



Review

Hierarchical micro-architectures of electrodes for energy storage

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HIGHLIGHTS

- Critically reviewed the architectures of electrodes.
- Summarized parameters of LIBs and SCs.
- Compared electrochemical performance of LIBs and SCs.
- Identified structures that are favorable for improving energy storage.

ARTICLE INFO

Article history:

Received 20 December 2014

Received in revised form

27 February 2015

Accepted 11 March 2015

Available online

Keywords:

Energy storage devices

Electrochemical performance

Charge transport

Lithium ion batteries

Supercapacitors

Micro-architectures

ABSTRACT

The design of electrodes for the electrochemical energy storage devices, particularly Lithium ion batteries (LIBs) and Supercapacitors (SCs), has extraordinary importance in optimization of electrochemical performance. Regardless of the materials used, the architecture of electrodes is crucial for charge transport efficiency and electrochemical interactions. This report provides a critical review of the prototype architectural design and micro- and nano-material properties designated to electrodes of LIBs and SCs. An alternative classification criterion is proposed that divides reported hierarchical architectures into two categories: aligned and unaligned structures. The structures were evaluated and it was found that the aligned architectures are superior to the unaligned in the following characteristics: 1) highly-organized charger pathways, 2) tunable interspaces between architecture units, and 3) good electric-contacted current collectors prepared along with electrodes. Based on these findings, challenges and potential routes to resolve those are provided for future development.

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1. Introduction

The demand for environmentally friendly energy has been increasing continuously for three decades [1,2]. The greenhouse effect, climate change, and potential exhaustion of fossil fuels require humans to develop novel clean and sustainable energy resources [1,3]. Electrochemical energy storage devices, especially lithium ion batteries (LIBs) [2,4–16] and supercapacitors (SCs) [2,3,5,10,11,16–21] are the most attractive alternatives to store energy with higher efficiency. Their potential applications include microchips [6], portable electronic devices [7–9], electric vehicles [6–8,10,15], and smart grid systems [7,10], among others. To date, electrochemical performances of LIBs and SCs are less than satisfactory. For example, LIBs have high energy density but low power

density, longer charging period, and low cycles [10] while SCs have high power density, shorter charging time, and tremendous number of cycles but low energy density [10]. Since LIBs and SCs rely on the electrodes kinetics [7] and transportation [22], intensive focus [4,5,7,8,11,12,15,21] has been on novel electrode materials and innovative electrode architectures for the eventual purpose of the creation of new electrochemical devices. The ideal design should possess characteristics of high energy and power density, good charging/discharging rate stability, long-cycle lifespan, light weight, small size, reliable performance safety, advanced mechanical and chemical stability, low cost, and environmental friendliness [7].

With the recent advancement in nanostructured materials, electrodes have emerged with favorable potentials. When an electrode decreases its dimension to micrometer length scale, alternative properties emerge, such as 1) high specific surface area that can lead to higher power density [2,15]; 2) new reactions that are impossible in bulk electrodes [2]; 3) short charge transport

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distances that can result in higher power density [2,15]; 4) longer cycling lifetime by partially eliminating the large volume expansion/extraction during charging/discharging because of higher strength of nanostructures to adapt large strain without pulverization [2,15,23], etc. Besides the importance of the intrinsically good properties of electrode materials, the architecture, also known as the structure/morphologies, of the electrode also need to be considered with creative ideas [2,9–11].

In this review, architectural designs and constructions of the electrode materials for LIBs and SCs will be investigated in terms of aligned and unaligned. The aligned architectures mean the ones from the prototype design with an organized base, e.g. 1-D nanorods [24]/nanowires [25]/nanotubes array [26], 3-D nanorods array [27], nanowalls [28]. Such aligned architectures possess the highly-organized arrangement. The unaligned architectures imply the randomly located sites in the initial electrode design, e.g., 3-D microcubes [29], nanospheres [30], 3-D inverse opal [31], nanotubes network [32]. For most designs of unaligned architectures, the electrodes are mainly composed by the aggregation of large amount of basic units. Their corresponding electrochemical performances and impacts to the charge transport will be introduced. At the end, summary and future outlook will be presented based on the state-of-the-art electrode architectures.

2. Basic principles on lithium ion batteries (LIBs) and supercapacitors (SCs)

2.1. Lithium ion batteries (LIBs)

Lithium ion batteries are the most promising electrochemical energy storage devices. They were first introduced commercially by Sony Inc. in 1990 [3]. Being mass produced for more than two decades, the typical components of a LIB are solidified into an anode (negative electrode) made by porous carbon/graphite, a cathode (positive electrode) made by layered lithium-transition metal oxide (e.g. LiCoO_2 [23,33], LiFePO_4 [34]), and a non-aqueous liquid electrolyte [8,11,35]. As illustrated in Fig. 1a, Li ions are extracted from the cathode during charging. These ions transport through the electrolyte to the anode and accomplish the intercalation. The driving force of such intercalating transport is the potential provided by the external power source, i.e., the power source donates electrons that move from the electrode towards anode. This results in the charge of transition metal ions (e.g. Co^{3+} to Co^{4+}) as well as the carbon atoms (C to C^-). According to Fig. 1b, during discharging Li ions are released from the anode and intercalate back to the cathode. Simultaneously, the electrons emerged from both the reduction of transition metal cations and the oxidation of carbon anions provide the energy of external loading [8].

