



Electrochemical properties of large-sized pouch-type lithium ion batteries with bio-inspired organic cathode materials



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HIGHLIGHTS

- Scale-up of batteries with flavin-based organic cathode materials was investigated.
- Large-sized pouch cell with lumichrome was fabricated using aqueous SBR-CMC binder.
- EC-DMC electrolyte exhibits better electrochemical performance than TEGDME.
- A battery module was able to power LEDs (850 mW) as an energy source system.

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ABSTRACT

To investigate the feasibility of scaling up bio-inspired organic materials as cathode materials in lithium ion batteries, large-sized pouch cells are successfully prepared via tape casting using lumichrome with an alloxazine structure and aqueous styrene butadiene rubber-carboxymethyl cellulose (SBR-CMC) binders. A battery module with a two-in-series, six-in-parallel (2S6P) configuration is also successfully fabricated and is able to power blue LEDs (850 mW). Lumichrome shows no structural changes during the fabrication processes used to produce the positive electrode. The large-sized pouch cells show two sets of cathodic and anodic peaks with average potentials of 2.58 V and 2.26 V vs. Li/Li⁺, respectively. The initial discharge capacities are 142 mAh g⁻¹ and 148 mAh g⁻¹ for ethylene carbonate-dimethyl carbonate (EC-DMC) and tetraethylene glycol dimethyl ether (TEGDME) electrolytes, respectively, similar to that of a coin cell (149 mAh g⁻¹). The EC-DMC-injected pouch cells exhibit higher rate performance and cyclability than the TEGDME-injected ones. The TEGDME electrolyte is not suitable for lithium metal anodes because of electrolyte decomposition and subsequent cell swelling.

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1. Introduction

Li-ion batteries are very attractive power sources for a variety of applications, including portable electronic devices and large-scale batteries [1]. In this technology, energy storage is mainly limited by the positive electrode [2], which is composed of inorganic transition metal oxide-based materials, such as LiCoO₂ and LiMn₂O₄. However, inorganic materials are associated with large environmental footprints and resource restrictions. To overcome such problems efficiently, organic materials have recently attracted attention as alternative active materials because of their light weight, structural flexibility, and chemical tunability. H. Chen et al.

have suggested using electro-active carbonyl compounds, e.g., Li₂C₆O₆, synthesized from biomass because these materials show reversible capacities as high as 580 mAh g⁻¹ [3]. M. Lee et al. have proposed the use of flavin-based organic materials, e.g., lumiflavin, to imitate a cellular energy-transduction mechanism because these materials exhibit gravimetric capacities as high as 174.32 mAh g⁻¹ [4]. However, to date, organic material-related research has been performed using Swagelok- and coin-type cells (CR2016 or CR2032) on a laboratory scale to evaluate their electrochemical properties [3,4]. Additionally, positive electrodes have mainly been prepared by mixing the active material, conductive additives, and polytetrafluoroethylene (PTFE) binder, followed by compression onto a stainless steel or aluminum mesh current collector [5–8].

Tape casting is widely used to fabricate positive and negative electrodes in commercial lithium ion batteries ranging in size from small to large [9]. To obtain a slurry suitable for tape casting,

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poly(vinylidene fluoride) (PVDF) and *N*-methyl pyrrolidone (NMP) have been typically used as the binder and organic solvent, respectively, because PVDF binder has good electrochemical stability and high adhesion to electrode materials and current collectors [10]. Recently, switching from a non-aqueous- to an aqueous-based binder system, e.g., water-soluble carboxymethyl cellulose (CMC) and styrene butadiene rubber (SBR), was attempted to take advantage of the environmental friendliness and non-flammability of such a system [11–13].

The present study was performed to investigate the feasibility of scaling up flavin-based organic materials for use in positive electrodes and to determine the effects of an aqueous binder system on the structural stability of flavin-based organic materials. The electrochemical properties of large-sized pouch cells were also evaluated using two types of electrolyte.

2. Experimental

Lumichrome (7,8-dimethylalloxazine; Sigma-Aldrich) was used as the active material, and Super P™ Li carbon black (TIMCAL Graphite & Carbon) was used as the conductive agent. A mixture of carboxymethyl cellulose (CMC, DS = 0.7, $M_w = 250,000$; Sigma-Aldrich) and styrene butadiene rubber (SBR, BM-400B; Zeon Corporation, Japan) in a 1:1 weight ratio in distilled water was used as the binder.

The cathode electrode was prepared by mixing 40-wt% active material, 40-wt% conductive agent, and 20-wt% polymer binder. Lumichrome, Super P™ Li, SBR, CMC, and zirconia balls (diameter: 10 mm) were placed in a bottle with a ball-to-weight ratio of 8.0. Additional distilled water was added to obtain a mixture appropriate viscosity. Slurry mixing was performed on a planetary centrifugal mixer at a rotation speed of 750 rpm for 40 min. The mixed slurry was deposited onto an aluminum foil (area: 60 mm × 80 mm, thickness: 20 μm). The 30 μm thick coated electrodes were then dried in a vacuum oven at 80 °C for 24 h.

