Data-driven health estimation and lifetime prediction of lithium-ion batteries: a review

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Abstract

Accurate health estimation and lifetime prediction of lithium-ion batteries are crucial for durable electric vehicles. Early detection of inadequate performance facilitates timely maintenance of battery systems. This reduces operational costs and prevents accidents and malfunctions. Recent advancements in "Big Data" analytics and related statistical/computational tools raised interest in data-driven battery health estimation. Here, we will review these in view of their feasibility and cost-effectiveness in dealing with battery health in real-world applications. We categorise these methods according to their underlying models/algorithms and discuss their advantages and limitations. In the final section we focus on challenges of real-time battery health management and discuss potential next-generation techniques. We are confident that this review will inform commercial technology choices and academic research agendas alike, thus boosting progress in data-driven battery health estimation and prediction on all technology readiness levels.

Keywords: Lithium-ion battery, Data-driven approach, Ageing mechanism, Battery health diagnostics and prognostics, Electric vehicle, Sustainable energy

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Nomenclatures

ANN	Artificial Neural Network
BOL	Beginning of Life
BMS	Battery Management System
CC	Constant Current
CV	Constant Voltage
CE	Coulombic Efficiency
DA	Differential Analysis
DV	Differential Voltage
DTV	Differential Thermal Voltammetry
DMP	Differential Mechanical Parameter
DOD	Depth of Discharge
EOL	End of Life
EIS	Electrochemical Impedance Spectroscopy
EV	Electric Vehicle
FFNN	Feed Forward Neural Network
GPR	Gaussian Process Regression
IC	Incremental Capacity
ICF	Incremental Capacity Curve Based on Measured Force
LAM	Loss of Active Material
LLI	Loss of Lithium Inventory
Li-ion	Lithium-ion
KF	Kalman Filter
LFP	Lithium Iron Phosphate
ML	Machine Learning
MA	Moving Average
NMC	Lithium Nickel Manganese Cobalt Oxide
PDF	Probability Density Function
PF	Particle Filter
RVM	Relevance Vector Machine
RUL	Remaining Useful Life
RNN	Recurrent Neural Network
SEI	Solid Electrolyte Interface
SOH	State of Health
SVM	Support Vector Machine
SVR	Support Vector Regression

1. Introduction

Lithium-ion (Li-ion) batteries have been widely applied as energy storage systems, such as electric vehicles (EVs) and hybrid electric vehicles (HEVs) [1] . The performance of Li-ion batteries deteriorates with time and use due to the degradation of their electrochemical constituents, resulting in capacity and power fade [2]. This is called battery ageing and is a consequence of multiple coupled ageing mechanisms influenced by different factors such as battery chemistry and manufacturing, as well as environmental and operating conditions. The point when the battery fails to meet the energy or power requirement for its application is commonly defined as the end of life (EOL). To ensure the safety and reliability of batteries despite ageing, health diagnostic and prognostic tools are required. State of health (SOH) estimation techniques have been developed to track the actual performance of batteries in operation. The SOH reflects the current capability of a battery to store and supply energy/power relative to that at the beginning of its life, calculated as the ratio of the actual cell capacity/resistance and its initial value. Generally, for applications where the available energy in the battery plays a fundamental role, such as in EVs, the capacity is often used for SOH characterization [3,4]. In applications where power is of interest, such as in HEVs, the internal resistance is usually employed as a SOH metric [3,4]. Typically, batteries are considered at EOL (and therefore sentenced to replacement) when their capacities drop below 80% of the initial values or when their internal resistances doubled [3]. Internal resistances can be measured by different methods (e.g. electrochemical impedance spectroscopy [5] and hybrid pulse power characterization [6]) and show high sensitivity to experimental conditions such as the cell state-of-charge [5], and the electrical contact resistance between the connectors and terminals of the cell, etc.. In contrast, capacity is a directly measured value by Coulomb counting method under galvanostatic charging/discharging conditions and thus perceived as a more straightforward descriptors for EOL and SOH diagnosis, although still dependent on usual control parameters such as currents and temperature. The existing literature on SOH estimation is rather extensive albeit focuses mostly on capacity estimation, which will also be our focus unless otherwise specified.

The prognostics of battery health concerns the battery energy/power degradation in the future and predicting how soon the battery performance will become unsatisfactory [7]. The health prediction requires the knowledge of the current and historical degradation signals, often obtained from the SOH estimator to forecast the future state of the system under certain operating conditions. The developed SOH estimation and health prediction algorithms are then implemented in the battery management system (BMS) for online monitoring. With the battery health and lifetime information, users can monitor the performance of the cells and can schedule any maintenance or replacements in advance.

A variety of SOH estimation methods have been developed over the years. One common way is through the use of models simulating the behaviours of cells [8,9], followed by various optimization algorithms and observers, such as the Kalman filter family [10] and particle filter [11] to identify the parameters and SOH states [4]. A widely-used approach is the use of electrochemical models that apply partial differential equations to simulate mass and charge transfer kinetics that are closely related to aging [12]. Also widespread are electrical models which use electrical-circuit analogs such as resistors and capacitors, to simulate the cell dynamics under different input currents [13]. This field has been particularly vivid and a number of review papers on this topic exist [14–16].

Data-driven methods for health estimation and prediction are gaining increasing interest in both academia and industry due to their advantages of flexibility and being model-free [17]. Here, we define them as the techniques requiring a large set of ageing data, having their effectiveness is heavily dependent on the quality and size of the dataset. Several technologies in this category are noteworthy. First, due to the correlation between the SOH and electrical, thermal and mechanical behaviours of a battery, differential analysis emerged as an effective tool which uses information on voltage, surface temperature and strain under different aging states. Next, by fitting a large amount of data collected under predefined experimental conditions, lifetime estimation models have become another popular technique with high computational efficiency and acceptable accuracy assuming similar operating conditions. Finally, due to their flexibility and nonlinear matching ability, machine-learning methods are among the most popular data-driven techniques for both health estimation and prediction. Specialized aging tests incorporating multiple factors affecting battery health are conducted to generate a suitable training dataset. Next, an underlying relation is synthesized by mapping these factors to the battery health state using different intelligent techniques. Data-driven methods are becoming one of the most prominent approaches to battery health estimation and prediction for real applications as they do not involve complex physical models.

To date, a few review papers on SOH and RUL estimation have been published, summarized in Table 1. Some [3,18–20] focus on one aspect (SOH or RUL) alone. As SOH estimation is often used as an input for ageing models/RUL predictors, these two topics are heavily correlated, and a review paper covering both is required. In the reviews where both are surveyed, e.g. [7,21], data-driven approaches are still not covered in depth.

Table 1. An overview of the published literature related to battery SOH estimation and RUL prediction

Topic	Reference	Content
Health estimation	Xiong et al. (2018) [18]	General review on SOH estimation methods
	Berecibar et al. (2016) [19]	General review on SOH estimation methods and ageing mechanism diagnosis tools
	Farmann et al. (2015) [3]	Review of SOH estimation techniques for EV and HEV application
	Rezvanizaniani et al. (2014) [22]	Battery health estimation and safety management by roughly focusing on physical models, data-driven and fusion methods
	Lu et al. (2013) [4]	Key functions of the BMS, such as the state of charge estimation, SOH estimation, cell balancing and fault diagnosis
	Barre et al. (2013) [23]	Ageing mechanism and SOH estimation for automotive applications
	Zhang et al.(2011) [7]	Battery state of charge estimation, health monitoring, fault detection, correction, and RUL prediction
Health prediction	Lucu et al. (2018) [20]	RUL prediction methods focusing on self-adaptive battery ageing models
	Lipu et al. (2018) [21]	General review of battery SOH estimation and RUL prediction methods

Here, we give an overview of data-driven estimation and prediction methods applied to Li-ion batteries and discuss their challenges. The review is intended to inform commercial technology choices and academic research agendas alike, thus boosting progress in data-driven battery health estimation on all technology readiness levels. We cover the following topics:

- In Section 2, Battery ageing mechanisms and the most common stress factors are discussed as the fundamentals for developing data-driven prediction methods.
- For battery SOH estimation, data-driven technologies including differential analysis, machine learning, and others are reviewed in Section 3, and their benefits and drawbacks are discussed.
- For battery health prediction, technologies including analytical models with data fitting, and ML methods are comprehensively surveyed in Section **Error! Reference source not found.**.
- A compilation of the existing issues and challenges is given in Section 5. Feasible and cost-effective solutions to address the current challenges are suggested as future work directions toward the improvement of data-driven based SOH estimation and RUL prediction technologies.

2. Li-ion battery ageing mechanisms and stress factors

The success of health estimation and prediction tools depends on how well the battery ageing processes and their causes are understood and translated mathematically [24]. Many studies have been dedicated to identifying fundamental cause-effect relations for the loss of performance. Here, we summarize the most common ageing mechanisms in battery and provide an overview of the main stress factors. The interested reader may refer to the literature dedicated to this topic [25–27].

A widely accepted categorization divides the main degradation modes acting in Li-ion as three: the loss of lithium inventory (LLI); the loss of active material (LAM) in the electrodes and the increase of cell internal resistance. LLI is mostly related to the consumption of Li-ions by side reactions, such as solid electrolyte interface (SEI) formation on the surface of the graphite negative electrode, electrolyte decomposition reactions or lithium plating [23]. Such side reactions irreversibly consume Li-ions, making them unavailable for subsequent charge/discharge. LAM normally originates from a combination of factors. One is the structural deterioration of electrodes due to volume changes of active materials during cycling. These induce mechanical stress, leading to particle cracking and reducing the density of lithium storage sites. Other factors include chemical decomposition and dissolution reactions of transition metals into the electrolyte and SEI modification [23,28]. The resistance increase of the cell can be caused by the formation of parasitic phases, such as SEI, at the electrode surface, as well as the loss of electrical contact inside the porous electrode [23].

Batteries deteriorate even when not in use ("calendar aging"). In contrast, cyclic aging refers to the ageing from the continuous battery charge/discharge cycling. Understanding both modes is extremely important for a better design and implementation of SOH estimation and RUL prediction tools. High storage state of charge (SOC) and high temperature are the main drivers of calendar ageing [29]. High SOC implies low Li content in the active material of the positive electrode (cathode). This increases the tendency of the electrode to chemically decompose electrolyte components. Behind that is the same chemical driving force that creates the higher cell voltage at higher state of charge, i.e., a higher driving force for Li to re-enter the electrode. Calendar ageing will inevitably occur throughout the battery life regardless of the operating mode, and all the factors in calendar ageing also affect cyclic ageing. The latter, however, is affected by additional factors, such as over-charge/discharge, current rate and cycling depths. These factors are not linearly correlated, which complicates the ageing process considerably [26].

High Temperatures: Accelerate side reactions, including (i) SEI layer growth rates on the anode, resulting in faster LLI and cell resistance increase [30], (ii) metal dissolution from the cathode [30], and (iii) electrolyte decomposition, with (ii) and (iii) leading to LAM and LLI. Extremely high temperatures may trigger "thermal runaway", the ultimate threat [31].

