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SURVEY PAPER

STATE-OF-ART SURVEY OF FRACTIONAL ORDER MODELING AND ESTIMATION METHODS FOR LITHIUM-ION BATTERIES

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This paper is dedicated to the memory of late Professor Wen Chen

Abstract

This paper presents a state-of-art survey of the research on fractional-order (FO) modeling with parameter identification, and FO estimation methods for state of charge (SOC), state of health (SOH), and remaining usage life (RUL) of lithium-ion batteries (LIBs) mainly in recent five years. FO electrochemical models and six different types of FO equivalent circuit models (ECMs) are introduced in detail. Then, the corresponding tuning algorithm for parameters of these FO models are also provided in brief. Moreover, FO estimation methods for SOC are listed and analyzed, mainly including FO observers, and FO Kalman filters (FO-KFs). SOH and RUL estimation is another vital aspect for LIBs ageing and degradation monitoring, thus FO estimation methods proposed in recent research within five years are all listed. Finally, some suggestions that may be helpful for further research are proposed in conclusion.

MSC 2010: Primary 26A33; Secondary 34A08, 60G22, 93A30, 93C95, 93E10, 93E12

Key Words and Phrases: fractional-order modeling; lithium-ion batteries; constant phase elements; state of charge; state of health; fractional-order Kalman filters

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1. Introduction

Battery is an emerging research aspect due to the increasing energy consumption in current applications, and batteries are the main energy storage device for several types of alternative energy, such as solar, wind, and hydroenergy. As Jeremy Rifkin said, in current third industrial revolution, three pivotal technologies: a communication internet, a renewable energy internet, and a mobility internet, all are connected to the Internet of Things (IoTs) [57]. Basically, batteries are everywhere to construct energy internet and ensure the energy supply for the other two internets in this big data era, so the design and control of batteries are the most concerning aspects for researchers around the world.

According to the charging and storage ways, batteries can be divided into four types: primary battery, secondary battery (rechargeable battery), fuel cell, and reserve battery. Since 21th century, people are pursuing more sustainable and portable batteries, thus lithium-ion batteries (LIBs) stand out among other kinds of batteries, such as lead-acid battery, Zinc battery, Nickel battery, and hydrogen-oxygen fuel cell. Besides, LIBs have high working voltage, high energy density, relatively low self-discharge, low maintenance and specific high current to applications, which make LIBs most widely used in applications [66]. However, LIBs have dynamic nonlinearity and ageing is always a concern. In Chemistry field, researchers are introducing new and enhanced chemical combinations to improve lithium-ion, while in electrical engineering field, researchers are aiming to learn more about LIBs, then monitoring and controling LIBs more accurately. This paper is mainly focused on the electrical aspect, that is, the modeling, and estimation methods for LIBs. Several performance index illustrating information of LIBs can be estimated, such as state of charge (SOC) [15], state of health (SOH) [83], remaining usage life (RUL) [38], degradation level or ageing level [69]. The well-known ampere-hour (Ah) integral method, and the open circuit voltage (OCV) measurement are proposed and commonly used for the estimation of SOC [10, 78, 94]. However, with growth spurt of mobile phones and electric vehicles (EVs), monitoring and management of LIBs in these appliances is faced with higher requirements, which stimulates various prominent modeling and estimation research [9, 35, 65, 85]. Among modeling and estimation research on LIBs, fractional calculus was firstly applied to present constant phase element (CPE), which starts a new fractional research era of LIBs.

Fractional calculus has been initiated more than 300 years, and started from mathematic definitions, that is, fractional derivate and fractional integral [29, 45]. Further extensions of fractional calculus, such as fractional-order (FO) state space model (SSM) [46], FO PID controller [52], fractional capacitor (CPE) [2], and fractional convection [30, 31], were gradually proposed and applied by electrical engineering researchers in recent 20 years [1, 6, 64]. As to fractional calculus for LIBs research, fractional modeling with identification and estimation of SOC, SOH, RUL are the two main aspects, which are the key monitoring points for further control and management of LIBs. The earliest FO battery research was designed for lead-acid battery about fractional system identification in 2006 [58], when lithium-ion battery has not been widely applied in EVs and mobile devices. After that, some research on fractional modeling of lead-acid battery have been proposed [12, 59]. With LIB gradually replacing lead-acid battery, fractional calculus was firstly applied to LIB for cell impedance analysis in 2007 [28]. Then fractional impedance analysis, fractional modeling, and fractional estimation methods for LIBs emerging [50, 95, 101], and Figure 1.1 shows the amount and research areas of published articles since 2006. The results are searched by fractional AND battery and fractional AND lithium battery in *Web of Science*, and are classified into LIB and Other Battery .

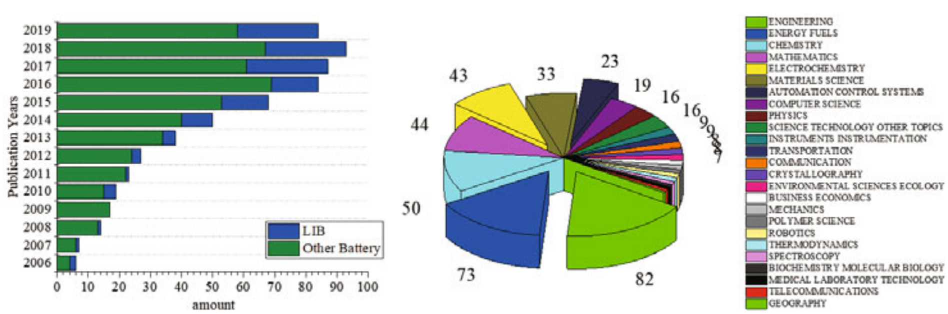

Fig. 1.1: Published articles and research areas of fractional AND battery since 2006 in *Web of Science*

From Figure 1.1, fractional research efforts on LIBs have developed rapidly in the past five years, and mainly focus on engineering, energy fuels, electrochemistry, and automation. As fractional research of LIBs is still at the beginning stage, it is worth drawing a literature review of these fractional research over the last five years. Although there are some existing early reviews introducing fractional techniques both on LIBs and supercapacitors (SCs) [4, 21, 77, 88], a specific and detailed analysis only for LIBs in recent five years is very necessary. Hence, this paper is written to present the novel fractional modeling and estimation methods for LIBs mainly between 2015 to 2019, and make an integration of these fresh research, then may provide some innovative suggestions for future fractional research on

LIBs. The rest of this paper is divided into four parts. Firstly, some necessary fundamental knowledge including SOC, SOH, RUL, and fractional basic definitions are introduced in the second part. Secondly, fractional modeling and corresponding identification methods are concluded and compared in the third part. Then, all fractional estimation methods of SOC, SOH, and RUL for LIBs in the past five years are provided in the fourth part, including fractional Kalman filter, fractional observer, online estimation, and so on. Finally, the last part provides current challenges and some suggestions for future work about fractional calculus applied to LIBs. It needs to be noted that all the modeling and estimation methods for LIBs mentioned in the following are referred to battery cells, rather than battery banks or battery packs in EVs.

