerical correlations with the EIS results. Furthermore, both EIS and the battery charge-discharge curves can be employed to quantify the battery's electrode losses and impedance increments, thereby further confirm the connection between them.[45,46] Yet, we do not exclude the possibility that this relationship is just a simple mathematical correlation between battery charging curves and EIS spectra.

This prediction method, which relies on model training, has the potential to accelerate the battery SOH diagnosis in vehicles applications.[47] The EIS information of the battery can be quickly obtained according to the charging curve of the battery.

Here, the implementation of DC measurement method demonstrates the potential as a substitute for EIS in application. The development of machine learning will extend the use of DC charging curve as replacement for EIS to study and diagnose the battery electrochemical behavior.

6. Perspective for the Development of Impedance Identification

6.1. Understanding the Underlying Mechanisms of Battery Impedance

Understanding of the underlying mechanisms of impedance is continuously evolving. Currently, it is broadly understood to be associated with specific chemical reaction processes which can be identified from the EIS measurement. In particular, the resistance of the Li-ion diffusion process is mainly related to the phase transition of the electrode material.[48] To further understand the Li diffusion resistance, it is necessary to clarify how the structural change of the electrode material affects the diffusion process of Li-ions in the electrode material. This requires intensive material structure studies combined with electrochemical measurement analysis. The transformation process of the material structure corresponds to the change in the diffusion coefficient of Li-ions.[49] In this way, the relationship between material structure and diffusion resistance can be established accurately.

The charge transfer is a slower process than the Li-ion transfer across the interfacial film, but faster than Li-ion diffusion in the solid phase.[50,51] The Li-ion desolvation and solvation processes involved in the charge transfer process remains unclear. The microscopic process of charge transfer is still inferred. Especially under different measurement techniques. It is necessary to adopt advanced measurement methods such as element labeling to track and clarify the charge transfer process. Then, the electrochemical process inside battery will be subdivided again to achieve a more precise diagnosis.

For the surface film impedance, the components and formation of the EEI (SEI and CEI) film are still unclear. Highly ordered pyrolytic graphite with a smooth surface is usually used to study the growth process and composition of SEI films formed in LIBs.[52] Cryogenic electron microscopy (cryo-EM) is a powerful tool to study in detail the structure and chemistry of SEI films, which helps to preserve the native state of sensitive SEI films and greatly suppress electron damage of traditional transmission electron microscopy (TEM).[53] X-ray photoelectron spectroscopy (XPS) and depth profile analysis is also useful for analyzing the composition of the SEI films.[54]

The study of the underlying impedance mechanisms relies on advanced detection equipment and powerful electrochemical analysis techniques. Clear microscopic descriptions of the mechanism will enhance the detail of the electrochemical process, leading to highly accurate models and reliable impedance calculations.

6.2. Development of Impedance Identification Methods

Time-consuming GITT measurements and DC impedance measurements are still widely used. In principle, the DC impedance measurement is similar to the EIS test, and various electrochemical processes are distinguished according to the duration of the current application. Therefore, DC impedance measurement is expected to replace EIS. However, it is difficult to distinguish the time constants of different chemical processes during DC impedance measurement. As shown in Figure 6, a large amount of measurement data can be used to calibrate

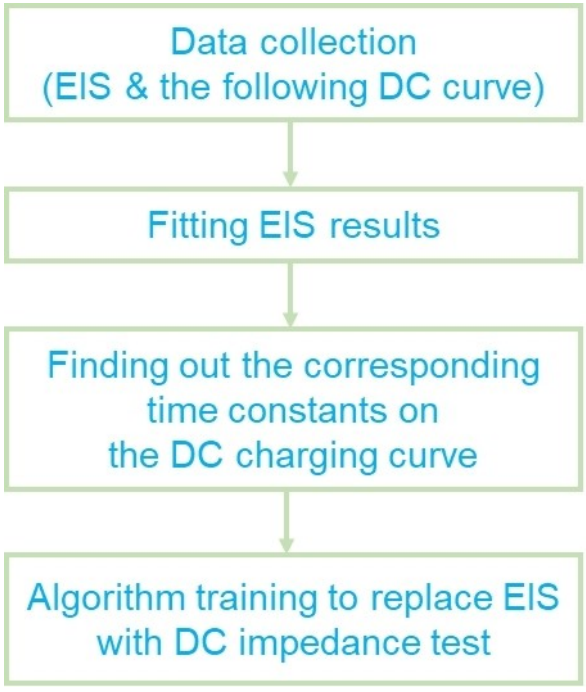

Figure 6. The algorithm training process for calculating DC impedance.

and establish an accurate function, which will be beneficial to the development of DC impedance technology. In addition, the advanced DC test algorithm also consider the impact of SOC change and the magnitude of the applied current during DC testing.

The test method of Li-ion diffusion coefficient GITT has been simplified, and the ICI technology with fast measurement is to establish a functional relationship under the unsteady state of the battery. With the development of electrochemical models and computing science, it is promising to find a more universal and rapid impedance measurement method.

6.3. Development of Impedance Identification in the Application

The battery's charging voltage curve can serve as a basis for predicting complete impedance spectra. While training the predictive model initially demands a substantial dataset, it ultimately enables accurate real-time impedance estimations during battery charging in practical scenarios. As a result, obtained EIS electrochemical diagnosis can furnish the battery management system with additional insights into battery health status. This enhanced feedback of battery information is instrumental in advancing intelligent battery management systems.

Furthermore, digitizing the potential connection between the charging curve and EIS could simplify the direct acquisition of battery impedance information through alternative means e.g., partial charging curve.

Simplifying DC impedance measurement as an alternative to EIS measurement or employing machine learning algorithms to estimate battery impedance from extensive charging datasets holds promise for achieving practical automotive battery impedance identification and advancing battery diagnosis and health estimation. Furthermore, by identifying the inherent relationship between the charge voltage curve and EIS, we can enhance our understanding of the electrochemical implications embedded within the charge and discharge curves. This deeper comprehension enables us to conduct in-depth health diagnosis and directly explore the internal degradation modes through the battery's charge and discharge curves.

7. Conclusions

Impedance measurements are effective methods for battery health diagnosis. From a mechanism perspective, both DC techniques and EIS can be utilized to identify battery impedance and their related electrochemical processes, and DC impedance measurement can yield results equivalent to EIS, such as, R_{ohm}, R_{sei}, R_{ct}, and R_{mt}. However, DC impedance measurement is underdeveloped at present and its implementation in real-life application is challenging, and many parameters (e.g., time constants of electrochemical processes) cannot be accurately determined. With the development of computational methods, DC impedance measurement is constantly being simplified, with the efficiency and accuracy being advanced. Machine learning methods can enable the prediction of EIS results based on DC curves by capturing the relationship in between. Overall, DC impedance measurement is promising and feasible to replace EIS measurement for battery health status diagnosis. It is expected that accurate battery health status will be obtained directly from DC testing in the near future.

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Conflict of Interests

The authors declare no conflict of interest.

Keywords: Battery diagnosis · Electrochemical impedance spectroscopy (EIS) · Direct current (DC) impedance · Constant current technique

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