Low Temperatures: Slow down the transport of Li ions in both electrodes and in the electrolyte. Where the electrolyte meets the graphite electrode, attempts of fast charging at low temperatures may thus create crowding of Li ions. This may cause (local) lithium plating of graphite [32] which comes with LLI. Continuous inhomogeneous lithium plating will eventually cause the growth of lithium dendrites, which may penetrate the separator and short circuit the cell.

Over-charge/discharge: When a cell is overcharged, the cathode is over-delithiated (no active lithium available) and the anode is over-lithiated (no more 'room' for lithium). The cathode material suffers from irreversible structural change when over-delithiated [33], followed by the dissolution of transition metal ions (such as Mn^{2+}) and active material decomposition [34]. Decomposition of the electrolyte and significant increase of the total internal resistance were found during the overcharging process [35]. Overcharging the cell can generate significant heat, due to Joule heating and the heat generated by a series of side reactions at both electrodes [36]. During over-discharging, the anode potential increases abnormally which leads to the anodic dissolution of the copper (Cu) current collector and formation of Cu^{2+} ions [37]. Upon recharging, the reverse reaction can form copper dendrites, which may lead to internal short circuit [38].

High currents: Excessive charge and discharge currents can cause localized overcharge and discharge to occur, leading to the same degradation reactions as generalized overcharge and over discharge. High currents come with more heat waste, which can raise the cell temperature and concomitantly the rates of ageing processes. Once Li-ion batteries use organic electrolytes, their relatively low heat capacity make them especially prone to rapid temperature increase upon current flow if compared to water-based batteries. For graphite anodes, fast charging also results in metallic Li plating due to the graphite's limited ability to accept Li ions at high rates, leading to LLI[39].

Mechanical stresses: Cells are subjected to stress from different sources, such as manufacturing (e.g. externally applied stack pressure) [40], electrode material expansion during operation [41], gas evolution in mechanically constrained cells and external loading during service. The highest stresses tend to be generated in the electrode particles near the separator, where cracking and fracture are most likely to take place [42]. When stress exceeds a certain limit, the electrode experiences material failure, associated with cracking or fracture. That results in significant degradation of cell performance and capacity fade [43].

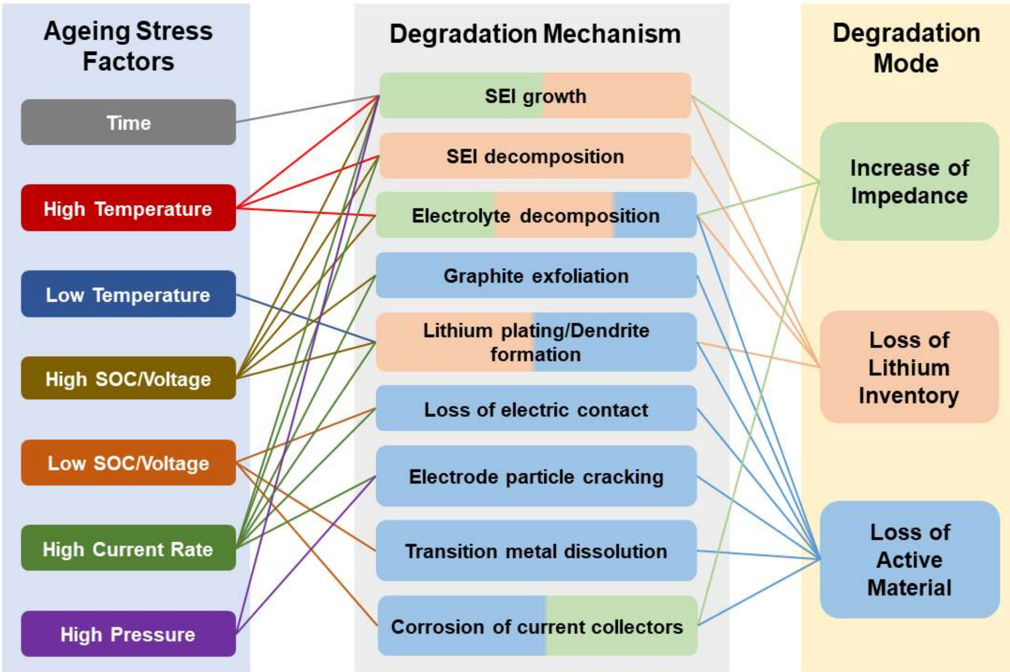

Fig. 1. Battery ageing impact factors during cycling and their associated degradation modes, adapted from [28]

The contributions of ageing stress factors to cell performance are outlined in Fig. 1, providing a proxy to describe the conditions which increase the ageing rate [24]. In large-format battery systems, the BMS is usually responsible for controlling the operating conditions to extend longevity and ensure safe operation. For instance, over-charge/discharge protection can be achieved via voltage regulation by the BMS: (dis)charging is stopped when one cell in the pack reaches a fully charged or discharged state. The thermal management system can actively heat/cool the batteries to ensure their temperature is in the range where the degradation rate is minimal. Developing an optimal charging protocol to achieve good trade-off between battery capacity fade and charging time [44–46].Understanding the impact of ageing factors is also essential to develop reliable health diagnostic and prognostic tools. Data-driven methods are largely based on the quantity and quality of experimental ageing data, but it is in practice impossible to test the batteries under the full range of potential operating conditions. For a specific application, some of the stress factors can play more important roles in the battery ageing than others. A quantitative relationship between operating conditions, stress factors, and ageing processes has to be focused on those with the greatest impact [24], this should be considered when designing the experimental testing scheme.

3. State of health estimation

Only high-fidelity data-driven approaches of SOH estimation are reviewed in this section, for conciseness. A differential analysis (DA) involves identifying features from the differentiated curves of the electrical, thermal or mechanical parameters during battery cycling, and correlating them with battery capacity fade. Machine learning methods, on the other hand, require training a model based on the extracted input features from the measured data of a BMS to describe the cell ageing behaviour and estimate the SOH. Other methods include constructing correlations between the intrinsic characteristics of the cell such as coulombic efficiency with its capacity fade. Before being applied to online SOH estimation, all the health estimators need to be built offline, based on experimental data after sufficient tuning and validation to ensure reproducibility and accuracy.

3.1 Differential analysis to identify features

DA in the context of batteries is based on the differentiation of curves containing the electrical, thermal or mechanical signals, obtained upon galvanostatically charging or discharging of a cell. Incremental capacity/differential voltage (IC/DV) analysis, differential thermal voltammetry (DTV) and differential mechanical parameter (DMP) analysis are most frequently mentioned. This subsection introduces the basics, application, and limitations of each DA method.

3.1.1 IC/DV analysis

IC/DV analysis provides a non-destructive means of characterization of cells and has been widely used for ageing mechanism identification [47,48]. IC is calculated by differentiating the change in battery capacity to the change in terminal voltage for a sufficient small-time interval, while DV is defined as the inverse of IC. The differentiation transforms voltage plateaus in charge/discharge curves into clearly identifiable peaks in

IC curves and valleys in DV curves. Peaks in the DV curve (plotted *vs*. cell capacity) indicate phase transitions in the electrodes, whereas peaks in the IC curve (plotted *vs*. cell voltage) represent the location of a phase equilibria [49]. Each peak in the curve has unique features, like intensity and position, reflective of a specific electrochemical process in the cell [50]. Both can provide ageing information, with one significant difference. IC curves refer to the cell voltage, which can be a direct indicator of the battery state. DV curves instead, refer to the cell capacity, which is a secondary indicator that varies with battery degradation and loses reliability as a reference in the course of ageing [51]. Through the progression of each peak in IC/DV curves throughout ageing, and observing the change of the active materials over time, degradation mechanism can be distinguished [52]. To study ageing, low charging/discharge current rates (e.g. C/20 or less) are typically used, as the peaks in the differential voltage spectrum are more pronounced and the polarization influence on IC curves is lower.

IC/DV analysis is also a powerful tool for online SOH estimation [53,54]: it can be easily implemented in a BMS by monitoring two parameters only (voltage and charge/discharge capacity) and is suitable for different types of Li-ion cells, regardless of the battery chemistry, size and cell design. However, for batteries with flat voltage *vs*. SOC regions (e.g., $LiFePO_4$ or $LiMn_2O_4$ cathodes), data processing via a two-point numerical differentiation is problematic once dV approximates zero, yielding results of infinite slopes. In general, differential curves are very sensitive to the sampling level, cell performance change and measurement noise. Therefore, smoothing is the first and most important step for SOH analysis, achieved by various filtering techniques, such as moving average [49], Gaussian filter [53] and Savitzky-Golay filter [55,56].

Once smoothed IC/DV curves are obtained, the evolution of features linked to capacity fade can be easily identified and tracked by, e.g. peak location [53], height [54] and integrated peak area [57]. For instance, Fig. 2 illustrates the evolution of IC curves of a high energy NMC cell under a charging current of C/3, where all the peaks are found to shift towards higher voltage levels and the peak heights decrease along with cycling [53]. Then, the SOH estimator is developed offline by constructing an analytical function between the battery capacity and the values of features of interests (FOIs) as a function of every possible degradation path. Berecibar et al. [58] estimated the SOH based on peak intervals from DV curves. Note that batteries in real-life applications are charged from different SOC levels, which leads to large changes of peak positions on DV curves due to the variation of charging capacity. That problem does not arise for the peak positions of IC curves since those are not plotted *vs*. capacity but *vs*. cell voltage, which has a more definite position [59] and is not subject to accumulated errors. Hence, IC-based health monitoring is available even during partial cycling. Furthermore, IC peaks are discoverable within a specific SOC range, e.g. around 60% [53], making IC more suitable for online SOH estimation than DV. Weng et al. [54] found the IC peak height to decrease with LFP battery capacity fading. Li et al. [53] used a linear regression relationship to describe the correlation between the battery capacity fade with the variation of peak positions from IC curves for NMC/graphite cells. Table 2 lists the testing conditions and features used in the published literature for single cell capacity estimation based on IC/DV analysis. As can be seen, this method is restricted to data obtained at low C-rates. At high current rates, the peaks are offset by the over-potential caused by the impedance of the cell, which is a stronger function of temperature than ageing [60]. Temperature can introduce significant errors in any real application of IC/DV curves.