2. Fundamental knowledge

This part is designed to provide some basic performance indexes of LIBs, like SOC, SOH, and RUL, also offer brief introduction to fractional calculus and fractional elements, like fractional Caputo definition, and constant phase element (CPE).

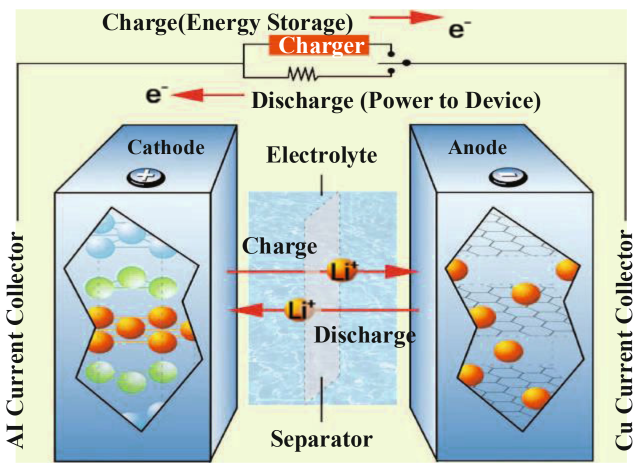

Fig. 2.1: Schematic of lithium-ion battery, which consists of four parts: negative electrode (anode), positive electrode (cathode), electrolyte, and separator (modified from [76]).

2.1. **Performance indexes of LIBs.** Figure 2.1 is a schematic of a LIB cell shown in [76], the cell includes four main elements: the positive electrode, negative electrode, electrolyte, and separator. During charging process, lithium ions are transported from cathode into electrolyte and then stored in anode, which builds up a potential difference between the positive and negative electrodes [76]. Discharging is based on the reversed process. The physical and chemical mechanism of a LIB cell can commonly be described by a two-dimensional (2-D) electrochemical model, that is, the Doyle-Fuller-Newman model, which is rarely used in real-time BMS due to the prohibitive computation [17]. Hence, single particle model (SPM) is derived by neglecting the electrolyte dynamics, so that the 2-D electrochemical model is simplified to one spatial dimension [56]. Due to the heterogeneity in the chemistry process, LIBs would show up some stochastic behaviors, like diffusion effect, leak current, and self-discharge, which can affect LIBs working states. To illustrate transient states of LIBs, several performance indexes have been defined. Three key indexes are introduced in this part, that is, state of charge (SOC), state of health (SOH), remaining usage life (RUL). Furtherly, fractional calculus is also applied to build the governing equations of a SPM for these reactions, specifically for the diffusion phenomena [49], and the details will be introduced in Section 3.1.

2.1.1. **State of charge.** SOC illustrates the remaining amount of available charge $Q(t)$ in a LIB, and cannot be directly measured. SOC can be expressed as the remaining percentage of a reference capacity Q_{ref} as follows [16],

$$SOC = \frac{Q(t)}{Q_{ref}} = \underbrace{\frac{Q(t_0)}{Q_{ref}}}_{SOC_0} + \underbrace{\frac{\int_{t_0}^{t} I(\tau)d\tau}{Q_{ref}}}_{SOC(t)} = SOC_0 + \quad SOC(t). \tag{2.1}$$

In practice, the relaxation period is too long thus it cannot obtain SOC_0 when LIB works in dynamic applications. Hence, how to measure real time SOC or design an estimator for SOC estimation is a key point of the battery management system (BMS) to prevent overcharge or overdischarge.

2.1.2. **State of health.** SOH is always considered with battery ageing and degradation process of LIBs, which can be affected by temperature, charging current, and discharge level [87]. SOH does not have the specific definition as SOC, but is usually defined as the ratio between remaining capacity and initial nominal capacity, as shown in the following [77],

$$SOH = \frac{C_p}{C_0}, \tag{2.2}$$

where C_p represents the available capacity, and C_0 represents the rated capacity specified per design and measured under the aforementioned conditions prior to operation. From equation (2.2), SOH decreases with fading capacity and rising ohmic resistance due to loss of active electrodes, solid electrolyte interphase (SEI), and irreversible lithium reactions [7].

There are four significant points for the ageing of battery indicated by SOH as listed below [47]:

(1) FL-BOL: First life beginning of life, serves as the reference for all the internal cell parameters monitored ($SOH = 100\%$).
(2) FL-EOL: First life end of life, represents the moment when the batteries are retired from the automotive use ($SOH = 80\%$) [70].
(3) SL-BOL: Second life beginning of life, represents the moment when the batteries are reimplemented on a second life applications.
(4) SL-EOL: Second life end of life, reproduces the moment of second life battery retirement.

2.1.3. **Remaining usage life.** RUL is the corresponding definition of SOH in aspect of battery ageing cycles amount. From Section 2.1.2, LIB needs to be replaced when reaching the FL-EOL point. Hence, there is a certain limited LIB cycles amount before the cycle life threshold, and RUL means the remaining cycles before this threshold. The RUL estimation is always companied with SOH estimation, and the estimation methods vary from internal resistance measurement of physics-based model, to data-driven prognostics [37]. The detailed introduction will be provided in the Section 4 of this paper.

2.2. Fractional calculus and fractional elements.

2.2.1. **Fractional calculus.** Fractional calculus is based on FO integrals and FO derivatives, also called non-integer-order integrals and derivatives [55], which can also be divided into left and right ones [32]. Since FO derivatives are applied to LIBs modeling and estimations rather than FO integrals, only left fractional derivatives are presented in the following. Basic fractional definitions commonly include Riemann-Liouville (R-L) definition, Grünwald-Letnikov (G-L) definition, Caputo definition [5], which may not equivalent to each other. Firstly, the three types of fractional derivatives are provided in the following. The R-L definition is expressed as [32]

$$ {}^{RL}D_t^{\alpha} f(t) = \frac{1}{\Gamma(n-\alpha)} \frac{\mathrm{d}^n}{\mathrm{d}t^n} \int_a^t \frac{f(\tau)}{(t-\tau)^{\alpha-n+1}} \mathrm{d}\tau, t > a, \tag{2.3} $$

where $f(t)$ is an arbitrary integrable function in $[a, b]$, $\alpha \in (n-1, n)$, ${}^{RL}D_t^{\alpha}$ represents the R-L type derivative operator, $\Gamma(\cdot)$ is the Gamma function. The G-L definition is expressed as [91]