The surface morphology of the as-prepared electrodes was examined by scanning electron microscopy (SEM; XL-30 FEG, Philips). The absorption spectra of lumichrome were obtained with a UV-visible spectrophotometer (V-650; JASCO) from 700 to 270 nm at a resolution of 0.5 nm. The electrochemical measurements were performed at room temperature of 23 °C using the large-sized pouch cell (capacity: 80 mAh) with eight lumichrome cathodes and five lithium metal anodes. Fig. 1 shows the schematic diagram of the pouch cell structure and as-prepared pouch cell. The pure lithium metal foils (thickness: 0.3 mm, width: 80 mm; Hohen) were commercially available and used without further treatment. The electrolyte solutions were 1-M LiPF₆ dissolved in EC-dimethyl carbonate (DMC) (1:1 vol%; Panaxetec) and 1-M LiPF₆ in tetraethylene glycol dimethyl ether (TEGDME; Panaxetec). The large-sized pouch cell (area: 73 mm × 110 mm) was assembled at room temperature of 23 °C in a dry room with a dew point less than −53 °C using polyethylene film (16 μm thick) as a separator. 12 ml of the electrolyte solutions were injected into an aluminum pouch, then vacuum applied to improve electrolyte wetting. Assembly of CR2032 coin-type cells was also carried out in an argon-filled glove box with a dew point less than −65 °C. The assembled pouch cells were charged and discharged at different rates from 10 mA g^{−1} to 2000 mA g^{−1} between 1.5 and 3.5 V versus Li/Li⁺ (Toscat-3000; Toyo) to determine the rate capability and cyclability at room temperature of 23 °C. Electrochemical impedance spectroscopy (EIS, Solatron 1287; Solatron) was performed by applying a small perturbation voltage (5 mV) in the frequency range of 100 kHz to 100 mHz.

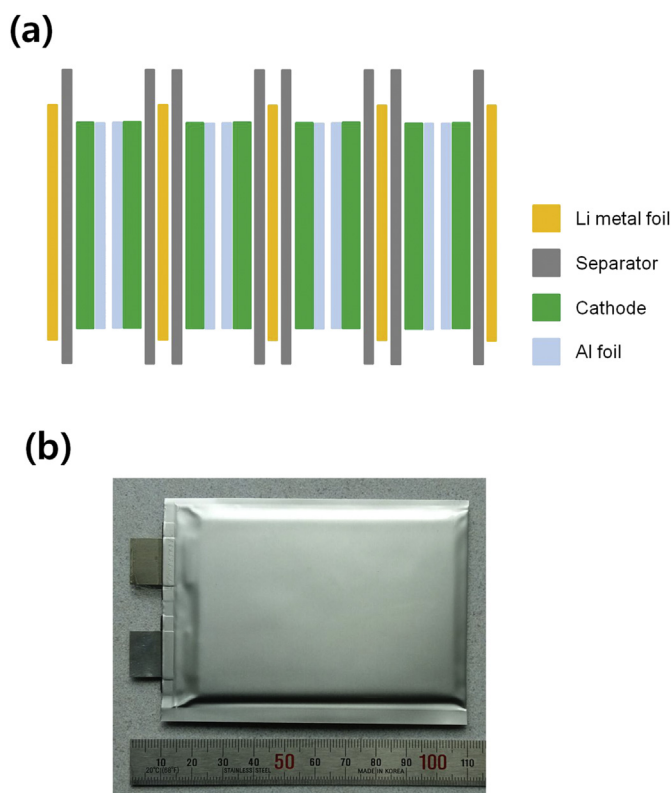


Fig. 1. (a) A schematic diagram of the pouch cell structure and (b) as-prepared pouch cell.

3. Results and discussion

Fig. 2 shows SEM images of the pristine lumichrome powder and as-prepared lumichrome cathode electrode. The average particle size of the flake-like lumichrome powder is approximately 1.0–2.0 μm. In the as-prepared cathode electrode, lumichrome particles are uniformly dispersed in the conductive agent, as shown by the arrow in Fig. 2(b).

UV-visible absorption spectroscopy is used to investigate structural changes associated with lumichrome during the electrode-fabrication processes. UV-visible absorption spectra of the pristine lumichrome powder and as-prepared lumichrome electrode in ethanol are shown in Fig. 3. The absorption spectra of the as-prepared lumichrome electrode are essentially identical to that of the pristine lumichrome powder. The two strong absorption bands, with maxima at 334 nm and 385 nm, are assigned to two independent π, π^* transitions of lumichrome [14]. It is evident that the fabrication processes used to produce the cathode electrode, which includes mixing, coating, and drying, are able to reliably preserve the original structural characteristics of the pristine lumichrome powder without any degradation.

Fig. 4 shows the charge/discharge profiles and differential capacity-voltage plots of large-sized pouch cells with two types of electrolyte, ethylene carbonate-dimethyl carbonate (EC-DMC) and TEGDME, and 2032-type coin cell with EC-DMC for the first cycle, which consists of a charge/discharge at a current density of 10 mA g^{−1} between 1.5 V and 3.5 V versus Li/Li⁺. EC-DMC and TEGDME injected pouch cells show discharge capacities of 142 mAh g^{−1} and 148 mAh g^{−1}, respectively. EC-DMC injected coin cell show discharge capacity of 149 mAh g^{−1}. As shown in Fig. 4(b), large-sized pouch cells and 2032-type coin cell exhibit two sets of cathodic and anodic peaks with average potentials of 2.58 V and

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