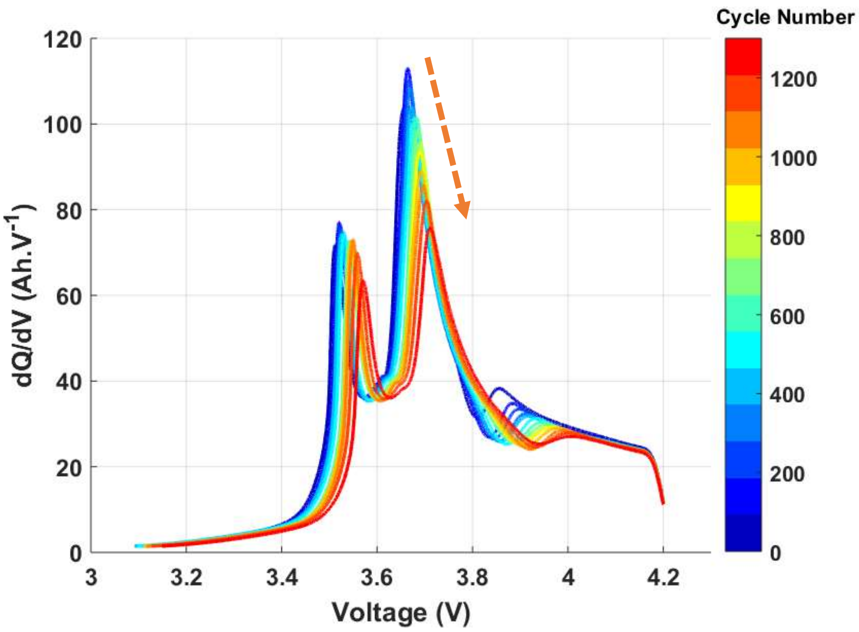

Fig. 2. IC curves for a high energy NMC/graphite cell under charging current of 0.3 C after different cycles at 25°C, and the decreasing of peak intensity and shifting of peak location can be observed during cell ageing. [53]

Table 2. Summary of the features and estimation methods used for single cell capacity estimation using IC/DV analysis

Differential analysis	Current rate	Features	Battery Chemistry	Ref.
IC	C/10	Peak height, peak area	LFP	Jiang et al. [61]
IC	C/2	Peak position	NMC	Li et al. [53]
IC	1C	Integrated area surrounding the peak	NMC	Tang et al. [57]
IC	C/2	Peak height	LFP	Weng et al. [54]
IC	C/20	Peak position and height	NMC	Zhang et al. [62]
DV	C/5	Regional capacity	LFP	Berecibar et al.[58]
DV	1C	Normalized location interval of two consecutive transformation parameter	LFP	Wang et al.[63]

3.1.2 Differential thermal voltammetry (DTV) analysis

DTV analysis can be used as a complementary tool to existing SOH diagnostic techniques, which combines the concept of IC analysis with temperature measurements to infer thermodynamic information about the electrode materials [55,64]. The DTV technique probes the cell surface temperature during galvanostatic (dis)charges, and it is obtained by differentiating the temperature (T) with respect to voltage (dT/dV) and plotting against cell voltage. DTV was designed to easily and quickly reveal the most pronounced entropy-related information during cell operation. Maher and Yazami [65] showed that the entropy (change) profiles of aged cells show variation in peak positions and amplitudes similar to those displayed by IC/DV profiles. For instance, shrinking peaks can be assigned to the increasing number of point defects in the active materials [66,67]. DTV provides similar information as IC analysis yet with the additional information of entropic nature. Each peak in the DTV might be attributed to a particular phase transition of either the negative or positive electrodes, or be a result from the combination of both when full cells are investigated using this technique, as shown in Fig. 3 (a) and (b).

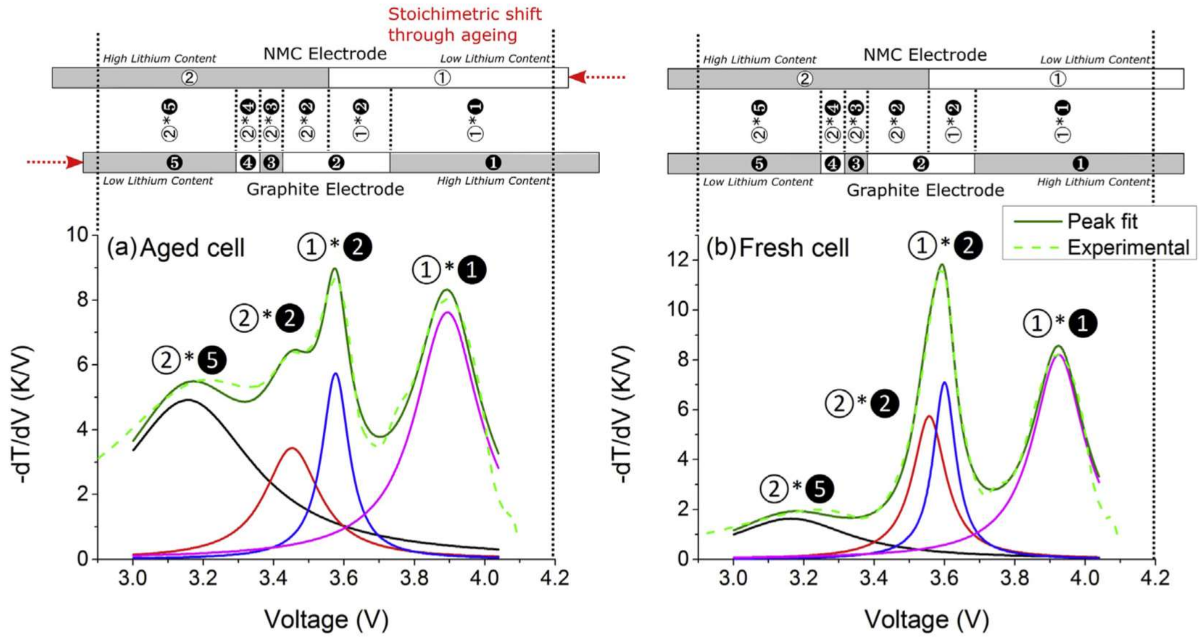

Fig. 3. DTV results and peak fitting obtained from a 2 C constant current discharge in both (a) aged and (b) fresh cells. Each peak can be assigned to a phase transition from negative and positive electrodes combined [68].

The peaks with greatest change in the peak parameters (e.g. position, height and width) can be used for diagnosing the cell degradation, such as capacity fade, resistance increase and inhomogeneous electrode performance, showing potential for SOH estimation in real applications [55,60]. Merla et al. [55] carried out DTV analysis and found that the peak position and height, both correlated with the cell impedance rise, change significantly with battery ageing. In their following work [68], they demonstrated the applicability of DTV for monitoring the health information of single cells connected in parallel. In a technique similar to DTV, Wu et al. [69] extracted an indicator from the temperature variation curve (dT/dt) using thermistors for battery SOH estimation and found that the time period from starting the charging process to the minimum in the dT/dt curve show a linear correlation with the battery health state. Worth mentioning the validity of this method has not been fully demonstrated on the cells with partial charged condition and no heat flux sensor was employed.

Generally, DTV is invoked as experimentally easy and applicable for parallel connected cells with the advantage of enabling higher current rate tests than the ones required for IC/DV analysis, for instance [68]. It only requires monitoring the parameters of voltage and temperature during the galvanostatic charge/discharge process, which shows great potential for online applications. Moreover, DTV does not require strict isothermal conditions; in fact, isothermal conditions ($T \approx$ const.) have no meaning for DTV analysis. The heat flux from the cell will only begin to affect the results if the cell temperature rises significantly above the ambient [55]. Appropriate DTV signals require the heat flux to the surroundings to be low in comparison with the rate of heat generation that dominates the dT/dV term, meaning that there is no necessity of super-efficient cooling of the batteries, making such analysis experimentally easier. However, DTV analysis is easily influenced by the testing temperature environment, and fluctuations of ambient temperature can introduce large noise hampering the extraction of meaningful data and further interpretation.

3.1.3 Differential mechanical parameter (DMP) analysis

Some recent work has also been directed toward understanding and modelling the mechanical behaviour of batteries for battery SOH estimation, such as the variation of cell level strain (ε) and stress [70,71]. The intercalation/de-intercalation of Li-ions in/from the electrode active materials is associated with volume change, expanding and contracting in repeatable patterns [72]. Mechanical stress in a cell evolves as a result of electrode expansion against a constraint normal to the plane of electrodes. It can be measured by load sensors on the cell surface.

Cannarella et al. [71] proposed to estimate battery SOH through the measurement of cell strain, after showing that the stress resulting from electrode expansion is linearly correlated with the SOH. The physical basis for this relationship is thought to be SEI growth. A few studies have investigated the first and second derivative of strain with respect to charge ($d\varepsilon/dQ$ and $d\varepsilon^2/dQ^2$) [56,73] and strain differential to voltage ($d\varepsilon/dV$) [72]. They postulated these curves bear similarities to IC/DV analysis and might indicate phase transitions in electrode materials. The fixtures for measuring cell level vary somewhat across this research area. Oh et al. [73] measured the strain of the cell during cycling by high-precision displacement sensors and used the $d\varepsilon/dQ$ curves to identify the phase transitions in the negative electrode. They pointed out that the measured strain caused by cell swelling is a significant factor in cell performance and claimed that strain derivatives have potential for SOH estimation. Sommer et al. [72] measured the cell strain through optic fibre sensors and plotted the derivatives $d\varepsilon/dV$ as a function of voltage. They claimed that the peaks in the differential curves representing an increase in strain at some voltages can be assigned to phase transitions. Schiffer et al. [56] measured strain via a linear variable differential transformer: an electromechanical sensor that converts mechanical vibrations into variable electrical signals. The second derivative of the strain with respect to capacity ($d\varepsilon^2/dQ^2$) was shown to exhibit similar shifts in peaks as those expected in the DV curves during the cell degradation process but in a more consistent and reliable manner. The $d\varepsilon^2/dQ^2$ curves were applied for identifying phase transitions in electrode materials at higher current rates than the DV analysis, making the method more time efficient.

However, strain measurement is only applicable to unconstrained cells that can expand freely. In a battery pack this expansion is limited, making it difficult to measure strain. Stress, on the other hand, resulting from expansion in a constrained space can be measured instead. Samad et al. [74] measured it with a force sensor on the end plates of a battery pack, and developed a method for battery capacity estimation by using IC curves based on measured force (ICF, dQ/dF). A linear relationship was found between the battery capacity fade and the increase of peak voltage in both ICF and IC curves as shown in Fig. 4. They claimed that the data processing of the ICF curve was easier than IC analysis, as ICF has a better signal to noise ratio than DV curves as the amplitude of the force signal is much bigger than that of the voltage signal. The identified ICF peaks are at a higher SOC level (around 70%) than the peaks in IC curves (around 40%) and indicate that an ICF-based SOH monitoring method could be updated more frequently during regular use of an EV or HEV where the SOC does not usually fall below 50%. While strain derivatives may contain the same information as IC/DV analysis, there is a barrier to using this method as it requires an additional apparatus to collect the required data. Despite these disadvantages, the strain derivative analysis remains a practical tool when cells are cycled under higher current rates, which is the main hurdle for IC/DV analysis [56].