Normally, the electrodes of LIBs are rated by the specific capacity [11] (i.e., gravimetric capacity, in mAh g^{-1} or mAh cm^{-2} [10]) measured from the charge/discharge graphs (normally is galvanostatic charge/discharge). A larger specific capacity means more charge can be stored in unit mass/area of electrode material before the potential exceeds its reversible window [11].

2.2. Supercapacitors (SCs)

The concept of supercapacitor was discovered by Becker in 1957 when he used the high-surface-area carbon coated a metallic substrate as a current collector [18]. Supercapacitors are capacitors which contain a liquid electrolyte solution in a dielectric layer. Supercapacitors are also known as electrochemical capacitors or ultracapacitors [21]. The terms super and ultra reflect their exceptional ability to store energy. There are two types of SCs categorized according to the mechanisms of charge storage and electrode

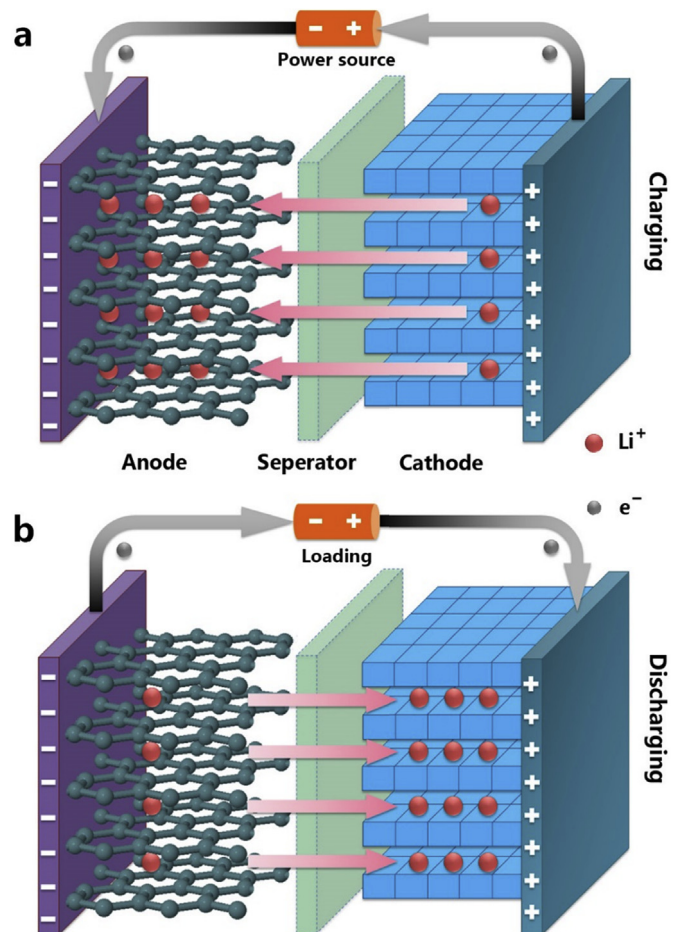


Fig. 1. Schematic illustration of the basic structure of a Lithium ion battery along with the charge transport mechanism inside it. Panel a shows the transport directions of both Lithium ions and electrons during charging; Panel b illustrates the processes of Lithium ions transport and electron supply to external loading.

materials applied. One is electron double layer capacitors (EDLCs) [36], the other is pseudocapacitors [3,36]. EDLCs are electrochemical capacitors that store charges electrostatically by reversible electrolyte ions adsorption onto active electrode with high surface area. Under the influence of an applied electric potential upon the two parallel plates, there is a spontaneous movement of charges in both solid electrode and liquid electrolyte towards one another. In this case, the electric double layers emerges at the interface of electrode and electrolyte. Electrons move from the positive plate to the negative simultaneously. During discharging, the electrons move back to positive plates from negative counterpart through the external loading. This existing and transporting type of chargers is almost a duplicate of that of conventional electrostatic capacitors. Owing to the extreme narrow distance between the double layers and high contact area, the capacitance of EDLCs is extremely large. Currently, commercial EDLCs use high-surface-area active carbon as the electrode material [11]. Well-established theoretical models were constructed by some pioneers of Helmholtz, Gouy, Chapman, and Stern in the 19th and early 20th centuries [17]. In contrast, pseudocapacitors mainly utilize the fast, potential-variable, and reversible surface or near-surface redox reactions rather than electrostatic charge storage to achieve high capacitance [21]. The charge storage ability of pseudocapacitors mainly depends on the material and its structure as well as the specific surface area of electrodes [17]. Transition metal oxides [37]

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