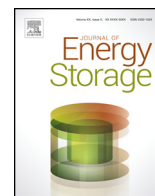




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Improvement of lithium-ion battery performance using a two-layered cathode by simultaneous slot-die coating

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ABSTRACT

The electrochemical performance of lithium-ion batteries (LIBs) could be improved using a two-layered cathode produced by a simultaneous slot-die coating method. The composition in each layer contained different proportions of two conductive additives; one with a high percentage of micro-sized graphite, and the other with a high percentage of nano-sized carbon black. Of the four cell structures fabricated, the two-layered cathode with an upper layer having more carbon black and a lower layer having more graphite particles was found to give the best current discharge capacity and longer life cycle. This finding could have potential applications in new LIBs design.

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1. Introduction

Lithium-ion batteries (LIBs) have been developed for extensive industrial applications in recent years. An important application of LIBs is in the electrification of automobiles including electric vehicles (EVs), hybrid electric vehicles (HEVs), and plug-in hybrid electric vehicles (PHEVs). For this type of batteries to be viable in automotive applications, it must possess superior electrochemical properties such as high life cycle, high energy and high power density, as well as sustainability after prolonged period of usage [1–3].

There were different approaches reported in the literature to improve the performance of LIBs, either by changing the formulation of electrodes or altering their structures. The most common approach was to change the electrode compositions including the active materials, binders and conductive additives [4–6], and to ensure that a good slurry dispersion was achieved [7]. The active materials in cathode generally exhibited poor conductivity and adding some conductive agents would be necessary [4,8]. The two most common commercial conductive agents were the nano-sized carbon black (Super-P) and the micro-sized graphite (KS-6). Some studies showed that adding more graphite (KS-6) could improve the battery cycle life, and adding more

carbon black (Super-P) could increase the battery capacity under high C-rate discharge. Hence, an optimum combination of these two additives must be found in order to achieve a high overall battery performance [9,10]. Attempts were made to improve the LIBs' performance by using different binder contents [11,12], different conductive additives [13,14] and/or changing the binder to conductive additives ratio [15–19]. Nevertheless, it seemed difficult to enhance substantially both power and energy performances using a single formulation cathode. A thicker electrode was found to provide a high energy capacity but to yield a low performance at high C-rate condition; whereas the opposite trend was observed for a thinner electrode [4,6].

Another approach to increase the performance of LIBs was to mix two active materials in a composite electrode and created a multilayer electrode [20–23]. The traditional way of making a multilayer cathode was by producing a single-layered cathode through a conventional slot-die coating followed by drying; then a second layer was similarly produced and combined with the first. However, when the two single-layered cathodes were bound together, binder diffusion of the second layer into the first could occur, resulting in an increase in the electrode resistance [21]. This problem could be overcome by designing a two-layered cathode with two separate coated layers. The two-layered structure appeared to provide more degrees of freedom in the selection of formulation in each layer. An efficient method of constructing a two-layered cathode was to coat the two layers simultaneously using the slot-die coating technology. This method of coating had

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been developed for a long time, and was successfully applied in many areas [21,24–26].

It was mentioned in the literature [9,10] that mixing different compositions of two conductive additives of vastly variable sizes and shapes as in the case of Super-P and KS-6 in a single-layered cathode could result in different electrochemical performances. This observation prompted the current thinking of using a two-layered cathode produced from simultaneous multilayer slot-die coating, with each layer having the same two conductive additives but different compositions. Several cells were then constructed using a two-layered cathode and a single-layered anode for testing. The battery performances in terms of cell discharge capacity, electrochemical impedance spectra (EIS), C-rate and cycle life were evaluated and compared.

2. Experimental

2.1. Cathode materials

There were four major ingredients in the formulation of cathode. The active material was $\text{LiNi}_{0.5}\text{Mn}_{0.3}\text{Co}_{0.2}\text{O}_2$ (LNMC 532, TX10, Umicore) of average particle size $11.9\ \mu\text{m}$ (d_{50}), and BET surface area $0.28\ \text{m}^2\ \text{g}^{-1}$. The two conductive additives were synthetic graphite (Timrex KS-6, Timcal) of average particle size $3.3\ \mu\text{m}$ (d_{50}) and surface area $21.8\ \text{m}^2\ \text{g}^{-1}$, and carbon black (Super-P, Timcal) of average particle size $40\ \text{nm}$ and surface area $64.1\ \text{m}^2\ \text{g}^{-1}$. The binder used was poly-vinylidene fluoride (PVDF1300, Kureha) and the organic solvent was *N*-methyl-2-pyrrolidone (NMP, Taiwan Maxwave).

2.2. Cathode slurries preparation and slot die coating

Two different cathode slurries of varying compositions, as given in Table 1, were prepared for the experiment. The difference between Slurries A and B was the relative amount of the conductive additives KS-6 and Super-P.