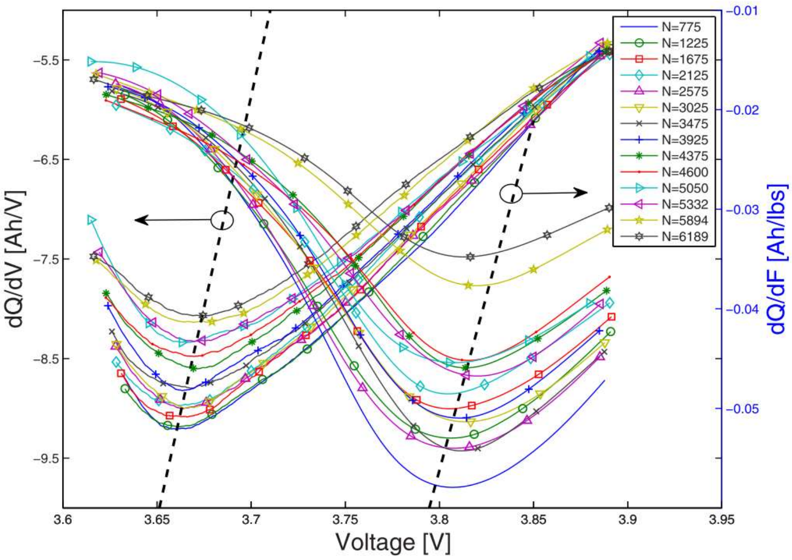

Fig. 4. The IC (dQ/dV) and ICF (dQ/dF) curves of the tested cell during 1C discharge capacity test after different cycles, the peaks in both the IC and ICF curves shift linearly as the number of cycles inreases. [74]

In a nutshell, each differential analysis has its merits and demerits and should all be taken as complementary techniques to aid the reasoning behind battery aging. To enable battery users to select the appropriate method for a given application, the characteristics of these three DA methods are compared in Table 3.

Table 3. Summary of the characteristics of differential analysis techniques proposed for online battery capacity estimation

Methods	Advantages	Disadvantages
IC/DV analysis	• Easy to monitor, only needs two parameters (voltage and capacity); • Can be applied to batteries with different types, sizes and chemistries; • Works for partial charging/discharging conditions; • Easy to be implemented in BMS for online applications.	• Limited to low current rates (< 1 C); • Sensitive to measurement noise – requires smoothed curves; • Influenced by the operation temperature; • Computing dV for chemistries with large voltage plateaus (e.g., LFP cells) might yield infinite solutions.
DTV analysis	• Easy, only needs two parameters (voltage and temperature); • Can be used for monitoring cells in parallel; • Applicable for partial charging/discharging conditions; • Easy for BMS implementation	• Needs additional and calibrated temperature sensors; • Sensitive to testing temperature variations; • Challenges in overcoming noise in the temperature measurement.
DMP analysis	• Can be applied for cells with a high initial SOC; • Not limited to low and constant current rates; • Applicable to high current rates.	• Needs additional equipment for the mechanical parameter measurement; • Not applicable to cells constrained with hard covers; • Difficult for online application.

3.2 Machine learning (ML) methods for health estimation

ML is a method of data analysis that automates analytical model building. It is based on the idea that systems can learn from data, identify patterns and make decisions or predictions with minimal human intervention. Fig. 5 illustrates the basic workflow required for the application of machine learning for the online SOH estimation. The first step is data collection. Measurable battery parameters, such as temperature, current and voltage data recorded by the BMS during operation are recorded and used as the inputs for training the model. However, not all data are relevant to cell ageing. A second step is to extract the features representative of the ageing process. The third step is to train a machine learning model to describe the relationship between the battery SOH and the extracted features. Once the model is trained, the last step is implementing it in a BMS for online application.

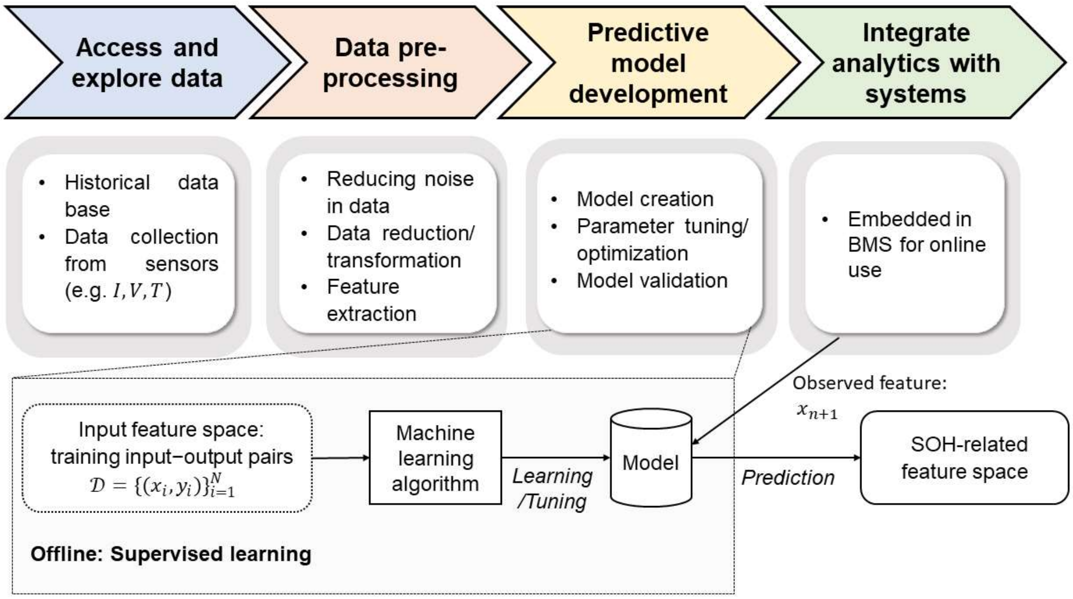

Fig. 5. Generic workflow for health prediction models using ML-estimated features. The learning machine senses the environment and stores data in memory and constructs the mapping from the feature space to the state space.

Feature extraction is a critical step and significantly affects the SOH estimation performance. More meaningful and accurate input data will produce more relevant and accurate predictions. This section presents an up-to-date overview of machine learning-enabled SOH estimation from the perspective of different input features for model training.

Group 1: Model fitted features. Several studies used model fitted features such as internal resistance, capacitance and SOC to train their machine learning SOH estimators. These features cannot be directly accessed from the BMS sensors and must be inferred by an underlying electrical model and online parameter/state estimation algorithms. For example, Pan et al. [75] and Yang et al. [76] used battery health model parameters such as ohmic resistance, polarization resistance and polarization capacitance as input features to train the machine learning algorithm. This approach requires the utilization of an electrical model with online state estimation algorithms, such as recursive least square algorithm or extended Kalman filters.

Group 2: Processed external features. Processed external features are normally extracted from differential charging curves under constant current rate, such as IC/DV curves [77,78] and voltage gradient curves (dV/dt) [79,80]. As discussed in Section 3.1.1, the variation of peak features has a strong connection with the ageing process. Berecibar et al. [78] and Wang et al. [77] trained the proposed ML models with a selection of features from IC/DV curves for cell capacity estimation. Wu et al. [79] used geometric features such as arc length and curvature from the voltage rate of change curves as input data for model training.

Group 3: Direct external features. Direct external features can be recorded directly by the sensors in a BMS during operation without the use of models. These include terminal voltage, current and temperature. For instance, You et al. [81] cycled batteries dynamically according to various driving patterns and used the measured BMS data (current, voltage, and temperature) to train a machine learning model, allowing the battery health estimation during dynamic operating conditions. Richardson et al. [82] and Li [83] proposed to train the model with the voltage-capacity data recorded in a specific voltage region under static charging conditions.

To highlight the advantages and disadvantages of the ML-based SOH estimation methods from the perspective of extracted features, a comparison is illustrated in Table 4. The model fitted features are not directly available but must first be calculated based on the BMS data. Those calculations rely on complex models and are thus not well suited for real-time applications. For the models using processed external features (group 2), a constant current is generally required, restricting their application to galvanostatic charging. In addition, due to the limited computational capability of the present BMSs, many external features are hard to obtain in operation. It seems that ML models with the variables measurable on-board are therefore more suitable for the implementation in more sophisticated devices, such as EVs. A SOH monitoring method which can directly utilize the direct external features (group 3) for battery SOH estimation becomes highly desirable, while the battery modeling and data pre-processing steps should ideally be avoided to reduce the computation effort.

Table 4. Comparison of ML algorithms for SOH estimation of batteries

Categories	Descriptions	Advantage	Disadvantages
Group 1. Model fitted features	• Extra models are needed for simulating the dynamic behavior of the battery such as electrical model.	• Dynamic performance of batteries can be considered.	• The model for simulating the battery working behaviour and the identification of model parameters are computationally expensive for online applications.
Group 2. Processed external features	• The features from differential curves such as peak position, intensity can also be used as input data	• Small number of input features are required for model training.	• Not suitable for dynamic operating conditions; • Constant current charge/discharge is required; • Some of the features can be hard to obtain during operation due to limited capability of the present BMSs.
Group 3. Direct external features	• Recorded directly by sensors in actual BMS; • Smooth methods can be applied to increase the data quality.	• Easy to obtain from BMS; • Suitable for online application.	• The number of input features can be large and therefore increase the computational cost.

When the dataset is collected and represented appropriately, a particular ML model needs to be selected. A wide range of models exists, which can be broadly categorized into supervised-learning and unsupervised-learning models. In supervised learning, the training data consist of sets of input and associated output values. The goal of the algorithm is to learn a mapping from inputs to outputs with an acceptable degree of fidelity [84]. The form of the output values can be within a discrete set (such as categorizing a cell as failed or non-failed) or a continuous set (such as the capacity or resistance value). When the output is categorical, the problem is known as classification; when it is real-valued, it is known as regression. All the battery health estimation and prediction problem fall into the regression category, as they produce a numerical value of SOH or lifetime. Contrary to the supervised learning algorithms, where data scientists determine which variables or features to train the models on and use them to develop predictions, the unsupervised learning algorithms are only fed in the given inputs and their goal is to find "interesting patterns", identify trends or clustering in the data without additional inputs. So far, supervised learning is the most mature and powerful approaches, and used in the majority of machine-learning studies in the battery health diagnostics and prognostics, and therefore the methods described here refer in particular to this type. Various supervised ML techniques have been utilized for battery SOH estimation, including Artificial Neural Networks (ANN) [81], Support Vector Machine (SVM) [85], relevance vector machine (RVM) [86], k-Nearest Neighbours (kNN) [87], Gaussian Process Regression (GPR) [82] or random forest regression (RFR) [83]. Section 4.2 will provide more details of these ML algorithms, given that they are also widely applied for RUL prediction.

3.3 Others

Apart from differential analysis and ML methods, some novel approaches correlating the intrinsic characteristics of a Li-ion cell with its SOH were also proposed. Coulombic efficiency (CE) evolution, calculated by dividing the discharge capacity by the charge capacity, has been found to present a close relationship with battery degradation. Yang et al. [88] studied the correlation between battery degradation and the long-term CE evolution and used CE as an indicator for battery degradation rate of LFP batteries. An empirical model was constructed for capacity estimation based on the measured CE and battery cycles, where the CE was assumed to be constant. Hu et al. [89] demonstrated that the sample entropy taken from the hybrid pulse power characterization data can be correlated with cell capacity loss and therefore used for online SOH estimation. In addition, some studies resorted to additional devices for battery health estimation. Ladpli et al. [90] developed a built-in acoustic-ultrasonic guided wave technique to monitor battery SOH and discovered a non-linear correlation between the remaining battery capacity with the guided wave signal features. However, these studies are still in the early stage and require dedicated testing procedures and instruments for health estimation, which are cumbersome for online application.

