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Effect of back-side-coated electrodes on electrochemical performances of lithium-ion batteries

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HIGHLIGHTS

- Back-side-electrode influences on electrochemical performances of full cells.
- Back-side-coated cathode improves the Coulombic efficiency and discharge capacity.
- Back-side-coated anode hampers the Coulombic efficiency and discharge capacity.

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ABSTRACT

In order to investigate the effect of back-side coating of cathodes and anodes upon electrochemical performances of lithium-ion batteries (LIBs), four different pouch-type lithium cobalt oxide ($LiCoO_2$, LCO)/graphite unit cells with different coating conditions are prepared and compared in a systematic manner. Their electrochemical performance, in terms of Coulombic efficiency, capacity realization, capacity retention ability, and rate capabilities, is investigated. From the results, we confirm an opposing relationship existing between back-side-coated cathodes and anodes, in that the coated cathodes provide improved cell performance, while the coated anodes impede it. This is attributed to the fact that, as is generally understood, cathodes act as noble lithium (Li) ion suppliers for LIBs, while anodes consume a large portion of Li ions to form surface layers during the first charging process. Furthermore, we also confirm that the magnitude of the back-side coating effect for LCO and graphite are somewhat different. The double-side-coated cathodes have a notable positive effect on cell performances in contrast to the negative effect seen with the double-side-coated anodes. As a result, unit cells employing double-side-coated cathodes with single-side-coated anodes show the best performance, followed by those based on double-side-coated cathodes with double-side-coated anodes rather than single-side-coated cathodes with single-side coated anodes.

1. Introduction

Lithium-ion batteries (LIBs) have been a promising power source for advanced energy storage technologies over the last two decades and, due to their high specific energy density and high energy efficiency, have been successfully applied in a variety of fields, from small-scale consumer electronics to large-scale applications, such as electric vehicles (EVs), and energy storage systems (ESSs) [1–3].

Since their first use in the early 1990s, judging on a basis of commercialized 18650-type cylindrical cells, LIBs have made steady gains in energy density [4,5]. It is worth noting, however, that cell manufacturers have been able to accomplish the majority of this progress by changing cell design parameters such as electrode thickness, porosity, particle size, slurry composition, and tab design rather than by fundamental changes to the existing cathode and anode materials, which have remained lithium cobalt oxide ($LiCoO_2$, LCO) and graphite [4,6–8]. This reminds us of a crucial, but

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often neglected, fact that cell design parameters play a pivotal role in determining the cell performance of LIBs.

In order to maximize the energy and power density of LIBs resulting from the maximum loaded amount of active cathode and anode materials, cell manufacturers favor a cylindrical and/or prismatic shape of battery [1,9,10]. Within these cells, electrodes and separators are coiled into a strip-wound design—the so-called “jelly-roll”—or are flatly laminated into a rectangular geometry stack (or combination of both ways: the so-called “stack and folding” type) [9,11]. Although much effort has been made to minimize the usage of electrochemically inactive constituents (such as current collectors and separators), LIBs have an inherent fault in that they possess an excess of isolated active material that does not face counter-electrodes, as shown in Fig. 1 colored in red (in web version). This arises from the fact that electrodes are coated on both sides of the current collectors, and the area of the anode is slightly larger than that of the cathode, which ensures easy coverage during cell assembly without the surface displacement between cathodes and anodes. This improves both processing and cost efficiency.