The cathode slurry was prepared by first dissolving the binder PVDF in the solvent NMP, and the active material and conductive additives were subsequently mixed into the binder solution. The detailed procedure of the mixing process was presented in a previous paper [7]. The slurry solid content was fixed at 60 wt.% and 55 wt.% for Slurries A and B, respectively. The mixing was deemed to be complete when the slurry viscosity as measured in a cone-and-plate rheometer (ARES-2, TA Instrument) became independent of time at constant mixing speed or shear rate.

A two-layered slot-die coater was used for coating the slurries. The coating width was set at 10 cm. A sketch of the two-layered simultaneous slot-die coating operation was shown in Fig. 1, with the front and side views of the coater shown in Fig. 1(a) and Fig. 1(b), respectively. The slurries were delivered by two screw pumps (Heishin, Japan), and coated on an aluminum foil. The coating speed was controlled at $1.7\ \text{cm}^{-1}$. The coated layer was then dried in a pilot line consisting of two oven sections each of 2 m long. The temperatures in the first and second sections were set at $100\ ^\circ\text{C}$ and $120\ ^\circ\text{C}$, respectively. The air flow speed in the drier was

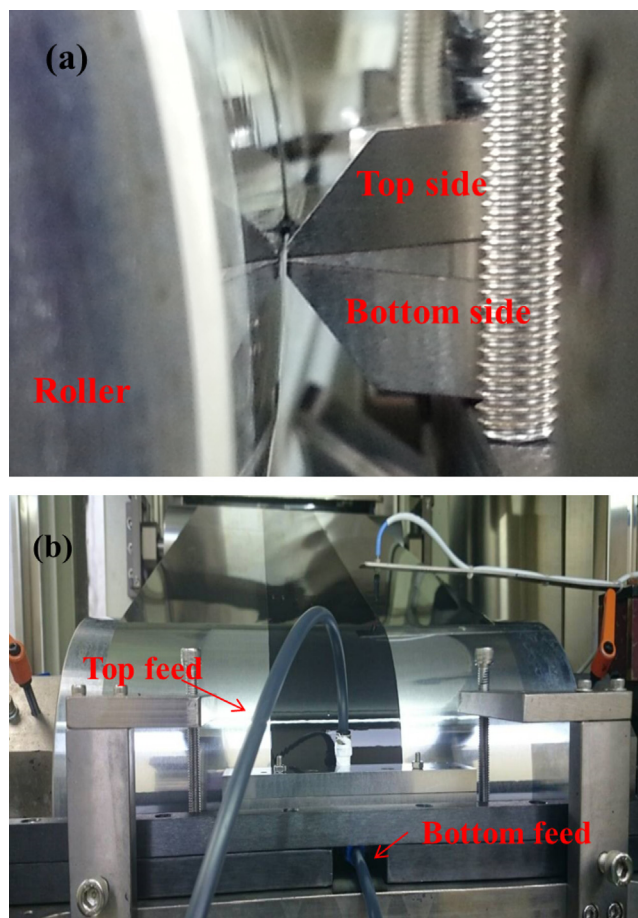


Fig. 1. Two-layered simultaneous slot-die coating: (a) front view; (b) side view.

$130\ \text{cm}^{-1}$. The total thickness of the dried coating layer was set around $95\ \mu\text{m}$, and the density of the compressed cathode sheets was around $2.5\ \text{g}\ \text{cm}^{-3}$.

Fig. 2 shows the four different cathode structures produced after the coating and drying operations. Fig. 2(a) and Fig. 2(b) are the single-layered cathodes of pure Slurry A and pure Slurry B, and Fig. 2(c) and Fig. 2(d) are the two-layered cathodes with Slurry A on top of Slurry B, and vice versa, respectively.

2.3. Physical properties of the cathode electrodes

The morphological characteristics of the cathode was examined by first solidified the sample in liquid nitrogen bath and then cut into pieces for surface topology measurements. The measurements were carried out using a field emission scanning electron microscopy (FE-SEM, JSM-6500F, JEOL).

The peeling test of the sample was performed in accordance to the ASTM D3359 standard. The sample was attached to a 2 cm wide tape (Scotch, 3M) and pulled by a testing machine (Imada), and the peeling force was recorded.

2.4. Preparation of the anode electrode

In order to perform the full electrochemical cell test, a cell must be assembled by combining the cathode with an anode. The anode slurry was prepared by mixing the ingredients in the same manner as the cathode. The active material in anode was meso-phased graphite powders (MGP-A, China Steel Chemicals), and the binder was PVDF (PVDF 9200, Kureha). Only a single conductive additive,

Table 1
The components of cathode slurries.

Constituents	Slurry A (wt.%)	Slurry B (wt.%)
LNMC 532	89	89
KS-6	5	2
Super-P	2	5
PVDF 1300	4	4

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