4. Health prognostic techniques

The battery health predictor is another key part of the BMS which provides the information on the remaining service time of the battery system. The existing research on battery health prognostics includes the battery remaining useful lifetime (RUL) prediction [91] and capacity (fade) forecasting [92]. The RUL is typically predicted based on a modelled degradation signal reaching a predefined failure threshold, and obtained by using the estimated life of the training units minus the current life position of the test unit. Battery capacity forecasting tools are developed to predict the future changes in SOH as a function of the usage history. Battery health prognostics cannot exist on its own and it needs input from the SOH estimator. Their relationship is illustrated in Fig. 6.

In general, two basic frameworks exist for the battery health prognostics, one based on analytical models and another based on ML methods. The first group requires the development of an ageing model, constructed by fitting an analytical function to a large set of ageing data (e.g. capacity fade) measured under laboratory conditions. In contrast, ML-based methods are model free and can learn from the ageing data itself to forecast the battery health change. The benefits and drawbacks of these methods are compared below. Their challenges are also addressed in this section.

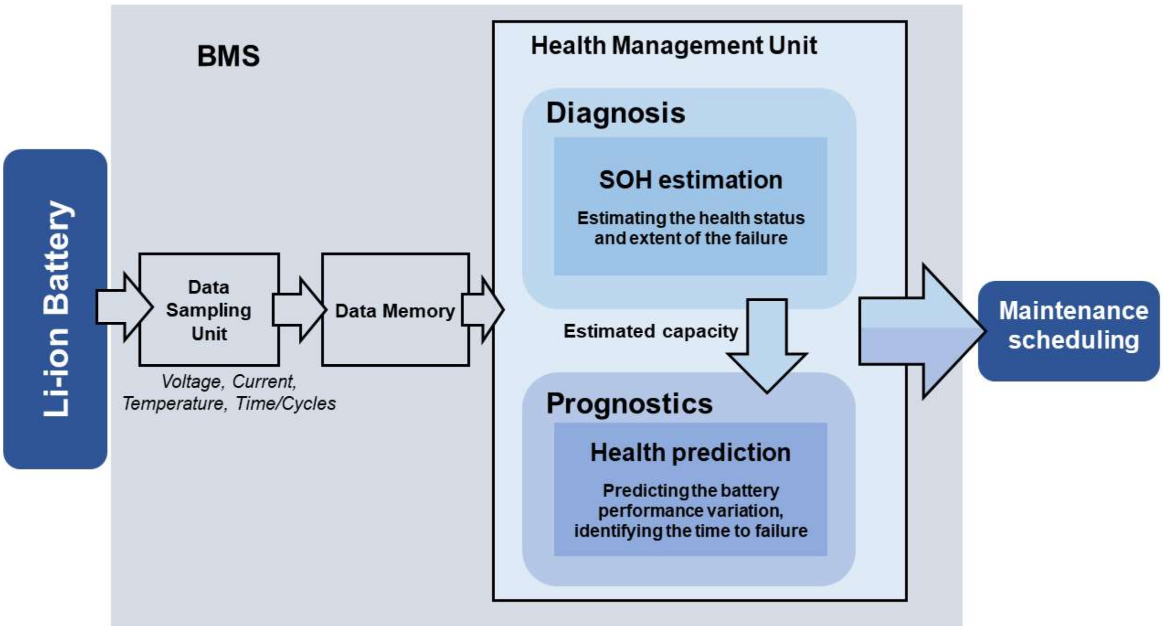

Fig. 6. BMS health diagnostics and prognostics algorithm framework [93].

4.1 Analytical models with data fitting

Analytical-model methods use a mathematical function correlating ageing status of a battery and its service time or cycle number. We discuss the analytical models developed by dividing them into two categories: semi-empirical lifetime estimation models and empirical ageing models with filtering. The former is an open-loop approach, where the model type and parameters are determined by fitting a large amount of ageing data. The model parameters cannot be changed once the model is constructed. Empirical ageing models with filtering are, instead, a closed-loop, and the parameters of this type of models are updated whenever new data becomes available during battery operation.

4.1.1 Semi-empirical life estimation models

Semi-empirical lifetime estimation models capture the direct relationship between the ageing stress factors and battery SOH to obtain a single mathematical expression of the battery performance level over its lifetime. They are constructed by interpolating and fitting data through a fixed set of experimental tests. In principle, for successful lifetime estimation, a comprehensive battery ageing analysis covering a wide range of operating conditions is required. However, it is extremely difficult to consider the effects of all impact factors (as discussed in Section 2). For simplification, only the most important factors are usually considered based on the specific application.

An outline for the development and application of semi-empirical model is illustrated in Fig. 7. Most of the existing studies model the battery cyclic and calendar aging independently, and combine the two to make predictions under a dynamic load profile [6,94,95]. Cells are stored or cycled under specific conditions that help exploring the influences of different ageing factors such as temperature, state-of-charge, charge/discharge current rates. The cell capacity loss is then calculated as a function of time, cycle numbers, or Ah-throughput. Ah-throughput represents the amount of charge delivered from one electrode to the other during cycling. The choice of fitting equations depends on the measured capacity degradation. The parameters of lifetime estimation models are determined by fitting a large amount of ageing data but are difficult to be changed once the model is constructed. During operation, parameters such as current cycle number or Ah-throughput are registered and used as input for current capacity loss estimation (therefore heath state estimation). Moreover, when feeding the model with the battery using conditions and loads, the battery lifetime can be also predicted.

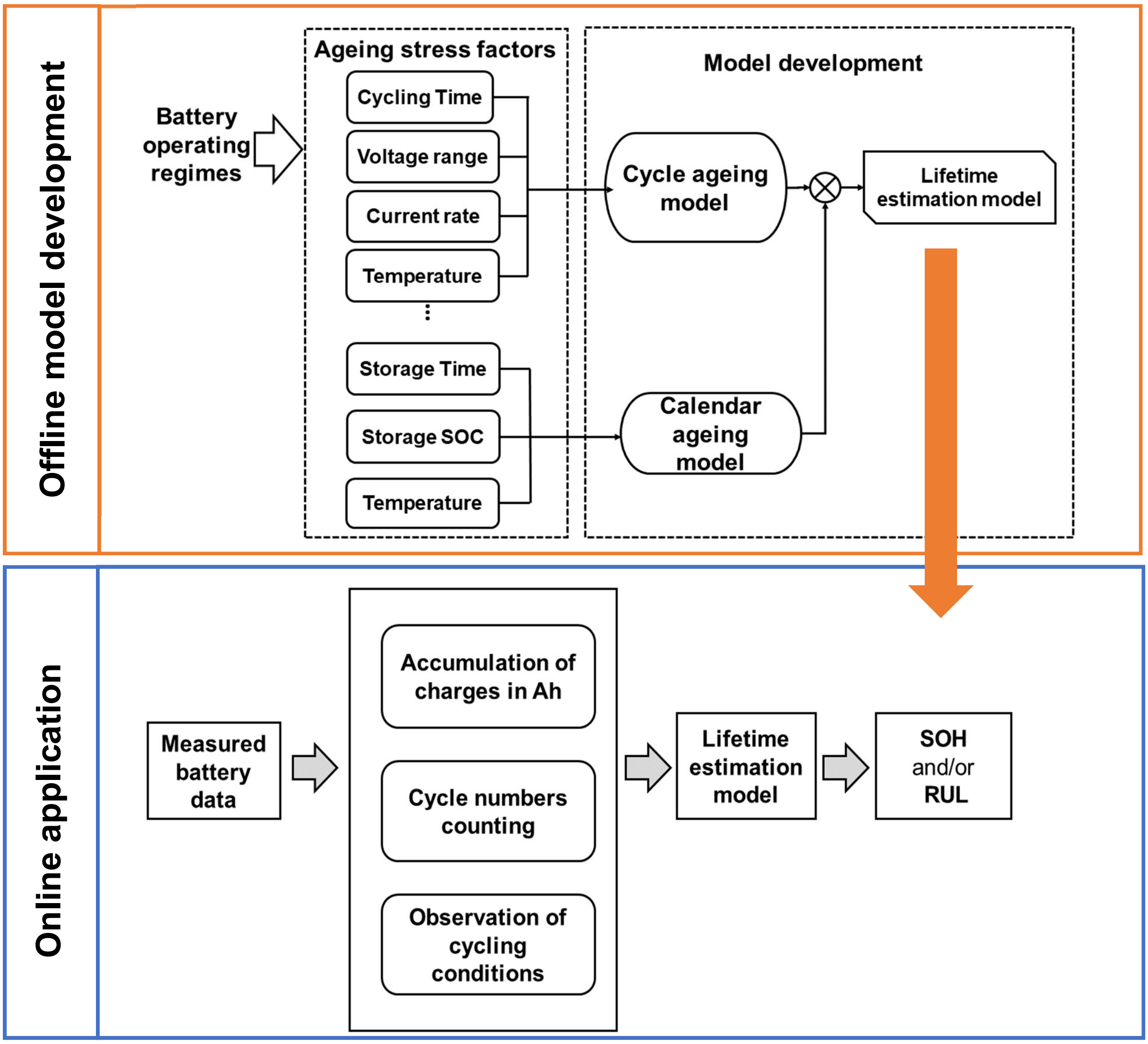

Fig. 7. Diagram of the concept of lifetime estimation model from offline development to online prediction [93,96] .

a. Calendar ageing

The capacity loss due to calendar ageing is usually proportional to a power law relation with time t, weighted by the influences of temperature T and storage SOC, what can be represented by some stress factor k_{cal} [95]:

$$Q_{\text{loss}}^{\text{cal}}(t) = Q(t) - Q\,(0) = k_{cal}(T, SOC)t^{z_{\text{cal}}}, \tag{1}$$

where $Q_{\text{loss}}^{\text{cal}}$ indicates the capacity loss during calendar ageing. $Q(t)$ and $Q(0)$ are the cell capacity at time t and at its BOL, respectively. The exponent z_{cal} is a dimensionless constant. The dependence of k_{cal} on temperature T is empirically modelled through the Arrhenius equation [97–99]:

$$k_{\text{cal}} = A \cdot \exp\left(\frac{-E_a}{RT}\right), \tag{2}$$

where A is the pre-exponential factor and E_a is the "effective" activation energy. "Effective" reflects the fact that there is no single underlying physical or chemical process that could be modelled in an actual kinetic model. Instead, the interplay of all the contributing processes produces an overall observable outcome with a temperature dependence similar to an elementary kinetic process. In contrast, the SOC dependence on calendar ageing lifetime is typically fitted by linear functions [100], exponential functions [99] or by the Tafel equation [95].