$$ {}^{GL}D_t^{\alpha} f(t) = \lim_{h \to 0} h^{-\alpha} \sum_{j=0}^{[\frac{t-t_0}{h}]} (-1)^j \begin{pmatrix} \alpha \\ j \end{pmatrix} f(t-jh), \tag{2.4} $$

where ${}^{GL}D_t^\alpha$ represents the G-L type derivative operator, $[\frac{t-t_0}{h}]$ is the approximate recurrence term for integer part, and $\begin{pmatrix} \alpha \\ j \end{pmatrix} = \dfrac{\alpha!}{j!(\alpha - j)!}$ represents the coefficient of the recursive function. Caputo definition is another widely used one in engineering and control field due to the same initial conditions with integer-order (IO) derivative. The Caputo definition is expressed as [91]

$$ {}^{C}D_t^\alpha f(t) = \frac{1}{\Gamma(n-\alpha)} \int_a^t \frac{f^{(n)}(\tau)}{(t-\tau)^{\alpha-n+1}} \mathrm{d}\tau, t > a, \tag{2.5}$$

where ${}^{C}D_t^\alpha$ represents the Caputo type derivative operator. According to [32], if $f(t)$ is suitably smooth, i.e. $f \in C^n[a,b]$, then the R-L derivative and the G-L derivative are equivalent, that is, ${}^{RL}D_t^\alpha f(t) = {}^{GL}D_t^\alpha f(t)$; the R-L derivative and Caputo derivative have the following equation

$$ {}^{RL}D_t^\alpha f(t) = {}^{C}D_t^\alpha f(t) + \sum_{k=0}^{n-1} \frac{f^{(k)}(a)(t-a)^{k-\alpha}}{\Gamma(k+1-\alpha)}, \tag{2.6}$$

where $n-1 < \alpha < n$, $f \in C^{n-1}[a,t]$ and $f^{(n)}$ is integrable on $[a,t]$. However, for LIB research, G-L definition is easy to be discretized in time domain, thus it is most commonly applied to fractional time-domain models or fractional estimators for LIBs. Besides, the Laplace transform of Caputo definition under zero initial conditions is $\mathcal{L}{}_0^C D_t^\alpha f(t) = s^\alpha F(s)$, which is suitable for fractional research in frequency domain. Hence, Caputo definition and G-L definition are more widely used in LIB modeling and estimations. The reason is that the initial conditions of Riemann-Liouville definition have more complicated forms than that of Caputo definition as shown in equation (2.6). Besides, the computational load of FO derivatives are heavier than integer-order ones, so some numerical methods have been designed in [29, 32].

2.2.2. **Fractional elements.** Besides basic fractional calculus definitions, "fractor" is another essential element for modeling and estimation of LIBs. It is well-known that capacitors and inductors are not ideal ones in practical system, and the relationship between voltage and current is not just first-order derivative or integral, especially in low frequency or high frequency. Hence, the term "fractor" arose following the successful synthesis of a FO capacitor or an inductor, and the transfer function of fractor is given by [2],

$$ Z_F(s) = \frac{1}{F s^\alpha}, \tag{2.7}$$

where F is the impedance of the fractor, named as fractance . Fractor is also called constant phase element (CPE), and FO capacitor and fractional-order inductor are the two types of fractor in nature, respectively. Equation (2.8) shows the voltage-current relationship of a typical FO capacitor [91]:

$$\begin{cases} i(t) = C_w \frac{d^\alpha u(t)}{dt^\alpha}, \quad 0 < \alpha < 1, \ t \quad 0 \\ \frac{U(s)}{I(s)} = \frac{1}{C_w s^\alpha}, \end{cases} \tag{2.8}$$

where C_w is a constant related to the capacitance, and α represents the order of the FO derivative in Caputo definition. From equations (2.7) and (2.8), CPE was firstly proposed to replace the IO capacitors inside LIB models and explain the low-frequency dynamics of LIB. A similar fractional element is called Warburg element, which is a 0.5th order CPE in a typical Randles model for LIB [67]. In recent five years, Warburg element turns to be any fractional order instead of 1/2, same as CPE. Hence, CPE is the basis of FO electrical circuits for LIBs, which will be further introduced and discussed in the following sections.

3. **Fractional order modeling**

In BMS, it is always necessary to build a model with parameter identification before other estimations, monitoring, and charge or discharge control. Plenty of research has been published in the modeling and parameter identification aspects of LIBs [39, 80, 92]. As fractional calculus is extended from integer calculus [32], FO modeling and corresponding identification are also vital extensions. In an early survey of FO techniques applied to LIBs, lead-acid batteries, and SCs [101], four kinds of typical FO circuit models for LIBs have been offered. While in this section, a more complete set of FO models are presented, including some new research published in recent two years. Then the corresponding parameter identification methods for these LIB models are also provided in Section 3.3.

FO modeling of LIBs mainly can be divided into two aspects, that is, electrochemical model (equations) and equivalent circuit model (ECM). The main difference is that, ECM is applied electric components to build an equivalent circuit instead of electrochemical equations to re"ect the electrochemical reactions inside LIBs. Thermal and ageing models for LIBs are also investigated recent years due to the important roles in the kinetics of charge transfer process and side reactions introduced in Section 2.1 and shown in Figure 2.1, [49]. Figure 3.1 presents the available capacity of four 18650 LIBs (rated capacity = 2Ah) in five temperatures, that is, 15°C, 25°C, 30°C, 35°C, and 40°C. It illustrates that the available capacity of a battery cell varies much in different temperatures, especially in low temperatures which may happen in EVs battery pack during winter.

In this way, the model parameters and performance indexes estimations (SOC, SOH, RUL) would be in"uenced by the temperatures, so that FO thermal models is another necessary aspect for LIBs. While integer-order thermal and ageing models are also two vital aspects for modeling of LIBs, the extended FO ones are still very few and worthy of further investigation. Moreover, the existing research on FO thermal and ageing models are considered together with electrochemical model [49], and this paper is inclined to more electrical engineering aspects, so the thermal and ageing models are just brie"y discussed together with the FO electrochemical model in Section 3.1.

