



Development and characterization of Li-ion capacitor pouch cells



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HIGHLIGHTS

- The specific energy and energy density are 30 Wh kg⁻¹ and 39 Wh L⁻¹, respectively.
- The capacitor was discharge at rates over 100 C rate.
- The capacitor was cycled over 10,000 cycles with capacitance degradation <20%.

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ABSTRACT

High energy density Li-ion capacitor (LIC) pouch cell prototypes were assembled with lab-scale equipment using activated carbon cathode and hard carbon/lithium stabilized metal powder (SLMP) anode. The specific energy and energy density as high as 30 Wh kg⁻¹ and 39 Wh L⁻¹ have been achieved, respectively. The pouch cells can deliver over 50% of the maximum stored energy at a discharge rate over 100 C-rate. After 10,000 cycles, the LIC pouch cell still has 80% of the initial capacitance. The average leakage current is 0.3 μA cm⁻² during the first 72 h.

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1. Introduction

Lithium-ion Capacitors (LICs) have attracted much attention as a new power source due to its high energy density compared to conventional electric double-layer capacitors (EDLCs), and its high power density compared to batteries as well as having the long cycle life. LICs contain a lithium-ion battery (LIB) anode electrode and an EDLC cathode electrode [1–3]. Extensive research has been done to optimize the electrochemical performance of the LICs [4–16]. Recently, Balducci et al. [17] reported the use of carbon coated iron oxide-based electrodes for the anode for LICs.

Schroeder et al. [18] used soft carbon for the anode for LICs to achieve high power density and long cycle life. Smith et al. [19] have done research into the electrochemical performance and thermal behavior of LICs, and the maximum energy density of the LIC pouch cells was approximately 10–15 Wh kg⁻¹. The LIC cells used in Smith's group were obtained from JM Energy and contained a third electrode of Li metal to pre-dope the anode electrodes, which was first proposed by Fuji Co. [6].

Previously, we have reported a LIC with carbon cathode and hard carbon/SLMP anode electrodes with high energy density, high power density and long cycle life [20,21]. This was made possible by replacing the conventional activated carbon anode of EDLCs with hard carbon (HC) with stabilized lithium metal powder (SLMP) on its surface. The added SLMP increased the open circuit voltage of the LIC and ensured less salt to be consumed when the LIC was charged, which was the reason for the high energy density. The charge–discharge maximum voltage of the LIC could be achieved as

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high as 4.1 V with stable cycling performance [22]. During our LIC cell assembly, a conventional two-electrode structure was used, and the SLMP was applied onto the surface of the hard carbon anode electrodes and some of Li from the SLMP was intercalated into the hard carbon when the electrolyte was filled into the cell. This high energy density concept of the LICs with such novel structure has already been demonstrated by laboratorial coin cells [22]. However, the LIC pouch cells with such novel electrode composition and the two-electrode structure have not been reported by any research group. Therefore, in this paper, we wish to report the detailed performance of two LIC pouch cells LIC250 and LIC395. The comparison of the energy and power performance between LIC250 and LIC395 is also included in this paper.

2. Experimental

Commercial active materials were used for both the positive and negative electrodes as received. The positive electrode (PE) was prepared by coating a slurry mixture of activated carbon (AC) and polytetrafluoroethylene (PTFE) as a binder by the mass ratio of 90:10 on an Al foil substrate (Exopack™, 20 μm in thickness). Two kinds of AC materials including YP-50F (Kuraray Chemical Co, Japan) and AB-520 (MTI Corporation; USA) were used to manufacture the cathode electrodes. The slurry mixture of the negative electrode (NE) was made of hard carbon (HC, Carbotron P(J), Kureha Japan) and PTFE as a binder by the mass ratio of 92:8. After the slurry was prepared, it was coated onto a Cu foil substrate that had a thickness of 10 μm. Then the electrodes were dried at 100 °C for 2 h in oven with flowing air. After all the electrodes were dried, a hot-roll pressing was applied to the AC and HC electrode sheets to make them into the desired thickness, 150 μm for AC and 90 μm for HC. All the electrode sheets were kept in the dry room and punched out into 8 × 5 cm (active area) electrodes with additional 1 cm long tab. All the electrodes were dried again at 120 °C for overnight in a vacuum oven then the stabilized lithium metal powder (SLMP) was applied onto the surface of all the prefabricated HC anode electrodes by doctor blade method and then roll-pressing in a dry room as shown in Fig. 1 before being assembled into the pouch cell. The SLMP, which is purchased from FMC Lithium, is Li powder with a passivation layer at surface and the average size of the Li powder is ~40 μm. Fig. 2(a) displays the 8 × 5 cm anode electrode with additional 1 cm long Cu tab before loading the SLMP. After all the SLMP was pressed onto the anode electrode, there was a uniform thin layer of SLMP on the surface of anode electrode as shown in Fig. 2(b).

