



Electrochemical performances and capacity fading behaviors of activated carbon/hard carbon lithium ion capacitor



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ABSTRACT

Lithium ion capacitor (LIC) is one of the most promising electrochemical energy storage devices, which offers rapid charging-discharging capability and long cycle life. We have fabricated LIC pouch cells using an electrochemically-driven lithium pre-doping method through a three-electrode pouch cell structure. The active materials of cathode and anode of LIC cell are activated carbon and pre-lithiated hard carbon, respectively. The electrochemical performances and the capacity fading behaviors of LICs in the voltage range of 2.0–4.0 V have been studied. The specific energy and specific power reach 73.6 Wh kg^{-1} and 11.9 kW kg^{-1} based on the weight of the active materials in both cathode and anode, respectively. Since the cycling performance is actually determined by hard carbon anode, the anode potential swings are emphasized. The capacity fading of LIC upon cycling is proposed to be caused by the increases of internal resistance and the consumption of lithium stored in anode. Finally, a large-capacity LIC pouch cell has been assembled with a maximum specific energy of 18.1 Wh kg^{-1} and a maximum specific power of 3.7 kW kg^{-1} based on the weight of the whole cell.

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1. Introduction

Electrochemical energy storage devices with fast charging-discharging capability, high energy density and long cycle life are highly required for various applications, such as electric or hybrid electric vehicles (EV/HEV), wind power stations, and consumer electronic products. Lithium ion capacitor (LIC) is an asymmetric electrochemical capacitor containing a capacitive electrode and a lithium-intercalation electrode, such as activated carbon (AC) electrode as cathode, and pre-lithiated carbonaceous-based electrode as anode [1]. The energy density of LIC can reach $12–15 \text{ Wh kg}^{-1}$, which is 2–3 times higher than that of EDLC. Hence, LIC has drawn extensive attentions due to the attractive merging of lithium ion battery (LIB) and electric double layer capacitor (EDLC) [2–11].

Several factors can influence the electrochemical performances and cycling stabilities of LICs, including types of anode materials, pre-lithiation degree, electrode potential window, and instability of the lithiated carbonaceous anode [12–15]. Kim et al. [16] investigated the electrochemical properties of natural graphite, artificial graphite and hard carbon (HC) employing half- and full-cell configurations. It was found that HC anode showed 70% capacity retention at 40C-rate (throughout this study, nC-rate means that the rated capacity is charged or discharged completely in 1/n hours), and 83% capacity retention after 10,000 cycles. Schroeder et al. [17,18] studied the electrochemical performances and the cycling stability of LIC containing soft carbon (petroleum coke, PeC) and graphite as anodic materials. It showed that PeC-based LIC displayed remarkable cycling stability close to that of EDLC. Zhang et al. [19] prepared flexible packaged LIC cells with AC cathode and pre-lithiated mesocarbon microbeads (MCMB) anode with the mass ratio of 1:1. The capacitor with the pre-lithiation degree of 56.5% revealed better high-rate capability and cycling stability. Aida et al. [20] observed large capacity loss during charge-discharge cycling when AC cathode was highly polarized at a potential over 4 V vs. Li^+/Li . Cao and Zheng [13,21] assembled LICs

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with AC cathode and HC/stabilized lithium metal powder anode. It revealed that the maximum potential for AC cathode was limited by the electrolyte stability, while the minimum potential for HC anode was determined by the growth rate of the solid electrolyte interphase (SEI), i.e., the formation current. Extra capacity in anode was beneficial for improving the cycle life of LIC cell. Sivakkumar and Pandolfo [22] evaluated LICs with AC cathode and pre-lithiated graphite anode. The capacity fade of LIC during continuous cycling was directly correlated with the reduced utilization of AC material, which was caused by the poorly formed SEI film on anode surface.

In our previous works [23,24], we have proposed a novel three-electrode pouch cell structure, in which a lithium metal foil was placed near the anode and separated by a separator. The structure and the fabrication process of the full-cell with flexible package can be found in our previous publications [24,25]. The pre-lithiation process is accomplished during the electrochemical discharging process of HC anode. The lithium metal foil acts as both counter electrode and lithium source. Lithium ion is electrochemically driven to migrates from Li electrode to HC electrode through the holes in current collector and separator. Besides, the potentials of cathode and anode can be monitored simultaneously by using the lithium metal electrode as a reference electrode during charging-discharging processes of the full-cell. We have noticed that the control of the electrode potential swing is critical for a LIC cell to obtain optimal electrochemical performances and better cycle-ability. For example, the potential limits of cathode and anode depends on ambient temperature, charge-discharge rate, pre-lithiation degree and mass ratio between cathode and anode. In our observation, the capacity fading upon cycling occurred mainly

in the initial 1,000 charge-discharge cycles, especially in the first 500 cycles. After then, the capacity reaches a relative stable state. Similar behaviors for LIC have been reported in other publications [18,26,27]. Hence, we have carefully examined the electrochemical properties of LIC during the first 500 cycles in this work. We have studied the drifts of potential swing at different C-rate and upon cycling. The internal resistance of a LIC cell has been obtained from the charge-discharge data and the electrochemical impedance spectroscopy (EIS) analysis. The effects of pre-lithiation capacity on the anode potential and the cycling stability have also been discussed. The scanning electron microscopy (SEM) images reveals that the needle-like lithium deposits can be formed on the surface of HC electrode. Additionally, a large-capacity pouch LIC cell has been assembled by using the optimal parameters.