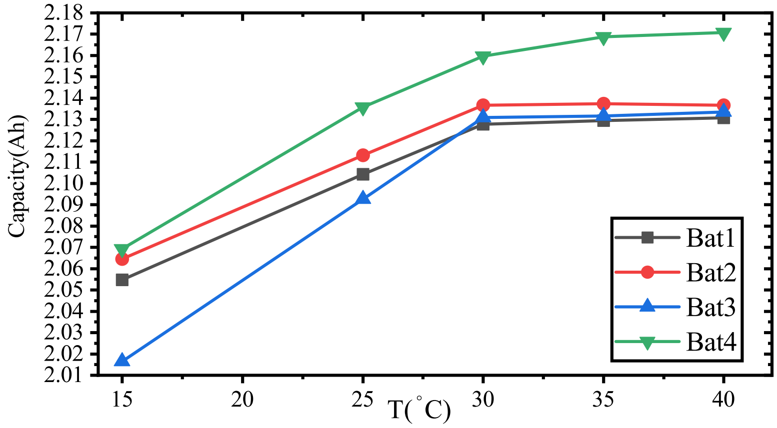

Fig. 3.1: The available capacity of four 18650 LIBs (rated capacity = 2Ah, named as Bat1, Bat2, Bat3, and Bat4) in five temperatures, that is, 15°C, 25°C, 30°C, 35°C, and 40°C.

3.1. **Fractional-order electrochemical model.** This type of FO model was firstly proposed by Sabatier et al. in [62]. The FO model is converted from a typical electrochemical model, called single particle model (SPM), which was built based on the electrochemical reactions inside a lithium-ion cell. A typical SPM generally includes four partial differential equations (PDEs) describing four key variables of the electrode and electrolyte, that is, lithium concentration c_{se} in the spherical particle by the diffusion law, lithium concentration c_e in electrolyte, charge conservation in electrode (electrode potential ϕ_s) by the Ohm s law, and charge conservation in electrolyte (electrolyte potential ϕ_e), respectively [63]. Then all of the four differential equations are linked by the Bulter-Volmer equation. As said in Section 2.1, a SPM is derived by neglecting the electrolyte dynamics and treating each electrode as a spherical particle that stores Li^+ as shown in Figure 3.2. From [49], FO electrochemical modeling for LIBs is based on the solution of Fick s first law of diffusion, and lithium ions concentration gradient in the particle can be described by the following:

$$\frac{\partial c_{se}}{\partial t} = \frac{D_{se}}{r^2}\frac{\partial}{\partial r}(r^2\frac{\partial c_{se}}{\partial r}) \begin{cases} \frac{\partial c_{se}}{\partial t}\big|_{r=0} = 0 \\ D_{se}\frac{\partial c_{se}}{\partial r}\big|_{r=R_s} = \frac{j^{Li}_{mean}}{a_s F} \end{cases} \tag{3.1}$$

where c_{se} is the lithium concentration, D_{se} is the diffusion coe cient, r is the radius of the sphere, and j^{Li}_{mean} is the average current density.

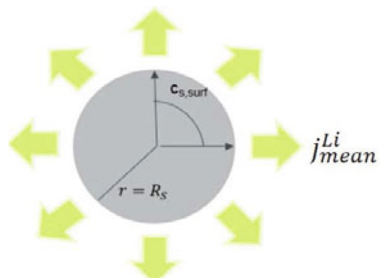

Fig. 3.2: Single particle model with concentration gradient through the sphere (modified from [49]).

The analytical solution of equation (3.1) is a transfer function, linking the mean value of the lithium current density in the electrode $J^{Li}_{mean}(s)$ to lithium concentration c_{se}. Based on the traditional SPM, Sabatier et al. have found that this transfer function can be approximated using the fractional transfer function [60]

$$H_{csi,e}(s) = \frac{c_{se}(s)}{J^{Li}_{mean}(s)} = \frac{K_{1i}(1+\frac{s}{\omega_{csei}})^{0.5}}{s}. \tag{3.2}$$

Based on equation (3.2) and we assume that the electrolyte potential is constant, a SPM can be simplified to a single-electrode model shown in Figure 3.3, if removing the negative electrode contribution [16, 61]. In this way, the FO electrochemical model has a concise structure without using large number of model parameters, but still holds the accuracy to re”ect electrochemical dynamics.

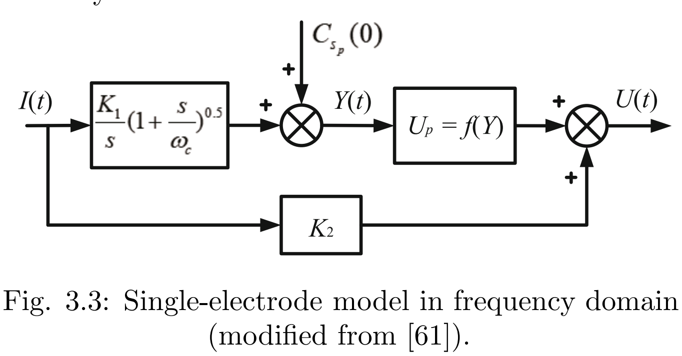

Fig. 3.3: Single-electrode model in frequency domain (modified from [61]).

Then, Sabatier et al. have published a new research related with this fractional-order electrochemical model in 2018 [49], which combined with an e cient simple thermal model and an ageing model designed for generation of fast charging algorithms. Like the FO electrochemical model, thermal and ageing models are presented in partial differential equation (PDE) forms. For example, the e cient thermal model is mainly based on a heat transfer equation as

$$mC_{\mathrm{p}}\frac{dT(t)}{dt} = Q_{\mathrm{gen}}(t) \quad Q_{\mathrm{loss}}(t). \tag{3.3}$$

where m is the mass of the cell, C_{p} is the specific heat capacity, Q_{gen} and Q_{loss} are the generated heat and convective exchanged heat with the environment, respectively. As to the ageing model, the structure is driven from the degradation caused by formation of solid electrolyte interphase (SEI) layer growth on the anode shown in Figure 2.1, and the detail PDEs can also be found in [49]. Similar fractional transfer function approximation has been applied to electrochemical equations in [34], and a simplified state space model of battery terminal voltage and load current was proposed and transferred into discrete form for further research. Since this type of FO model for LIB is a simplified one for SPM, it may also be useful to other kinds of enhanced SPM, like SPM with electrolyte dynamics (SPMe), SPM with electrolyte and thermal dynamics (SPMeT) [54]. Moreover, the FO thermal and ageing models still have lots to be explored and are worthy of further research. While the fractional calculus applied in the electrochemical models can better illustrate the dynamic reactions or thermal in" uences for LIBs, the FO PDEs of the FO electrochemical models remains computationally expensive for real-time BMS, which is also an aspect to be improved.

3.2. **Fractional-order equivalent circuit model.** As to the FO ECM, it is another electrical engineering way to analyze LIBs dynamics, while temperature and ageing are converted into parametric functions or SOC, SOH estimations [102]. Almost all ECMs structures were proposed according to the electrochemical impedance spectrum (EIS) of LIBs as shown in Figure 3.4, because the EIS reveals the electrochemical dynamics in frequency-domain and varies with SOC, SOH, RUL, temperature of LIBs. So various kinds of ECMs were proposed to explain the three main parts of the EIS, that is, high-frequency inductive tail, mid-frequency reaction, and low-frequency Warburg diffusion dynamics. Here four forms of ECMs are presented in Figure 3.5, and each form can be separated into certain types of ECMs in the following sections.