b. Cycle ageing

Battery cycle life is sensitive to the operating conditions and is complicated to predict as it involves more variables than the calendar lifetime estimation. The main aspects considered are typically the temperature, cycle number/time, charge/discharge current rate, cycling voltage range and average SOC during cycling. Cycle number is generally used as a measure of time for cycle lifetime modelling, although in some cases Ah-throughput is used instead. An often used cycle ageing model expresses the capacity loss as a power law relation with throughput:

$$Q_{\text{loss}}^{\text{cyc}}(L) = Q(L) - Q(0) = k_{\text{cyc}}(T, I, DOD) \cdot L^{z_{\text{cyc}}}, \tag{3}$$

where $Q_{\text{loss}}^{\text{cyc}}$ indicates the capacity loss during cyclic ageing, and it is an overall capacity difference over time/cycles. L can be either cycle number or Ah-throughput. k_{cyc} represents the effects of ageing factors on the battery degradation process and I is the cycling current. DOD represents the depth of discharge during cycling. Again, the exponent z_{cyc} is a constant extracted from experimental data fitting. Similarly to calendaring ageing, Arrhenius equation is often used to empirically account for the temperature influence [101]. The current rate and DOD dependence on cyclic ageing can be modelled with exponential [95,102] or polynomial [102] functions. Besides, polynomial functions were also used to describe the capacity fade under the influence of cycle DOD and cycle number [97], as shown by (4)

$$\boldsymbol{Q}_{\mathbf{loss}}^{\mathbf{cyc}}(\boldsymbol{L}) = \sum_{i=0,j=0}^{n,m} \left(\boldsymbol{a_i} \cdot \boldsymbol{L^i} + \boldsymbol{b_j} \cdot \boldsymbol{DOD^j}\right), \tag{4}$$

where L is the cycle number, a_i and b_j are fitting constants; n is the order of L-factor and m is the order of the DOD-factor. Fig. 8 illustrates the surface fits constructed with extended measurement data where cycle ageing is modelled as the considered ageing factors.

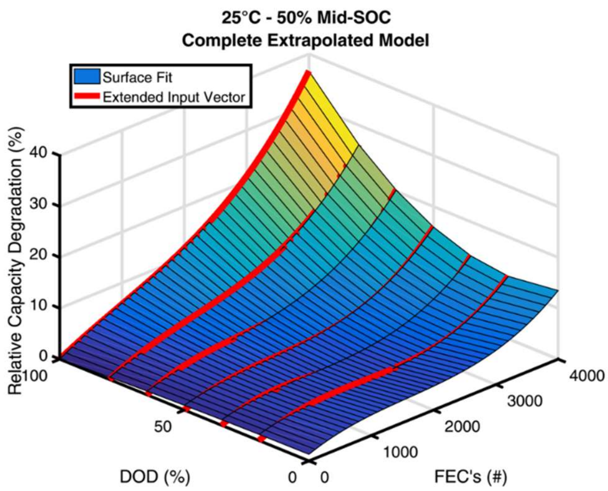

Fig. 8. Illustration of the 3D-surface fitting of the developed cyclic ageing model for lifetime prediction by considering the stress factors of cycling DOD at a fixed temperature and mid-SOC [97]

4.1.2 Empirical ageing models with filtering

Empirical ageing models constantly update their model parameters when new estimated/measured capacity data is available. First, a preliminary ageing model is constructed by fitting the experimental data to an appropriate function that describes the capacity degradation, usually expressed as a function of cycle number or time and fitted model parameters. Linear, exponential and polynomial functions are generally used, summarized in Table 5. Next, the model parameters characterizing the degradation behaviour during operation should be continually updated as part of the prognostic process. This is achieved by various optimal state estimation technologies every time when a new estimated or measured capacity value supplied by the BMS is available. After every update, these models with tuned parameters can provide a more accurate prediction of RUL.

Table 5. Models and filters used in the literature for battery RUL prediction. c_k is the capacity at k^{th} cycle, and a_n are the model parameters, c_k indicates the normalized capacity at the k^{th} cycle, adapted from [20].

Model Equation	Filter	Reference
$c_k = a_1 - a_2 \cdot k$	Fixed-lag Multiple Model PF	Hu et al. [103]
$c_k = 1 - a_1[1 - exp(a_2 \cdot k)] - a_3 \cdot k$	Interacting Multiple Model PF	Su et al. [104]
	Gauss-Hermite PF	Hu et al. [105]
	Fixed-lag Multiple Model PF	Hu et al. [103]
$c_k = a_1 \cdot exp(a_2 \cdot k)$	PF	Saha et al. [106] Zhang et al. [107]
	Spherical Cubature PF	Wang et al. [108]
$c_k = a_1 \cdot exp(a_2 \cdot k) + a_3 \cdot k^2 + a_4$	PF	Xing et al. [109]
$c_k = a_1 \cdot exp(a_2 \cdot k) + a_3 \cdot exp(a_4 \cdot k)$	Bayesian Monte Carlo	He et al. [110]
	Unscented Kalman filter	Chang et al. [111]
	Unscented PF	Zhang et al. [112]
	Heuristic Kalman optimized PF	Duong et al. [113]
	Interacting multiple model PF	Su et al. [114]
	Gauss-Hermite PF	Ma et al. [115]
	Interacting multiple model PF	Su et al. [104]
$c_k = a_1 \cdot k^3 + a_2 \cdot k^2 + a_3 \cdot k + a_4$	PF	Y. Sun et al. [116]

A whole family of Bayesian filters, ranging from the Kalman filter (KF) [117], particle filter (PF) [117] and their variants (see Table 5), provides a general framework for dynamic state estimation problems. In Bayesian inference, the observations are used to estimate and update parameters with a form of a probability density function (PDF) [118]. The choice of the filter depends on the dynamics of the system and the shape of the noise distributions, as well as the filter itself. For a linear system with Gaussian noise, KF is the best candidate. For instance, Burgess [119] proposed a linear capacity fade model with KF to estimate the RUL for valve regulated lead-acid batteries. However, the fading process of Li-ion cells is often non-linear, and variant KF such as extended KF and unscented KF, have been proposed to address this. For the KF family, the state space PDF remains Gaussian at every iteration and the filter equations propagate and update the mean and covariance of the distribution [120]. Note that the errors of RUL prediction come from multiple sources during data acquisition and transmission. Hence, the overall noises thus do not always show Gaussian behaviour. Applying KF algorithms in such scenarios may cause the filter to diverge [91].

The health prognostics process involves solving non-Gaussian problems based on a nonlinear system, which is the strength of the more widely used PF algorithms. PF is a sequential Monte Carlo method that combines Bayesian inference with importance sampling. In PF, the Bayesian update is processed sequentially with particles that have probability information of unknown parameters. When a new measurement is available, the posterior from the previous step is used as the prior information at the current step and the parameters are therefore updated by multiplying it with the likelihood [118]. Numerous studies using PF and its variants have been carried out for RUL prediction.

Saha et al. [121] found that the sum of impedance parameters exhibits a linear correlation with the battery capacity measured at 1 C-rate Development of a lifetime Model for Lithium-ion batteries and can be used as a health indicator for capacity prediction. An exponential impedance growth function was used to describe ageing, which was combined with the PF framework to make predictions of the battery RUL. The result is illustrated in Fig. 9, and it shows that the prediction accuracy of the model can be improved by increasing the considered dataset. The accuracy of empirical ageing models with filtering is highly dependent on the fitting model. For cells with complex ageing behaviour, one single model may not be enough to describe the degradation process. To address this, Hu et al. [103] proposed two empirical models (linear and exponential/linear hybrid model) for representing the capacity fade behaviour of Li-ion cell and a fixed-lag multiple model PF was applied on nonlinear filtering for batteries whose capacity fade behaviour switches between multiple fade models.

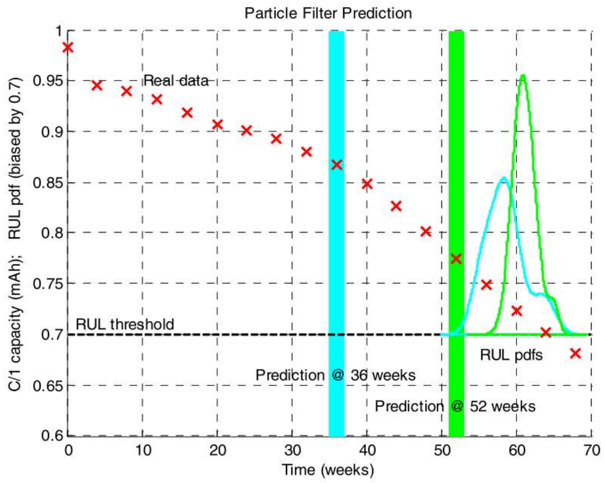

Fig. 9. Schematic illustration of RUL prediction with PF [121]

4.2 Machine learning methods for health prognostics

While ML methods can be utilized for both SOH estimation and RUL prediction[122,123], there is a large difference between the two applications in terms of input features and the desired output. As described in Section 3.2, the input features for SOH estimation should be extracted from the BMS during operation and the outputs are the estimated capacity at a given time. However, the ML methods for RUL prediction generally require the estimated or measured SOH information such as the capacity values as the inputs to predict remaining lifetime or cycles. Supervised ML models can be either non-probabilistic or probabilistic. In the former, the outcomes are determined through known relationship among states and events without modelling the underlying probability distributions. Some methods include ANN, SVM, elastic net and others. Yet, an important aspect of RUL diagnostics is not only predicting the RUL value but also presenting the uncertainty level of the prediction. For this reason, probabilistic models like Gaussian Process regression (GPR) and relevance vector machine (RVM), both deriving from the Bayesian framework, are gaining increased attention for the uncertainty quantification.

4.2.1 Non-probabilistic approach

4.2.1.1 Autoregressive based models

Autoregressive (AR) based modelling is a time series model that uses a linear combination of observations from previous time steps as input to predict subsequent time step values. The AR model has the advantages of easy parameterization and low computational complexity. Long et al. [124] used an optimized AR model for battery capacity fade prediction, where the model order can be changed adaptively by applying particle swarm algorithm. However, the AR model is linear while the battery capacity fading process is generally nonlinear, and this difference will make the model under-fitting especially for the long-term prediction. To solve this problem, an autoregressive integrated moving average (ARIMA) framework was proposed that combines the AR model and the moving average method. Instead of using past values of the forecast variable in a regression, the moving average uses the past forecast errors in a regression-like model. For instance, Zhou et al. [125] combined ARIMA model with empirical mode decomposition to improve the prediction accuracy.

4.2.1.2 Artificial neural network

ANN is designed to mathematically mimic the activity of the human brain, with artificial neurons (the processing unit) arranged in input, output and hidden layers as shown in Fig. 10 (a). The input layer takes the pre-processed data and serves as a conduit to the hidden layer(s) [126]. In the hidden layers, each neuron contains a mathematical model for determining its output based on its input, and can be expressed by a weighted linear combinations that are wrapped in an activation function [126]. The higher the weight of the neuron, the greater its sensitivity to this specific input. The prediction data comes out of the model from the output layer. During the learning process, the model parameters are tuned by considering the number of hidden layers, the number of neurons in each layer, the weights of interconnections between neurons and the type of activation function.

