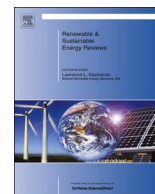




Contents lists available at ScienceDirect

Renewable and Sustainable Energy Reviews

journal homepage: www.elsevier.com/locate/rser

A review of supercapacitor modeling, estimation, and applications: A control/management perspective

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ARTICLE INFO

Keywords:

Energy storage
Supercapacitor
Modeling
State estimation
Industrial applications

ABSTRACT

Supercapacitors (SCs) have high power density and exceptional durability. Progress has been made in their materials and chemistries, while extensive research has been carried out to address challenges of SC management. The potential engineering applications of SCs are being continually explored. This paper presents a review of SC modeling, state estimation, and industrial applications reported in the literature, with the overarching goal to summarize recent research progress and stimulate innovative thoughts for SC control/management. For SC modeling, the state-of-the-art models for electrical, self-discharge, and thermal behaviors are systematically reviewed, where electrochemical, equivalent circuit, intelligent, and fractional-order models for electrical behavior simulation are highlighted. For SC state estimation, methods for State-of-Charge (SOC) estimation and State-of-Health (SOH) monitoring are covered, together with an underlying analysis of aging mechanism and its influencing factors. Finally, a wide range of potential SC applications is summarized. Particularly, co-working with high energy-density devices constitutes hybrid energy storage for renewable energy systems and electric vehicles (EVs), sufficiently reaping synergistic benefits of multiple energy-storage units.

1. Introduction

Energy storage systems play an important role in a diverse range of industrial applications [1,2], as either bulk energy storage or distributed transient energy buffer. Specific energy, specific power, lifetime, reliability, and safety are among the main criteria considered when picking energy storage [3]. Rechargeable batteries, especially lithium-ion batteries, are currently a popular option due to their high energy density and acceptable cycle life [4]. Nevertheless, they have limits of relatively low power density and relatively high internal resistance that could heavily curtail their power-delivery capability under large current loading. Moreover, battery life is highly susceptible to high current-rate and transient loading conditions [5]. In order to overcome these shortcomings, redundant design is often adopted in practice for pulse and peak power fulfillment, which inevitably incurs additional expense. On the other hand, supercapacitors (SCs), also known as ultracapacitors (UCs) or Electric Double-Layer Capacitors (EDLCs), are being

actively studied and unanimously envisaged as a promising energy storage technology, owing to their desirable merits including high power density and high degree of recyclability [6,7]. They have additional advantages, such as low internal resistance, wide operating temperature window, and high efficiency, despite that they have relatively low energy density [8]. These advantageous characteristics render them particularly suitable for working independently or in tandem with other high-energy devices for power sinking/sourcing in real plants [9].

In order to ensure efficient, safe, and reliable operation of SC systems, an enabling management system is necessary [10]. Its main tasks include cell equalization management, thermal management, power control synthesis, safety supervision, and so on, all of which hinge on systems and control engineering. For example, accurate and efficient modeling is fundamental for management system development regarding electrical, thermal, and aging issues [11]. Besides, precise state estimation provides insights for cell non-uniformity

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Received 11 June 2016; Received in revised form 6 February 2017; Accepted 29 May 2017

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suppression and optimal power control of SC systems. There is a large body of literature reporting on advances in SC modeling, state estimation, and their industrial applications. This paper provides a comprehensive survey of SC studies in the recent literature, with the primary objective to systematically summarize the state of the art in SC modeling, state estimation, and industrial applications from a control/management perspective. Three prominent contributions distinguish our endeavor from existing review articles [12–15]. First, we review virtually all the modeling approaches applied to SCs, including electrochemical, equivalent circuit, intelligent, and fractional-order models, especially underscoring the most recent modeling outcomes. Second, we cover the latest literature on State-of-Charge (SOC) estimation and State-of-Health (SOH) monitoring, and highlight the influencing factors that impact SC health. Third, we elucidate a broad variety of SC applications, especially focusing on the energy management development of SC-involved systems. This paper is anticipated to manifest research progress in the subject of SC management, catalyze novel transformative modeling/control ideas, and unlock more application opportunities.

The remainder of the paper is structured as follows: Section 2 gives a short overview of SC fundamentals. Section 3 reviews SC modeling approaches. The SOC estimation and SOH monitoring techniques are summarized in Section 4, followed by introducing SC industrial applications in Section 5. Section 6 summarizes the key points of this paper.

2. SC fundamentals

The electric double-layer (EDL) phenomenon was firstly described by Helmholtz in 1853, and patented by Becker (General Electric Company) in 1957, who used porous carbon material with high specific area as electrodes for double-layer structure formation [16]. Nippon Electric Company (or NEC) licensed a SC product as a memory backup device that marked the first commercial application in 1971 [17]. Structurally, the SC consists of two electrodes, a membrane separator, and electrolyte as shown in Fig. 1, which was also illustrated in our previous study [18].