3.2.1. **FO Thevenin model** ($n = 1$). If n in Figure 3.5(a) is 1, ECM in Figure 3.5(a) becomes a FO Thevenin model, which is also called 1-RC model in traditional integer-order model. The state space model (SSM) of FO Thevenin model in Caputo definition was proposed and approximated into a discrete system in [102]. Since FO Thevenin model is already a simplified one and the approximation accuracy requires heavy computation, a data-based FO Thevenin model was built in continuous-time form [26]. From Figure 3.5(a) ($n = 1$), the FO Thevenin model is proved to be a simpler fractional ECM for LIB on the basis of the EIS and hybrid pulse power characteristic (HPPC) test [74]. As it only considers high-frequency and mid-frequency reactions, and ignores Warburg element of diffusion effects [51]. So FO Thevenin model is seldom applied in recent five years in fractional modeling. In comparison, FO Partnership for a New Generation of Vehicles (PNGV) and FO Randles model are the more widely used models.

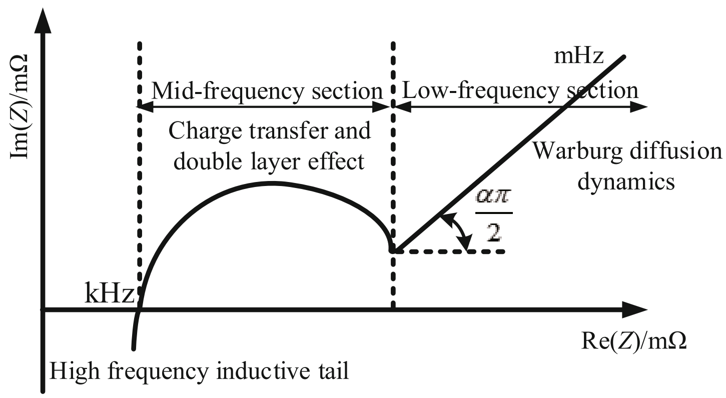

Fig. 3.4: A typical schematic diagram of EIS of LIB.

3.2.2. **FO PNGV and Randles model.** Figure 3.5(b) and Figure 3.5(c) are the FO Randles model and FO PNGV model, respectively. Both of them are systems with two fractional orders. Actually, traditional Randles model is already a FO system, because the Warburg element re"ecting diffusion dynamics was always considered as 0.5th order in the previous research. Since the diffusion effect varies due to different temperature, SOC, ageing level, the first step is changing the 0.5th order Warburg element into an arbitrary fractional order element, that is, another CPE [86]. Then, Wang et al. have found that the curve in mid-frequency shows a semi-ellipse rather than semi-circle due to capacitance dispersion, so the ideal capacitor re"ecting double layer effect was replaced by a CPE in [71], constructing the FO Randles model in Figure 3.5(b).

On the other side, FO PNGV model is also quite popular FO model for LIBs, and the corresponding state-space model (SSM) is denoted by the followings [36]

$$
\begin{aligned}
&\begin{bmatrix} \frac{d^m U_W}{dt^m} \\ \frac{d^n U_{cpe}}{dt^n} \end{bmatrix} = \begin{bmatrix} 0 & 0 \\ 0 & \frac{1}{R_{ct} C_{cpe}} \end{bmatrix} \begin{bmatrix} U_W \\ U_{cpe} \end{bmatrix} + \begin{bmatrix} \frac{1}{C_W} \\ \frac{1}{C_{cpe}} \end{bmatrix} I, \\
&U_0 = \begin{bmatrix} 1 & 1 \end{bmatrix} \begin{bmatrix} U_W \\ U_{cpe} \end{bmatrix} + R \;\; I + U,
\end{aligned} \tag{3.4}
$$

where U_W and U_{cpe} mean the voltages of Warburg element and CPE_1; C_W and C_{cpe} mean the capacitance of Warburg element and CPE_1; m and n mean the fractional order of Warburg element and CPE_1, respectively; I is the current of the FO PNGV model.

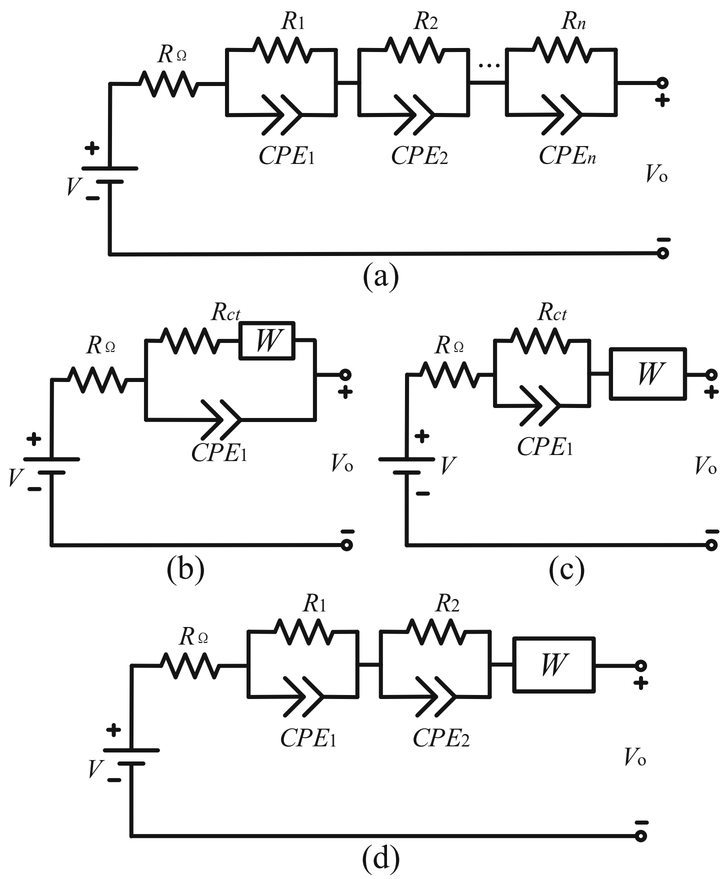

Fig. 3.5: Four forms of ECMs for LIBs, (a) n-RC^{α} Model, (b) FO Randles model, (c) FO PNGV model, (d) three-orders FO model ($CPE_i, i = 1, 2, ..., n$ and W are the fractional elements introduced in Section 2.2.2).