Two types of ANNs have been successfully applied for battery RUL prediction, including feed forward neural network (FFNN) and recurrent neural network (RNN). In

FFNN, the input data travels in one direction only. When FFNN is extended to include the feedback connections, it is called RNN as shown in Fig. 10 (b). RNN can keep and update the previous information for a period of time, making it a promising tool to capture the correlations in battery capacity degradation data. The battery degradation process generally covers hundreds of cycles, and the information of capacity degradation among these cycles is highly correlated. It is thus meaningful to extract and consider these correlations for accurate RUL prediction. Because it can learn the long-term dependencies in the data, RNN is a promising NN type to capture and update information from degradation data.

Based upon the analysis of terminal voltage of charging curve under various cycle numbers, Wu et al. [127] used FFNN to simulate the relationship between battery charge curves at a constant current and battery RUL. The total cycle number when the battery comes to its EOL was taken from experiments, and they used the FFNN to estimate the current cycle number of the battery. The RUL was then calculated by subtracting the current cycle number from the total cycle number. Liu et al. [128] proposed an adaptive RNN to predict the RUL of Li-ion cells, relying on a history of cell impedance data from multiple batteries as a starting point to predict the unknown impedance variation of a new battery. The proposed method can enhance the prediction accuracy by utilizing the previous system states through adaptive/recurrent feedbacks.

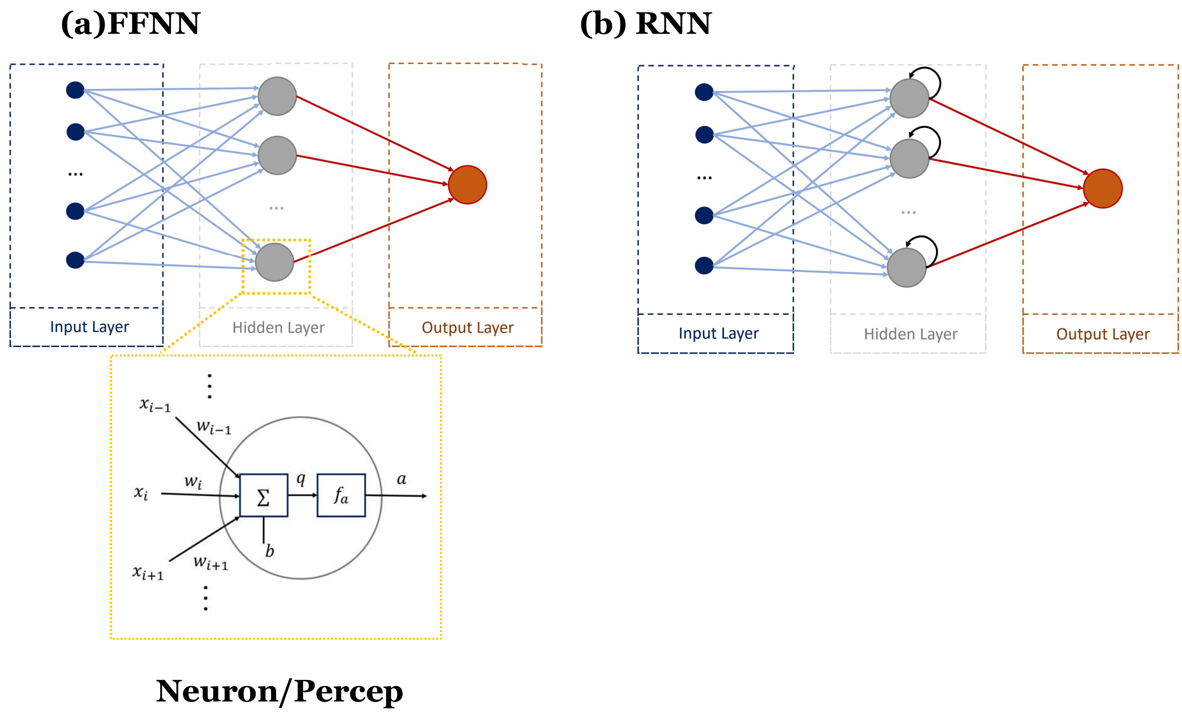

Fig. 10. A visual representation of (a) feed forward neural network and (b) recurrent neural network. The recurrent connections in the hidden layer allow information to persist from one input to another. The neurons are represented as circles [126].

One distinct characteristic of ANNs is their ability to learn from experience and examples to adapt to changing situations. They can be established automatically by training without the identification of model parameters and coefficients. However, they require a large amount of input data for training and verifying, and their accuracy is significantly affected by the training method and data. Moreover, the computational cost is still a bottleneck for large-scale applications of RUL prediction and the structure of an ANN plays an important role in its performance. Furthermore, the identification and optimisation of the model topology of ANN remains an open technical challenge. Generally, the structure is achieved through a time-consuming trial and error phase. Hu et al. [129] proposed to use novel genetic algorithm-based fuzzy C-means clustering technique to partition the training data sampled of a lithium-ion battery during the driving cycle-based test, and the clustering result was then applied to automatically learn the topology and antecedent parameters of the ANN model for battery state estimation.

4.2.1.3 Support vector machine (SVM)

SVM is a non-parametric ML technique based on kernels. A non-parametric model means that the number of parameters grows with the amount of training data. It has the advantages of being flexible and can model arbitrarily complex systems when providing enough data[130]. It performs classification by searching for the hyperplane separating classes of interest with a maximal margin. Kernel functions are often used in SVM to facilitate solving nonlinear problems, by transforming the nonlinear problem in a low-dimensional space into a linear problem in a higher dimensional feature space. Typically, the predictions are based on some functions defined over the input space, and learning is the process of inferring the parameters of this function. SVM makes predictions based on the function (5), as follows [131]:

$$y(x) = \sum_{n=1}^{N} \omega_n K(x, x_n) + \varepsilon \,, \tag{5}$$

where ω_n are the model weights connecting feature space to output, $K(\cdot)$ is a kernel function and ε is an independent noise term. Current health diagnostic and prognostic algorithms primarily use SVM as a regression tool for continuous values and are known as support vector regression (SVR). Regression is realized by searching for a minimum margin fit instead of a maximum margin classifier.

Based upon load collectives, Nuhic et al. [132] used SVR to learn the capacity degradation behaviour of a battery and then used the same estimation method to predict RUL. Measurements were carried out on Li-ion cells aged to different degrees to ensure a large amount of data for model training. Qin et al. [133] proposed an SVR model to capture capacity degradation. There, particle swarm optimization was used to optimize the kernel parameters of SVR, improving the RUL prediction. In order to improve the efficiency of training and prediction, Zhao et al. [134] took the feature vector selection to reduce data size by extracting two health indicators measurable online. An SVR model was utilized to capture the relationship between health indicators and capacity, resulting in reliable RUL prediction.

SVM is particularly appealing for its capability of handling small training datasets [135]. However, when the size of the training dataset increases, the number of support vectors increases accordingly. In order to improve the stability and robustness of SVR with large-scale training samples, decremental and incremental strategies [136,137] have been applied to integrate the relevant data sample for SVR training and ignore the irrelevant part. However, the computational cost is also increased by this procedure.

4.2.2 Probabilistic approach

Prognostic predictions need to cope with uncertainties coming from the measurement, the operation environment and the model itself – these arise from the structure of the model and uncertain parameters [110]. Probabilistic approaches use probability theory to express all forms of uncertainty, where probability distributions are used to represent all the uncertain unobserved quantities and their correlations with the data [138].

4.2.2.1 Gaussian process regression

Deriving from the Bayesian framework, GPR models have been widely applied to prognostic analysis as they are flexible, nonparametric and probabilistic [139]. GPR is a kernel based ML method, which can realize prognostics combined with prior knowledge based on a Bayesian framework and provide variance around its mean prediction to describe the associated uncertainty [120]. The Gaussian process can be seen as a collection of a limited number of random variables which have a joint multivariate Gaussian distribution [140]. Richardson et al. [122] applied a known parametric model to exploit prior information of capacity fade dynamics and then proposed three multi-output GPR models for RUL prediction by incorporating data from multiple batteries. In their later work [92], a GPR transition model was proposed to generate the underlying mapping between arbitrary current, voltage, temperature and capacity to predict the capacity degradation and battery RUL under dynamic conditions. However, the basic GPR method is unable to capture the local regeneration phenomenon during capacity degradation, where a battery shows a sudden and temporary incremental increase in capacity. To rectify this, Liu et al. [141] utilized a combination of covariance functions and mean functions in GPR for multi-step-ahead prognostics.

The performance of GPR is highly sensitive on the covariance function and the kernels should therefore be carefully selected to achieve high prediction accuracy [139]. The capacity fading process is complicated as it is influenced by many factors. The single covariance function would result in unreliable prediction for non-linear mapping with multidimensional input variables. Hence, it is recommended to construct an isotropic kernel with an advanced structure such as automatic relevance determination [142]. Note that an unsuitable optimization of hyper-parameters in the covariance function can result in over-fitting. To ameliorate this in GPR, one way is to minimize the negative log marginal likelihood [139].

4.2.2.2 *Relevance vector machine*

Relevance vector machine (RVM) was first introduced by Tipping [131] and is identical to SVM as shown in Eq. (5) but with a probabilistic approach. The RVM employs a Bayesian framework to infer the weights ω_n, with which the PDFs of the outputs instead of point estimates can be obtained. RVM provides performance comparable to SVM, while utilizing arbitrary kernel functions with high sparsity and also offering probabilistic predictions [143]. High sparsity means that a significant number of weights are zero, leading to more computationally efficient models.

Because of uncertainty representation, RVM is an effective approach for RUL prediction. Wang et al. [144] used RVM to derive the relevance vectors to represent the battery capacity fade, and predict capacity degradation values for the future cycles of relevance vectors. The uncertainties of the predicted degradation values are calculated and used to determine the parameters of a capacity degradation model. RUL estimations was achieved with the extrapolation of this model. To improve the long-term prediction performance of RVM, Liu et al. [145] proposed an incremental on-line learning strategy for RVM to inpromove the RUL prediction precision.

RVM provides good accuracy, high learning ability, sparsity, easy training process, and prediction result with probability distribution. However, one obvious drawback is that large datasets are required for training, leading to high time and memory demands.

Noteworthy, the computational complexity is on the order of N^3 , with N being the number of training samples [146].

5. Discussion

Numerous methods have been proposed for health diagnostics and prognostics of Li-ion cells. There is no single method to solve all current issues. A trade-off between the accuracy, computational effort and generalizability is usually required for each particular application. To better understand these trade-offs, this section summarizes and compares the characteristics of the existing data-driven methods. Based on their comparison, the challenges of the up-and-coming technologies based on data-driven battery health diagnostics and prognostics are discussed.

5.1Data-driven based battery health estimation

Accuracy and computational complexity are the main challenges for health diagnostics in real applications. Some of the benefits and drawbacks of each type of data-driven based approach are listed in Table 6, comparing ML and DA methods.