Two pouch cells LIC250 and LIC395 were assembled in the dry-room by stacking four double-side coated HC/SLMP anode electrodes, three double-side coated AC cathode electrodes and two single-side coated AC cathode electrodes. The activated carbon

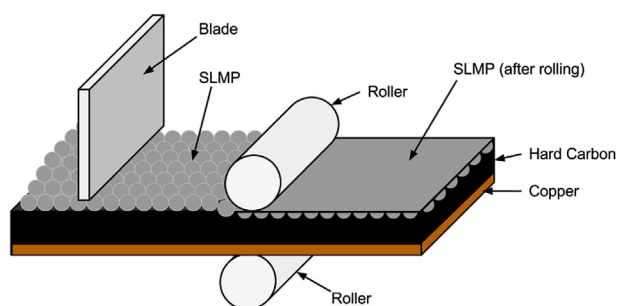


Fig. 1. Schematic view of coating process of stabilized lithium metal powder (SLMP) on the surface of a negative electrode.

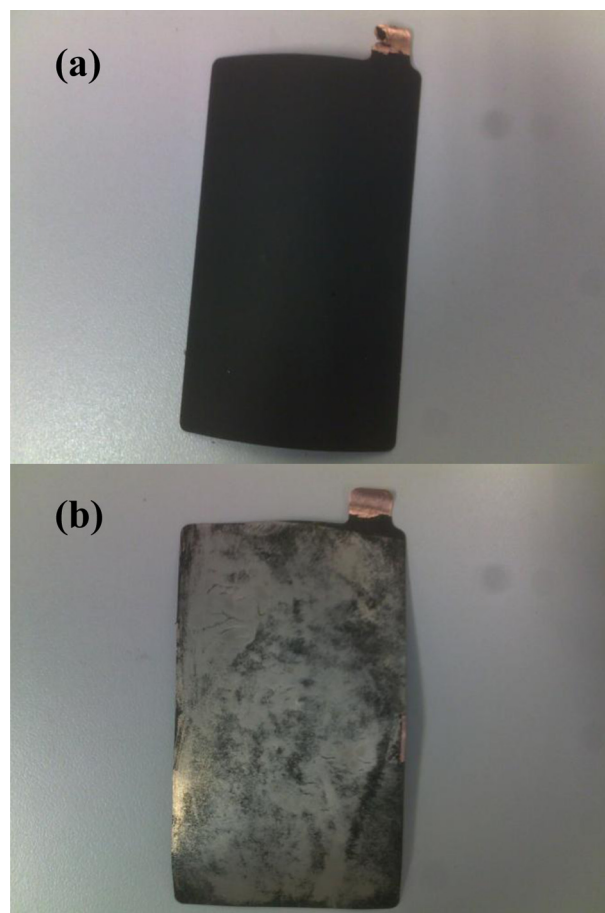


Fig. 2. (a) 8 × 5 cm HC electrode with 1 cm long tab and (b) HC electrode with SLMP pressed on surface.

material YP-50F was used to assemble the pouch cell LIC250, while LIC395 used cathode material AB-520. Fig. 3 shows the schematic representation and the stacking core of the pouch cells. It can be seen from Fig. 3 that the Cu and Al substrates were welded to nickel (Ni) and aluminum (Al) current collector tabs, respectively. After the stacking and welding processes, the cell core was housed in an aluminum laminated formed case that is suitable for the size of the electrodes. Then the vacuum heat sealing process was applied to the case with the cell core in order to remove the excess gas trapped in the cell after the pouch cells were filled with electrolyte. The separator used was a microporous membrane (Celgard 3501). The electrolyte was 1 M LiPF₆ in ethylene carbonate (EC):dimethyl carbonate (DMC) at a ratio of 1:1 by weight (LP30, SelectiLyte™, Merck Electrolyte). Both LIC250 and LIC395 were charged-discharged under various currents in order to obtain the energy density under different discharge C-rate. The LIC pouch cells were then charged and discharged under a charge-discharge C-rate of 20 C to obtain the cycle life performance and the leakage current of the LIC was also tested. The LIC pouch cells were charged and discharged using an Arbin Battery Testing Unit and the electrochemical impedance spectrum (EIS) for LIC250 and LIC395 was recorded in the frequency range of 0.1–10⁶ Hz using Gamry Instruments.

The specific surface area and pore structure of the two kinds of positive electrodes (YP-50F and AB-520) were determined with the nitrogen adsorption/desorption isotherms at 77 K (Quantachrome Autosorb-iQ). All the samples were degassed for 12 h before the adsorption experiments. The specific surface area was calculated by

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