It can be known from equation (2.8) that FO PNGV model is an incommensurate order SSM, while some researchers would make $m = n$ to simplify into a commensurate order one. Xiao et al. have made a comparison among FO PNGV, FO Thevenin, IO PNGV, and IO Thevenin models [79]. The results prove that FO PNGV model can describe the OCV variation and low-frequency dynamics of LIB, and better capture the dynamic performance than the other three kinds of models. Moreover, it is interesting that a simplified FO Randles model analyzed in frequency domain [53] may have some connections with FO PNGV model. The authors of [53] separated the charge transfer process with diffusion dynamics [14], which turns out to be the structure of FO PNGV model. The Nernst diffusion phenomenon is firstly involved in [53], then simulated by FO integrator, which turns out to be a 0.5 order integrator. Hence, it seems that the simplified Randles models in [53] is a specific FO PNGV model.

3.2.3. **2-RC^α model** ($n = 2$). If in Figure 3.5(a) $n = 2$, ECM in Figure 3.5(a) becomes a FO system with two fractional orders. This type of FO model was proposed because the low-frequency part of EIS is proved to be a part of a depressed semicircle with a large diameter rather than a straight line, by certain dynamic tests, such as hybrid pulse power characteristic (HPPC) tests [84, 96]. Hence, the Warburg element is represented by the parallel combination of a CPE and a resistance, which is also called ZARC element [8]. The structure has little difference with FO PNGV model, but would bring more computation burden for further investigation, especially when the two fractional orders are incommensurate. However, more research used this 2-RC^α models recently [40, 72].

3.2.4. **High order FO model** (n 3). For higher modeling accuracy to fit EIS, some researchers have proposed extended high-order FO models for LIB. Hu et al. have improved the 2-RC integer-order model by adding low-frequency component and replacing ideal capacitor with CPE [22, 33], which results in a high-order FO model with three fractional orders α, β, γ as in Figure 3.5(d). Another type of high-order FO model with three fractional orders is based on the FO Randles model [98]. Similar to [84], a ZARC element was added to describe the high-frequency part of EIS, which intersects with the mid-frequency part. Moreover, Jacob et al. have proposed a general FO battery EIS model with the structure in Figure 3.5(a), which has n CPEs with n fractional orders [25]. The number of parallel tanks depends on the required accuracy, and the parallel resistance can be neglected to build a Warburg term, so that this general EIS model with n fractional orders holds high ”exibility.

3.2.5. **Variable and speci“c FO models.** Considering the fractional orders may change with several working factors (time, temperature, ageing), some researchers have proposed variable FO models for LIBs. Lu et al. constructed a monotonous relationship between SOC and the fractional order, which was considered re”ecting the fractal morphology of charge distribution [42]. Then, Lu et al. also applied the fractional order as an indicator for electrode ageing [41]. In this way, variable FO model provides a rapid method to estimate SOC and evaluate ageing level. However, the real-time identification for fractional orders of the variable FO model is a tough work in some practical application which may require online adaptive algorithm [8].

Despite those models cited above, some specific FO models can also provide novel explanation for LIBs, like the three types of FO impedance models involving bounded diffusion with three kinds of particle geometry [18]. Also Zhang et al. combined kinetic battery model (KiBaM) with ECM to build a hybrid FO model [89]. Considering the linear requirement of EIS test, Xiong et al. replaced charge transfer resistance by Bulter-Volmer (BV) equation, and ohmic resistance by a piecewise quadratic function of current, resulting in a BV-FOM [82].

3.3. **Parameters and fractional orders identi“cation.** Before all the FO models applied further to estimation or control, parameter identification is the first step to ensure the accuracy of FO models. Compared to integer-order models, the added fractional orders increase the identification di culty for the researchers, so they are also searching effective tuning methods. Here state-of-art tuning methods are introduced brie”y, and some suggestions are offered.

In earlier period of FO modeling, the Thevenin model was often applied because the corresponding identification methods were simpler, e.g. step response curve [60], algebraic calculation [90, 93], least squares (LS) method [27], and gradient method [14]. With more complicated FO modeling including two or three fractional orders, optimization algorithm and adaptive observer were increasingly designed [3, 13]. In the observer aspect, a Kreisselmeier-type adaptive observer has been proposed for FO Randles model [67, 68]. As for the optimization part, genetic algorithm (GA) and particle swarm optimization (PSO) are the two most applied algorithms for FO models among the large amount of optimal algorithm [71, 98].

In recent three years, it tends to design more enhanced algorithms for 2-RC^{α} and high-order FO models, like LS-GA (combination of the LS methods and GA) [84], and mixed-swarm-based cooperative particle swarm optimization (MCPSO) [22]. These long-name algorithms have complicated procedures and high cost, which may not be available in practical battery working situations. Investigating online algorithms like LS improved adaptive method in [74], or automatic updating methods, like the automatic updating of parameters value at different ageing stages in [34] are more desirable. Moreover, the analysis of specific algorithm and in" uencing factors, such as Bayesian inference [25], and the historical data dynamics [24], are also very necessary for other researchers to refer in their new FO modeling for LIBs.

4. **Fractional order estimations**

Either fractional modeling or parameters identification, the aims are building an accurate model of LIB for further monitoring or control. For battery monitoring, SOC, SOH, and RUL are three main performance indexes to indicate dynamic working states of LIBs. Based on different kinds of FO models, FO estimation methods for SOC, SOH, and RUL are also proposed in recent five years, which will be presented in this section.

4.1. **SOC estimation methods.** The traditional SOC estimation methods for LIBs generally include four aspects: basic methods (Ah and OCV methods), model-based observers, model-based Kalman filter (KF) series, and machine learning (ML). From equation (2.1), it is obvious that the Ah method is the simplest direct way to estimate SOC, its definition is presented as follows [11]:

$$SOC(t) = SOC_0 + \frac{\int_0^t \eta i(t)}{Q_N} dt, \tag{4.1}$$

where SOC_0 is the initial SOC value, Q_N is the rated battery capacity, and η is the charge-discharge e ciency. Ah method is simple but it depends on current measurement accuracy and has accumulated error. Thus, the monotonous relationship of OCV to SOC was proposed, however, OCV needs to be measured after long time rest of LIBs, which is not possible in some practical working situations. Hence, model-based methods are the main focus in recent research, such as sliding mode observer (SMO), Luenberger observer, and various types of KFs, while Ah and OCV methods are usually applied as SOC reference in current research.

By analyzing the published articles of SOC FO estimation for LIBs from 2014-2019, these methods can mainly be divided into four types: SMO and Luenberger observers, other observer, FO Kalman filter series, and special estimator, as shown in Figure 4.1(b). It is obvious that FO-KF series methods were investigated and proposed mostly. To better illustrate the state-of-art research distribution, four main aspects of traditional methods are also collected from *Web of Science* by searching lithium-ion battery , state of charge and corresponding key words, as shown in Figure 4.1(a). Since various kinds of FO ECMs listed in Section 3.2 have been applied to LIBs modeling, the traditional model-based estimation methods can be directly transferred to FO estimation methods. However, other fractional filters and fractional neural network were already proposed but still not applied, which may be the new direction of FO methods. In the following, we respectively present the methods shown in Figure 4.1(b).