2. Experimental

The cathode was prepared by coating a slurry mixture on a 20- μm -thick porous aluminum foil (through-holes with a diameter of 100 μm). The mixture contains activated carbon (AC, YP50F, purchased from Kuraray chemicals, Japan), carbon black, and poly (vinylidene fluoride) with a mass ratio of 80:10:10. The anode was prepared by coating a slurry mixture on a 20- μm -thick porous copper foil (through-holes with a diameter of 100 μm). The mixture contains hard carbon (HC, purchased from ATEC Company, Japan), carbon black, sodium carboxymethyl cellulose and styrene-butadiene rubber with a mass ratio of 90:5:2:3. The employed electrolyte is 1 M lithium hexafluorophate (LiPF_6) in the mixture solvent of ethylene carbonate, dimethyl carbonate, and diethyl

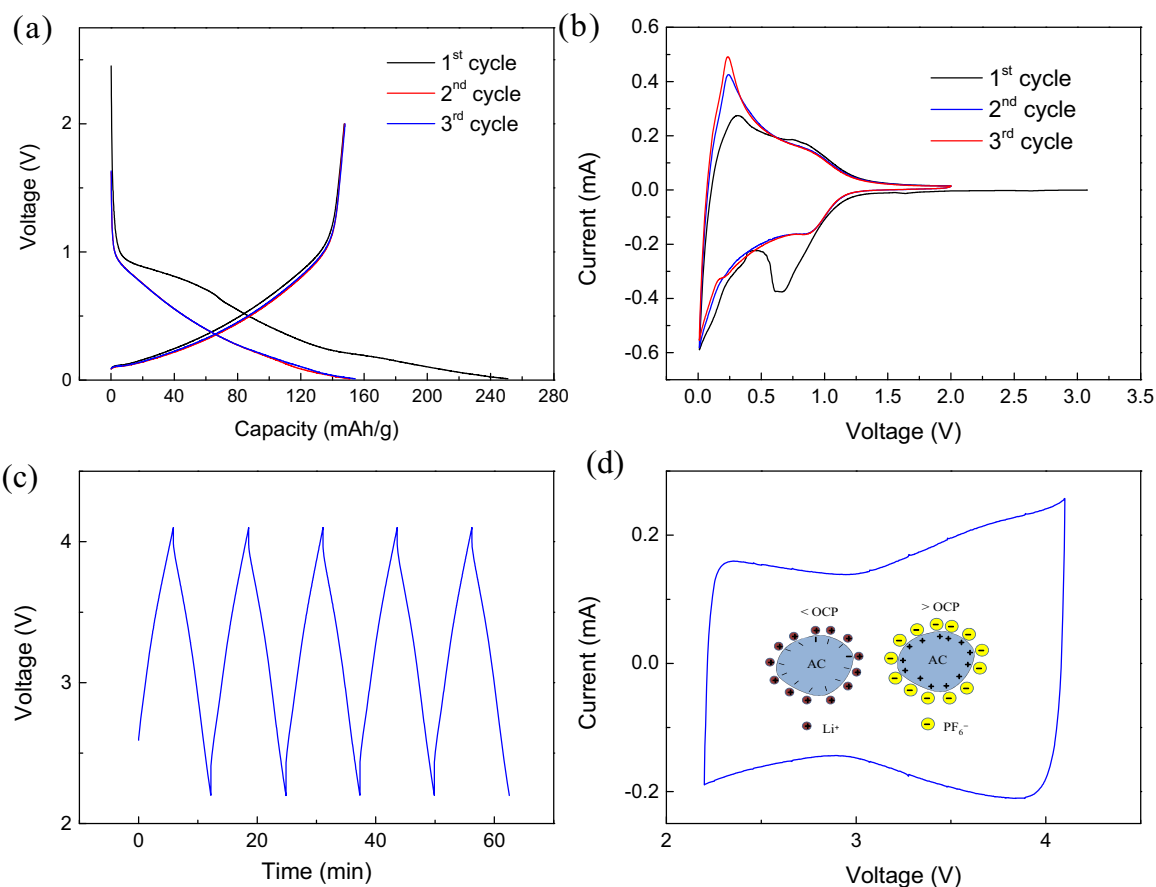


Fig. 1. Charge-discharge curves of (a) HC/Li coin cell in the voltage range of 0.01–1.5 V at 50 mA g^{-1} , (c) AC/Li coin cell in the voltage range of 2.2–4.1 V at 25 mA g^{-1} . Cyclic voltammetry curves of (b) HC/Li coin cell and (d) AC/Li coin cell at the scan rate of 0.5 and 0.1 mV s^{-1} , respectively. Inset of (d) is the schematic illustration of cathodic and anodic process happened on AC surface.

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