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Review article

A review of fractional-order techniques applied to lithium-ion batteries, lead-acid batteries, and supercapacitors

Changfu Zou[a,1], Lei Zhang[b,1], Xiaosong Hu[c,*], Zhenpo Wang[b], Torsten Wik[a], Michael Pecht[d]

[a] Department of Electrical Engineering, Chalmers University of Technology, Gothenburg 41296, Sweden
[b] National Engineering Laboratory for Electric Vehicles, Beijing Institute of Technology, Beijing 100081, China
[c] State Key Laboratory of Mechanical Transmissions, Chongqing University, Chongqing 400044, China
[d] Center for Advanced Life Cycle Engineering, University of Maryland, College Park, MD 20742, USA

HIGHLIGHTS

- Fractional-order techniques applied to electrochemical energy storage are reviewed.
- Mathematical fundamentals of fractional-order calculus are elucidated.
- Available fractional-order battery/supercapacitor models are characterized.
- Case studies are performed to quantitatively analyze the efficacy of different models.
- A research outlook for the modeling methodology and applications has been discussed.

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ABSTRACT

Electrochemical energy storage systems play an important role in diverse applications, such as electrified transportation and integration of renewable energy with the electrical grid. To facilitate model-based management for extracting full system potentials, proper mathematical models are imperative. Due to extra degrees of freedom brought by differentiation derivatives, fractional-order models may be able to better describe the dynamic behaviors of electrochemical systems. This paper provides a critical overview of fractional-order techniques for managing lithium-ion batteries, lead-acid batteries, and supercapacitors. Starting with the basic concepts and technical tools from fractional-order calculus, the modeling principles for these energy systems are presented by identifying disperse dynamic processes and using electrochemical impedance spectroscopy. Available battery/supercapacitor models are comprehensively reviewed, and the advantages of fractional types are discussed. Two case studies demonstrate the accuracy and computational efficiency of fractional-order models. These models offer 15–30% higher accuracy than their integer-order analogues, but have reasonable complexity. Consequently, fractional-order models can be good candidates for the development of advanced battery/supercapacitor management systems. Finally, the main technical challenges facing electrochemical energy storage system modeling, state estimation, and control in the fractional-order domain, as well as future research directions, are highlighted.

1. Introduction

Transportation electrification and grid integration of renewable energy sources constitute two renewed research efforts to reduce dependence on fossil fuels and mitigate global warming [1]. Market penetration of electrified vehicles (EVs) can help meet these goals if it is coupled with decarbonized electricity, for example, solar and wind power [2]. Electrochemical energy storage systems (EESSs) play a critical role in both EVs and renewable energy integration applications. They serve as energy sources to provide power supply and/or energy buffers to improve efficiency and the overall economy.

Rechargeable batteries and supercapacitors are typical EESSs that share a similar structure–both of them store and convert energy through diffusion and migration of ions. Each battery or supercapacitor cell is composed of positive and negative electrodes separated by an enabling separator that allows ion transfer but prevents electron conduction.

* Corresponding author.
E-mail addresses: changfu.zou@chalmers.se (C. Zou), xiaosonghu@ieee.org (X. Hu).
[1] Equally contributed to this work.

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Electrodes and their separators are often immersed in an electrolyte solution that contains mobile ionic species [3]. Among a number of different energy storage technologies, lithium-ion (Li-ion) batteries have currently been accepted as the leading candidate for commercial EESSs because of their superiority, especially in volumetric and gravimetric energy densities [4,5]. However, each EESS has unique features and characteristics, and may be well suited for particular applications. For example, while lead-acid batteries are primarily used in cases where cost, reliability, and abuse tolerance are crucial [6], supercapacitors are preferred in devices that require high power density and long cycling lifetime [7].

EESSs must be safe and highly tolerant of high/low temperatures. They must also be cost-effective and provide large energy/power density and long cycle life. To pursue these objectives, model-based state estimation/monitoring techniques and energy management schemes have been extensively studied in the literature, e.g., [8,9]. A common requirement of these tasks is to construct accurate yet simple mathematical models that are adaptable to thermal and aging phenomena inherent in EESSs.

Considerable research efforts have been dedicated to mathematically modeling EESS dynamics and have resulted in physics-based, equivalent circuit, and data-driven models [10–12]. Based on differentiation orders, these models can generally be assorted into integer- and fractional-order models. Integer-order models dominate the research and engineering application of electrochemical energy storage. Hu et al. [13] compared commonly used equivalent circuit models of Li-ion batteries in terms of accuracy, complexity, and robustness under vehicle driving cycles. Doyle et al. [14] and Zou et al. [15] formulated electrochemical models governed by a set of partial or ordinary differential equations for Li-ion cells. Zhang et al. [16] and Drummond et al. [17] discussed electrochemical models for supercapacitors. However, it has been incrementally recognized, such as by Freeborn et al. [18], that EESSs exhibit some mathematical characteristics in accordance with fractional-order systems. This fact solicits increased interest and endeavors to come up with novel EESS models in the domain of fractional calculus. As a result, the fractional-order modeling methodology may not only improve prediction accuracy but also preserve some physical meanings underlying model parameters.