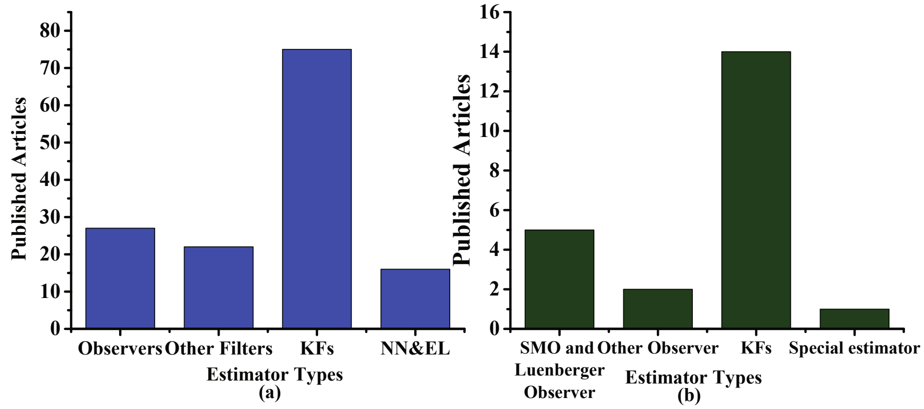

Fig. 4.1: Distribution of published articles about SOC estimation methods for LIBs, (a)Four main aspects of the traditional estimation methods: observers, other filters, KFs, and neural network (NN) & extreme learning (EL); (b)four main FO estimation methods: SMO and Luenberger observer, other observer, KFs, and special estimator.

4.1.1. **FO Luenberger observer and SMO.** The FO SMO was firstly proposed for SOC estimation of LIBs based on a 2-RC^{α} FO Model with commensurate fractional order α as shown in Figure 3.5(a)($n = 2$) [95]. Zhong et al. derived the differential equations for the 2-RC^{α} FO ECM under uncertainty $\delta_i, i = 1, 2, 3, C_e, C_d$ disturbance caused by nonlinear dynamics. Then, considering the fractional dynamic system [96],

$$D^{\sigma} X = f(X, I_{in}) + BW(t), \quad \sigma \quad (0, 1], \tag{4.2}$$

where X denotes state variable, $f(X, I_{in})$ is the system function, B and I_{in} are the constant matrix and the input vector, respectively. The proposed SMO is that

$$D^{\sigma} X = f(X, I_{in}) + L_i \mathrm{sgn}(X \quad X), \tag{4.3}$$

where X is the estimation for X, L_i is the SMO gain. Thus, the SMO in equation (4.3) was applied to the differential equation including SOC variable $S(t)$, to ensure the estimation error to approach zero. Zhong et al. have reduced SMO chattering by adjusting the SMO gain L_i, resulting an adaptive FO SMO [96], and also estimated the polarization voltage at the same time in a later article [97]. Similarly, a Luenberger observer was designed for a 2-RC^{α} FO Model with the same structure, but with incommensurate fractional orders α_1 and α_2 [72]. The structure of the Luenberger observer is also similar to equation (4.3) but without function sgn(·). Based on SMO and Luenberger observer, Zou et al. designed a nonlinear FO estimator for LIBs with FO PNGV model [100], and the nonlinear FO estimator has the form as follows:

$$\begin{aligned} D^{\alpha}x &= Ax + Bu + H_0(x,u) \\ &\qquad + L_l(y \quad y) + L_s\mathrm{sgn}(y \quad y), \\ y &= Cx + Du + f(x). \end{aligned} \tag{4.4}$$

The detailed parameters and variables definitions in equation (4.4) can be referred to [100]. From equation (4.4), the nonlinear FO estimator is actually the combination of SMO and Luenberger observer, and the original SSM of the FO PNGV model is

$$\begin{aligned} D^{\alpha}x(t) &= Ax(t) + Bu(t) + H_0(x,u), \\ y(t) &= Cx(t) + Du(t) + f(x_1(t)), \end{aligned} \tag{4.5}$$

where $x(t) = [SOC(t), V_{cpe}(t), V_W(t)]^T$, $y(t)$ is the output voltage V_o, and $f(x_1(t))$ is a nonlinear function related to SOC. It needs to be noted that equation (4.5) is a more useful SSM expression for FO PNGV model instead of SSM in equation (3.4), because SOC is inherently included in the state variables.

4.1.2. **FO Kalman “lters.** As the most commonly used kind of filters, FO-KFs for SOC estimation of LIBs includes several types of KFs, such as KF [44], extended KF (EKF) [36], unscented KF (UKF) [11, 73], cubature KF (CKF)[43], and dual KFs. All kinds of KFs are able to eliminate estimation error depending on the five basic equations including state time update, state measurement update, gain matrix update, time update of error covariance, and measurement update of error covariance [79]. While fractional-order KFs are still based on the five basic equations, the state update equations are in discrete forms with fractional differential. Since KFs are working in an iterative process with discrete forms, the original estimated FO continuous-time model needs to be discretized, which uses G-L definition in all research. Table 1 lists all types of FO-KFs applied to SOC estimation in recent five years, and the corresponding comments and analysis are provided in the following.

It needs to be noted that not all references of a type of FO-KF are listed in Table 1. Instead, a typical reference is chosen to be analyzed here, because the other references with same type are similar and listed in the references of this paper. The following comments and analysis are provided:

(1) FO-AEKF (FO Adaptive EKF) in [99] is able to change the process noise covariance matrices Q and measurement noise covariance matrices R with the estimation process, which is very intelligent and

ensures the high estimation accuracy in practical dynamic working situations.

(2) Dual FO-UKF in [8] is for SOC and fractional order estimation, dual FO-EKF in [23] is very e cient for SOC and SOH estimation at same time, and dual FO-KF in [33] is for SOC estimation and parameter identification. The dual structure can avoid applying other algorithm, reduce the system complexity, and improve the estimation e ciency.

(3) Dual FO-KF in [33] is also an adaptive KF (AKF) with the real-time parameter update, which is suitable for online SOC estimation.

(4) In all references, FO-KFs was compared with integer-order KF, EKF or UKF, resulting in faster converges speed and higher accuracy. The reason to choose FO-KF, FO-EKF, or FO-UKF needs to be further investigated, like for nonlinear dynamics, and a comparison between these FO-KFs may be necessary.

(5) Most of FO-KFs are based on FO ECMs, especially 2-$RC^{\alpha,\beta}$ model, so the design process may be similar. FO-KFs based on other kinds of FO models for LIBs can be discussed further, like the improve model by BV equation in [82], and the FO-AEKF designed in [34].