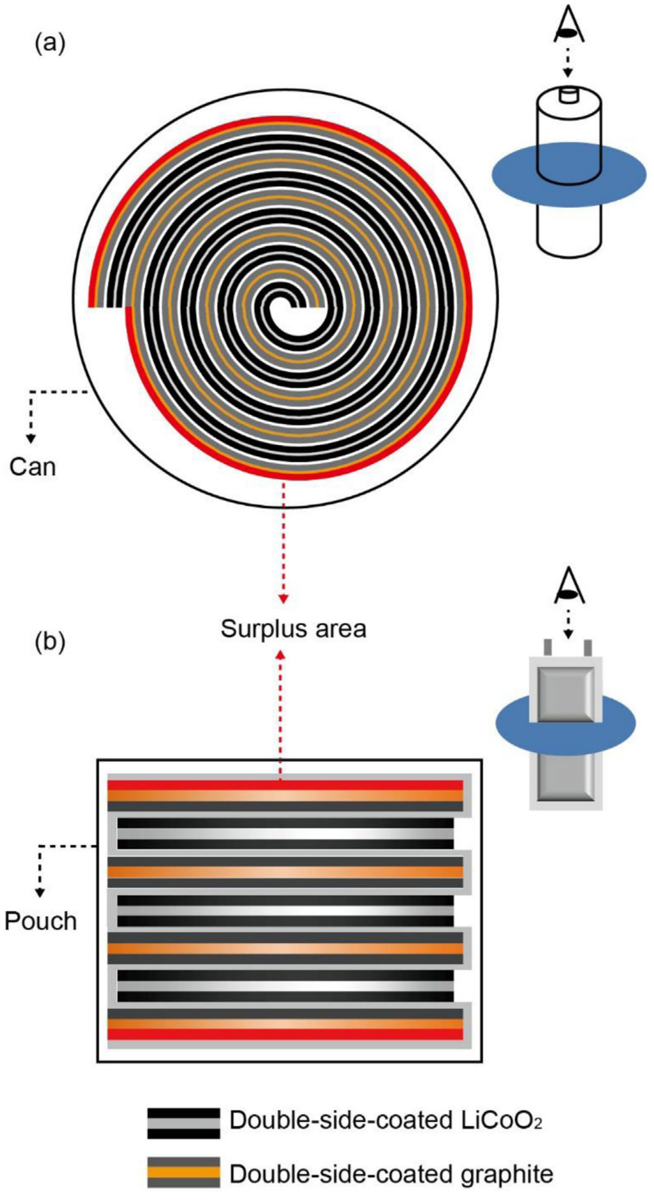

Fig. 1. Schematic diagrams describing a (a) cylindrical and (b) prismatic shape of battery.

In a quantitative point of view, for the large-scale LIBs consisted with multi-layered large-area electrodes, the ratio of the excess isolated active material to the overall active material becomes so small that the significance of the excess isolated active material can be negligible. Nonetheless, the large-scale LIBs still suffer from the significant cell performance variation in accordance with the electrode condition, the origin of which has so far been unclear. Along with this line, we have previously reported on the effect of the cathode/anode area ratio on the electrochemical performances of LIBs [12]. We observed that larger anodes can significantly reduce initial Coulombic efficiency and result in a poor rate capability. As a further step toward proper electrode preparation, herein, we have investigated the effect of back-side coating with active materials on the electrochemical performances of LIBs using a pouch-type cell, which emphasizing the role of back-side coating. We investigated this by exploring various cases of separator-electrode assembly, these being: 1) single-side-coated cathode/single-side-coated anode, 2) single-side-coated cathode/double-side-coated anode, 3) double-side-coated cathode/single-side-coated anode, and 4) double-side-coated cathode/double-side-coated anode.

2. Experimental

2.1. Electrodes preparation

Cathodes were prepared by coating *N*-methyl-2-pyrrolidone-based slurry (NMP, Sigma–Aldrich, USA) consisting of 92 wt.% lithium cobalt oxide ($LiCoO_2$, LCO, 10 μm), 4 wt.% conductive carbon (Super-P, TIMCAL, Switzerland), and 4 wt.% polyvinylidene fluoride (PVdF, Sigma–Aldrich) binder onto aluminum (Al) current collector foil (20 μm). The active material loading level of cathodes in this work was 12.2 mg cm^{-2}. The thickness of single cathode coating layer were controlled to be 40 μm with density of 3.05 g cm^{-3}.

Anodes were prepared in a similar way, by coating NMP-based slurry containing natural graphite (85 wt.%), Super-P (8%), and PVdF binder (7%) onto copper (Cu) current collector foil (10 μm). The active material loading level of anodes in this work was 6.1 mg cm^{-2}. The thickness of single anode coating layers was controlled to be 48 μm with density of 1.27 g cm^{-3}. More detailed information associated with cathodes and anodes is summarized in Table 1.

2.2. Cell assembly

For unit cells with various types of coated electrodes, cathodes and anodes varying in different coating conditions were punched as 4 cm^2 (2 cm × 2 cm) for cathode and 4.84 cm^2 (2.2 cm × 2.2 cm) for anodes to make sure perfect coverage of cathodes during assembly. Electrodes were dried at 80 °C for 12 h under vacuum before use.

Table 1
Materials, loading level, thickness, densities, and designed capacities of prepared cathode and anode.

	Unit	Cathode	Anode
Active materials	–	$LiCoO_2$	Natural graphite
Loading level	mg cm^{-2}	12.2	6.1
Single coating thickness	μm	40	48
Electrode density	g cm^{-3}	3.05	1.27
Nominal capacity	mAh cm^{-2}	1.53	1.93
Current collector	–	20 μm Al foil	10 μm Cu foil