This paper provides a comprehensive review of fractional-order techniques for typical EESSs, including Li-ion batteries, lead-acid batteries, and supercapacitors. Section 2 presents the mathematical fundamentals of fractional-order calculus. Section 3 introduces the common dynamic processes of EESSs and electrochemical impedance spectroscopy to elucidate the principles of fractional-order modeling. Available battery/supercapacitor models are sequentially surveyed, grouped, and characterized. After analyzing parameter identification techniques in Section 4, the accuracy and computational requirement of fractional-order models (FOMs) are quantitatively investigated via case studies in Section 5. Section 6 highlights the main technical challenges facing FOM-based management for EESSs, including modeling of coupled system dynamics, state estimation, and charge/discharge control, followed by concluding summaries in Section 7.

2. Mathematical fundamentals

This section exhibits the mathematical fundamentals of fractional-order calculus (FOC) to facilitate the understanding of concepts and technical tools used for modeling electrochemical energy systems. In particular, the definitions of impedance and fractional-order derivatives and the FOM's state-space representation will be discussed. A thorough exposition of FOC can be found in textbooks on fractional-order system modeling, analysis, and applications [19–21] and related survey articles [22,23].

Frequency-domain electric impedance. In the frequency domain, a general impedance, Z, in electrical circuits may be defined by the following proportional relation

$$Z \propto (j\omega)^{\alpha}, \quad \text{for } \alpha \in [-1,1],\ \omega \in \mathbb{R}, \tag{1}$$

where j is the imaginary number and ω is the radian frequency. The conventional equivalent circuit elements, including pure capacitors, resistors, and inductors, are special cases of Z, corresponding to $\alpha = 1$, 0, and -1, respectively.

As initially proposed by Cole and Cole [24], a fractional-order capacitive element can be characterized by the impedance in (1) as

$$Z_{\mathrm{CPE}} = \frac{1}{C_{\alpha}(j\omega)^{\alpha}}, \quad \text{for } \alpha \in (0,1), \tag{2}$$

where the exponent α is a fractional-order and C_{α} is a constant and is called a pseudo-capacitance with the dimension $\mathrm{F{\cdot}sec^{\alpha-1}}$ [25]. Z_{CPE} has a constant-phase angle at $\alpha\pi/2$ [26] and is often called a constant phase element (CPE). In comparison, the phase shift for pure capacitors is $\pi/2$.

Fractional-order derivatives. The fractional-order operator for the CPE in (2) is mathematically defined by ${}_0\mathfrak{D}_t^{\alpha}(\cdot) = d^{\alpha}(\cdot)/dt^{\alpha}$. An equation with ${}_0\mathfrak{D}_t^{\alpha}$ describes dynamic processes with infinite dimension. To facilitate analysis and numerical implementation, three different definitions, namely, Riemann-Liouville (RL), Caputo, and Grünwald-Letnikov (GL) fractional derivatives are often utilized for such an operator [19]. For instance, the GL fractional derivative takes explicitly the form

$${}_0\mathfrak{D}_t^{\alpha} f(t) = \lim_{T\to 0} \frac{1}{T^{\alpha}} \sum_{k=0}^{t/T} (-1)^k \left\langle \alpha,\ k \right\rangle f(t - kT), \tag{3}$$

where T is the sampling time interval, t/T is the maximum integer lower than t/T, and $\langle \alpha,\ k \rangle$ represents the Newton binomial term defined as

$$\left\langle \alpha,\ k \right\rangle = \frac{\Gamma(\alpha + 1)}{\Gamma(k + 1)\cdot\Gamma(\alpha - k + 1)}, \tag{4}$$

where Γ() is the gamma function with the definition of

$$\Gamma(\alpha) = \int_0^{\infty} \xi^{\alpha-1} e^{-\xi} d\xi. \tag{5}$$

For simplicity, ${}_0\mathfrak{D}_t^{\alpha} f(t)$ is written as $\mathfrak{D}^{\alpha} f(t)$ in the sequel. A comprehensive description of these definitions as well as their peculiarities has been presented in Refs. [19,27].

Unlike their integer alternatives, fractional derivatives are not local operators because they take into account the entire past trajectory of $f(\cdot)$ over the interval $[0,\ t]$, as seen in (3). This is the so-called *long memory property* of fractional derivatives. However, this property significantly increases the computational burden for engineering applications of FOMs, particularly for real-time model-based optimization and control. To improve implementation efficiency, a short memory principle was therefore proposed by Podlubny [28] to approximate (3) with high-order difference equations, which consider only recently past information in the state propagation. This approach has been shown to be effective in a number of examples in fractional-order modeling of Li-ion batteries [29] and supercapacitors [30]. Indeed, there is in general a trade-off between modeling accuracy and computational complexity around the memory length.

System representation and types. The state-space representation of a general fractional-order system can be written in the following form

$$\mathfrak{D}^{\alpha} x(t) = f(t, x(t), u(t)), \tag{6a}$$

$$y(t) = h(t, x(t), u(t)), \tag{6b}$$

where $x := [x_1, \cdots, x_n]$ is the state vector, $\mathfrak{D}^{\alpha} x := [\mathfrak{D}^{\alpha_1} x_1, \cdots, \mathfrak{D}^{\alpha_n} x_n]$, and u, y are separately the system input and output vectors. This representation is the same as integer-order system representations except for the fractional derivative on the left-hand side of (6a). If $\alpha_1, \cdots, \alpha_n$ are all positive integer multiples of a real number γ, then (6) is a commensurate fractional-order system of order γ; otherwise, it is said to be incommensurate, with more degrees of freedom to fit system dynamics [19,31,32].