4.1.3. **Other FO observers and special estimator.** Another observer based on FO electrochemical model was proposed by Sabatier. et al. in [16] and [61]. It is the same research group introduced in the electrochemical model part of Section 3.1, and the electrochemical model is actually a FO model with 0.5th order. The SOC estimation was implemented by employing two error injection schemes, that is, input current feedback and

No.	FOKFs Types	References	Estimated FO-ECM
1	FO-KF	[40]	2-RC^{α_1,α_2} model
2	FO-EKF	[48]	2-$RC^{\alpha,\beta}$ model
3	FO-AEKF	[99]	2-$RC^{m,n}$ model
4	UKF	[51]	FO PNGV model
		[82]	BV-FO Thevenin model
5	Dual FO-KF	[33]	three-orders FO model
6	Dual FO-EKF	[23]	2-$RC^{\alpha,\beta}$ model
7	Dual FO-UKF	[8]	2-RC^{α_d,α_e} model
8	FO-CKF	[43]	FO Thevenin model

Table 1. All types of FO-KFs for SOC estimation in recent five years

SOC feedback [16]. The structure of SOC feedback observer is much like a Luenberger observer. Here a special SOC estimation method is presented, that is, a rapid SOC estimation by fractional order [42], which has proposed the fractional order can indicate SOC. The basic principle is finding the relationship between SOC and fractional order, and building a look-up table for the iterative estimating process to search.

From SOC estimation methods presented above, it still has many other aspects to investigate. From Figure 4.1, the model-based methods dependent on the FO ECMs is the mainstream of current SOC FO estimation research, however, machine learning (ML) is another developing aspect and has much to do in future, such as fractional-order neural network and FO version of extreme learning (EL) for SOC estimation of LIBs.

4.2. **SOH and RUL estimation methods.** SOH and RUL are connected with each other, so they are discussed together in this part. Since the degradation of LIBs are nonlinear dynamics, SOH and RUL estimation cannot be considered as linear measurements. Hence, fractional calculus provides a novel way to estimate SOH and RUL during the ageing process of LIBs. However, fractional-order SOH and RUL estimation methods proposed in recent five years are not as many as those for SOC. One reason is that SOH and RUL do not have a unique definition that is easy to be quantified, the second reason is that battery ageing problems is di cult to be described by fractional calculus, like end of life (EOL) and second-life reuse problems [75]. From Section 2.1.2, SOH can be estimated by remaining capacity, resistance, and related to lifetime, degradation level, RUL of LIB. Here a SOH index is shown as [19]

$$SOH_R = \frac{R_{EOL} \quad R_{current}}{R_{EOL} \quad R_{init}}, \tag{4.6}$$

where R_{EOL}, $R_{current}$, and R_{init} are resistance values at EOL, current status, and fresh status. Hence, the resistance of FO model can be estimated to calculate SOH, which may be related with EIS measurement of LIB. Similarly, RUL, degradation, and ageing level can also be investigated by the resistance or even the fractional order [41, 65]. Table 2 lists all published articles related with FO estimation methods for SOH, RUL, degradation, lifetime, and battery ageing. Some brief explanations and analysis are listed in the following.

(1) The investigated indexes in Table 2 are SOH (No.1 & No.2), RUL (No.3), degradation (No.4 & No.5), and ageing level (No.6 & No.7), respectively.
(2) Incremental capacity analysis (ICA) in [70] was designed to recognize ageing mechanism and estimate SOH in real time.

(3) Overall impedance in [20] means the diameter of the semicircular part (in the mid-frequency) of EIS, which would increase due to the increase of charge transfer resistance.
(4) SEI (solid electrolyte interface) resistance was observed in [81], and it shows a linear relationship with the remaining capacity, which can indicate the degradation behavior.
(5) Fractional order was considered as an indicator of ageing level and SOH in [41], which is the research from the same group that has illustrated fractional order can also indicate SOC linearly in [42].
(6) In [13], Sutter et al. provided both ohmic resistance and charge transfer resistance evolution over lifetime.
(7) In [65], the fractional order α_ω was considered as an indicator of ageing level just like that in [41], but the difference is that the variable fractional order α_ω was investigated with the on-line capacitive resistance arc of EIS in frequency domain, which is similar to that in [20]. Hence, the fractional order α_ω was verified to be a meter of states and performance of LIBs, like SOH, RUL, etc.
(8) Although the SOH estimation, RUL estimation, degradation, and ageing problems of LIBs are related with each other, all the No.1-No.6 methods in Table 2 were designed specifically for the certain type of index. Hence, all the six kinds of methods would be extended for the other indexes, or for the combining estimation of these indexes. As to the seventh method in [65], it only indicates that fractional order α_ω is relevant with SOC, ageing level and discharge rate of LIBs, but did not provide specific estimation method for ageing level. Thus the findings in [65] remains open to more practical applications.

No.	References	FO-ECMs	Methods
1	[23]	2-$RC^{\alpha,\beta}$ model	Dual FO-EKF
2	[70]	2-$RC^{\alpha,\beta}$ model	incremental capacity analysis
3	[20]	FO PNGV model	overall impedance
4	[81]	2-$RC^{\alpha,\beta}$ model	SEI resistance
5	[41]	Warburg model	fractional order
6	[13]	FO Thevenin model	resistance
7	[65]	EIS	fractional order

Table 2. SOH and RUL estimation methods (2014-2019)

5. **Conclusion and future work**

This paper presents a state-of-art survey on fractional-order modeling and estimation methods for LIBs mainly in recent five years. FO electrochemical models and ECMs are the main model forms for LIBs, and a very detail presentation and analysis of six types of ECMs in Figure 3.5 are provided in Section 3.2. Moreover, all the FO estimation methods for SOC, SOH, and RUL of LIBs are introduced and analyzed in Section 4. FO observers and FO-KFs are the two main methods applied to SOC estimation, and eight kinds of FO-KFs are listed in Table 1 with brief comments and analysis. While SOH and RUL estimation methods listed in Table 2 are not as many as those for SOC estimation, there is still a lot to be investigated for fractional calculus in LIBs lifetime research. The following are some suggestions that may be helpful in future work.

(1) Online and real-time monitoring and estimation are the new trends for future BMS, thus FO modeling, parameter identification, and estimation methods that can work online are interesting for LIBs.
(2) Adaptive methods are required for further research, including adaptive parameters of the FO identification algorithm, the weights update of the FO iterative process during estimation, and the updates over the LIBs ageing.
(3) Other technologies may be combined with current models and estimation methods. FO filters except FO-KFs, FONN and FO-EL may be applied for parameter identification and SOC estimation.
(4) SOH, RUL, ageing, and degradation problems are lack of investigation by using fractional calculus, and the integer-order aspects already have lots results that can be referred from [77].

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