Table 6. Advantages and disadvantages of SOH estimation methods

Methods	Advantages	Disadvantages
Differential Analysis	• Easily implemented in a BMS; • Reasonable amount of literature available (mature technique); • Low computational effort.	• Requires controlled charging/discharging processes; • Temperature variation disturbs the estimation accuracy; • Requires noise filtering.
Machine Learning	• Good estimation accuracy; • Applicable in dynamic operating conditions; • No need of physical-based models.	• High computational effort; • Estimation accuracy is sensitive to the quantity and quality of training data.

ML provides higher estimation accuracy than DA as pointed out in [147] by comparing the IC analysis with two ML techniques (GPR and random forest regression). DA, especially IC/DV and DTV analysis, relies on the data measured during static (dis)charging which limits their usability. ML, on the other hand, can be used in a dynamic situation such as a driving cycle of an EV. Moreover, the temperature has a significant influence on the DA and will cause large bias, while ML can use temperature variations as the input features for model training and correlate it with ageing. However, the high computational effort required for ML methods is a major hurdle for their online application. In contrast, DA is easily implemented in a BMS by monitoring several cell parameters. A suitable SOH estimation method should be selected based on the application of the cells. When cells operate in moderate environmental conditions and in predictable patterns, for example in households, DA offers sufficient performance as the ageing trend can be captured with simple mathematical functions. For batteries under more complex operating conditions, such as in EVs, ML is a better solution due to its ability to approximate non-linear function surfaces.

5.2 Data-driven battery health prognostics

Methods for RUL prediction differ in their numerical complexity, prediction accuracy, and the ability to produce confidence intervals. Table 7 summarizes and compares the main characteristics of data-driven battery health prediction methods. Lifetime estimation models on the one hand and empirical ageing models with filtering on the other both rely on functions that capture the relations between battery capacity loss with its service time or the number of cycles. The prediction accuracy relies on the developed mathematical function. Note that lifetime estimation models belong to the open-loop type, developed offline using a large amount of ageing data collected in laboratory experiments. Such models have the advantage of low computational effort and ease of implementation in a BMS. Due to the ease of extracting model coefficients and the low computational effort, it is convenient to implement these models for online prediction. However, one major disadvantage is that they rely completely on the estimation accuracy of the developed model and do not include any recalibration mechanism. Additionally, the prediction accuracy is highly dependent on the amount of data used in their development. When the data is insufficient, extrapolating the fitted curve produces large errors. In contrast, empirical ageing models with filtering belong to the closed-loop type and can be recalibrated using battery characteristics in real-time. These models are designed to automatically achieve the desired output with the help of adaptive filters by comparing the estimated output with the actual measurement. The model parameters are updated during the operation to tune their predictions. However, these models construct functions by fitting the degraded capacity of the cell which restricts their applicability under more complex ageing conditions. Our recommendation is therefore hybrid approaches, for instance by combining these two types of models: a lifetime estimation model that could be constructed first based on a reasonable amount of experimental data and then, adaptive filters would be implemented to update the key parameters to provide more reliable prediction results. This seems a worthwhile and relatively unexplored path for future research. In this way, the model can be adapted to the real battery degradation conditions.

Battery health predictions based on ML methods do not assume any explicit mathematical model to describe the battery ageing behaviour and are mainly dependent on the quality of the historical test dataset. Non-probabilistic ML approaches can only provide an estimated point in regression. Ideally, however, the conditional distribution in order to capture the prediction uncertainty level, is a real challenge due to uncertainties from various sources such as measurements, state estimation, model inaccuracies, and future load uncertainty [120]. Probabilistic methods with an ability to yield PDFs, predict data points but also return confidence bounds around them. Because of this, probabilistic ML approaches are preferable as the estimated uncertainty can benefit battery users. However, the development of probabilistic ML methods is still in its infancy. Most existing studies test/validate their ML models on data obtained under the same conditions used for their training, which calls into question the robustness of these models in real applications where the operating conditions may vary significantly. It is therefore recommended to improve probabilistic techniques by training the models under complex ageing conditions. Additionally, the performance of these techniques is also highly sensitive to their structure and parameters. Suitable structure determination and parameter optimization strategies should also be explored to enhance their performance for future self-adaptive health or lifetime prediction.

Finding methods that can accurately predict the lifetime of batteries in an early stage is essential to accelerate the development, manufacture and optimization of emerging battery technologies. Interestingly, Severson et al. [148] have tackled the challenges using lasso and elastic-net regression approaches on a comprehensive training dataset that characterizes the performance of 124 commercial LFP/graphite cells aged under fast-charging conditions. The best regression model had correctly predicted cycle lives for 90.9% of the tested cells before any clear signal of capacity fade, within the first 100 cycles. Additionally, the developed classification model could classify cells as either having a short or a long lifetime based on the first five cycles of experimental data with test error of 4.9%. It is remarkable that this level of accuracy was achieved by analysing the discharging process from the experiments rather than by only considering capacity-fade data. This ML training approach is different from the conventional one. This work highlights the attractive applying ML techniques for lifespan prediction at early stages.

Table 7. A comparison of battery health prediction methods

			Advantages	Disadvantages
Analytical model with data fitting		**Semi-empirical model**	• Easy to be built up and quick to produce predictions; • Easy of extracting model parameters; • Low computational effort; • Easy to be implemented on BMS for online application by monitoring the parameter of cycling conditions, time and/or numbers.	• Extensive laboratory tests over the entire operating range are required, which are time consuming and economically costly; • Difficult to develop suitable laboratory ageing tests to analyse the interaction between different ageing processes and link them to lifetime expectancy on an experimental basis; • Poor generalizability. Developed models are restricted to a specific battery type and operating conditions.
		Empirical ageing model with filtering	• Only a small amount of ageing data is required for setting up the model; • Estimation errors are updated based on the real measurement.	• High computational effort and which increases the difficulties for online application.
ML	**Non-probabilistic**	**AR model**	• Simple structure; • Easy to identify parameters; • Easy to implement.	• Easy to cause under-fitting problems due to its linear regression type; • Poor generalization ability; • Bad long-term prediction ability.
		ANN	• Strong ability to consider nonlinearities; • RNN owns strong long-term RUL prediction ability due to recurrent links; • High prediction accuracy.	• Potential to cause over-fitting problem; • Poor uncertainty management ability; • Performance highly depends on the training process.
		SVM	• High prediction accuracy; • Non-parametric; • Robust to outliers; • Low prediction time.	• High computational cost; • Poor uncertainty management ability; • Requires cross validation procedure to determine hyper-parameters.
	Probabilistic	**GPR**	• Provide covariance to generate uncertainty level; • Non-parametric; • Being flexible.	• Performance is highly affected by kernel functions; • High computational cost.
		RVM	• Generate PDF directly; • Non-parametric; • Realize high sparsity; • Avoid cross validation process.	• Plenty of data is required for modelling; • Large time and memory consumed training process; • Easy to fall into the local optimization problem; • Potential to cause over-fitting problem.

5.3 Challenges and future developments

Although great efforts have been made in developing data-driven diagnostic and prognostic techniques, there are several major challenges in this field:

Ageing mechanism identification: Some of the purely data-driven methods, especially ML techniques, cannot provide in-depth information of the battery ageing mechanism. It is therefore desirable to find a way to combine the identification of ageing mechanism with online health estimation methods. As such, combining ML methods with physical mechanisms of degradation is indeed a promising direction for future research. As mentioned in Section 3.1, DA techniques, especially IC/DV and DTV, can reveal battery degradation mechanisms. It is therefore recommended to use DA under low current rates to uncover the ageing mechanism. Combining DA with ML is particularly promising as the former can help finding the most sensitive indicators for capacity loss which can then be used for ML SOH estimation or RUL prediction.

Self-improving models via online data: The degradation behaviour of Li-ion cells is sensitive to the operating conditions. It is still difficult to predict ageing under conditions different from the training dataset. The deviations between the laboratory conditions used to develop the models and real operating conditions limit the practical applicability of data-driven methods. This can be rectified in two ways: by improving experimental testing and by further algorithm development. When the size of the experimental dataset increases, covering the ageing information on a large range of operating conditions, the prediction capabilities of a data-driven approach increases accordingly. However, this also leads to a large experimental cost. On the other hand, improving the dynamic updating capability of the data-driven methods developed off-line is worth investigating further as it paves the way to self-improving models.

Health diagnosis and prognostics at module and pack level: Till now, most of the research on battery health diagnostics and prognostics has been done at cell level. However, in practice, these are generally connected in series and/or parallel to construct a battery pack for specific energy and power requirements. Understanding the ageing process of packs requires knowledge beyond the cell-level, considering additional impact factors, such as inconsistencies of cell characteristics, electrical imbalance and temperature gradients between cells [149]. All these issues complicate accurate ageing estimation and prediction models for packs. The advances in artificial intelligence and deep learning algorithms are foreseen to introduce some solutions to these problems. Deep neural networks are particularly suitable for highly complex non-linear fitting and can therefore achieve better accuracy for these problems. Several newly developed deep learning ANNs such as convolutional neural networks and generative adversarial networks have been successfully applied in the fields such as speech recognition [150] and image segmentation [151], owing to their strong self-learning abilities. However, to our knowledge, no attempts have been made so far to utilise them in batteries especially for module/pack-level health diagnostics. The use of ANNs or similar self-learning methods is also recommended for RUL prediction.

6. Conclusion

This article reviews data-driven technologies for battery health diagnostics and prognostics. Scientific literature covering the above topics is analysed, and each individual approach is discussed in view of its advantages and pitfalls. We provide an intuitive classification of the different strategies reported in the literature, and methods using differential analysis, analytical models, and machine learning are specially explored given the emerging interest on using them to assert more accurate models for Li-ion batteries lifespan.

We highlight that differential analysis methods can not only be used for battery health estimation but also for the fundamental identification of ageing mechanisms. They are generally computationally light and easy to implement, but also easily affected by the testing conditions such as temperature and current rate. Semi-empirical models can also be used for both health diagnostics and prognostics, but as they are open-loop in nature, their generalization ability is poor and therefore inflicting their performance when the battery is exposed to operating conditions different from those used to develop the model. Empirical ageing models with filtering can be recalibrated employing battery characteristics measured during operation. Machine learning methods are gaining increased attention for both health estimation and lifetime prediction problems, as they perform well in modelling highly nonlinear dynamic systems without assuming any mechanism a priori. Nevertheless, this increases the computational effort and their prediction accuracy is still highly limited by the adopted capacity fading model.

In a nutshell, the permanent reliable operation of a battery requires data-driven methods to be implemented in the battery management system for online application, but many corresponding technologies are immature. None of the methods is a one-size-fits-all solution; instead there are inherent trade-offs between complexity and the corresponding diagnostics and prognostics performance. Among all, the machine learning techniques, supported by a platform of open-source tools and data sharing, has the potential to revolutionize the battery health management system. We hope that this review provides useful reference points to support the design and operation of battery health diagnostics and prognostics systems, whilst informing the agenda of the battery research community at the same time.

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