The two electrodes are insulated by the membrane separator and impregnated to the electrolyte. The membrane separator only permits the ion mobility but prevents electric contact. SCs store electrical energy mainly through the formation of the double-layer capacitor structure at the interface between the electrodes and the electrolyte. This energy storage mechanism involves no chemical phase or composition changes, apart from fast and reversible Faradaic reactions existing on the electrode surface, which also contribute to the total capacitance. The characteristic of electrostatic charge transfer results in a high degree of recyclability [19]. Compared to conventional capacitors, the high capacitance of SCs originates from the high specific area of the electrodes, which is largely determined by the used electrode materials and their physical properties (e.g. conductivity and porosity). Advanced electrode materials have been the area of intensive study, and the latest progress has been periodically reviewed in [20,21]. Carbon materials with high specific area, conducting polymers, and metal oxides constitute the main categories for SC electrode materials [22]. Particularly, carbon materials have been successfully utilized in the commercially available SCs because of their advantages such as low cost, high specific area, availability, good conductivity, high electrochemical stability, and wide operating temperature window [23]. The porosity parameters, including pore size and pore-size distribution, equally exert an important influence on the practical SC capacitance, because these parameters can have a major impact on the active electrode surface accessible to the electrolyte. For example, Largeot et al. [24] pointed out that the capacitance culminates when electrodes have the pore size close to the ion size of the electrolyte. Electrolyte is another important component that affects SC performance. The general requirements for the electrolyte encompass large voltage window, high

ionic concentration, high electrochemical stability, low resistivity, low viscosity, low volatility, and low cost [25]. Aqueous electrolyte, organic electrolyte, and ionic liquids are mainly used, each with its own strengths and limitations. Generally, SCs with aqueous electrolyte exhibit better performance in terms of capacitance and power delivery, since the aqueous electrolyte can have higher ionic concentration and lower resulting resistance. However, the voltage window of aqueous electrolyte is as low as about 1.2 V, which significantly hinders the improvements of energy and power density, since the SC energy is proportional to the square of the voltage. In contrast, the organic electrolyte can offer a voltage window as high as 3.5 V, making it more preferable in SC manufacturing. Ionic liquid refers to the smelt salt at certain temperature which possesses several desirable properties, including low vapor pressure, large voltage window, high electrochemical stability, and so forth [26].

3. SC modeling

A common framework for describing and analyzing systems is always required by researchers and engineers. This framework is often mathematics, and referred to as mathematical modeling. For SC systems, modeling is essential for design prediction, condition monitoring, and control synthesis. Since a model is, at best, a surrogate for real systems, whose accuracy is subject to the assumptions and requirements, it must be generated for a specific purpose. As such, numerous SC models have been reported in the literature for different purposes, including capturing electrical behavior, thermal behavior, self-discharge, aging simulation, *etc.* For electrical behavior modeling, electrochemical models, equivalent circuit models, and fractional-order models are the most commonly used models. Generally, electrochemical models have high accuracy but low calculation efficiency, since they are able to capture the real reaction process inside UCs at the expense of coupled partial differential equations (PDEs). This hinders their applications in embedded systems for real-time energy management and control. In contrast, equivalent circuit models are derived from empirical experience and experimental data under certain conditions. This renders them inadequate for representing the UC dynamics under wide-range conditions, thus giving rise to model mismatch issues. Also, their parameters and states lack physical representations so that no internal information is explicitly available. However, the structural simplicity and decent modeling accuracy make them well-accepted for real-time energy management synthesis. The comprehensive SC models for control/management purposes reviewed in this paper are given in Fig. 2.

3.1. Electrochemical models

Helmholtz [27] discovered the EDL phenomenon and described it using a model where all the charges were assumed to be adsorbed at the electrode surface. This is identical to a conventional dielectric capacitor structure [28]. Gouy [29] and Chapman [30] further modified the Helmholtz model to account for the ion mobility in the electrolyte solutions as a result of diffusion and electrostatic forces. Boltzmann distribution equation was adopted to analytically depict the relationship between the ionic concentration and local electrical potential in the diffuse layer. Stern [31] combined the Helmholtz model and the Gouy-Chapman model, and divided the EDL into two characteristically distinct layers, i.e., the Stern layer (Helmholtz layer) and the diffuse layer (Gouy-Chapman layer), as shown in Fig. 3. The Stern layer accounts for the specific absorption of the ions on the electrode surface, whilst the diffuse layer incorporates the Gouy-Chapman model [32]. The total capacitance of EDL can be treated as the Stern layer and diffuse layer capacitances connected in series. An unrealistic ion concentration value may be obtained by deriving the Poisson-Boltzmann (PB) equation; this model treats the ions as point charges by ignoring their physical size, but the point-charge assumption is only

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