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Embossed aluminum as a current collector for high-rate lithium cathode performance



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HIGHLIGHTS

GRAPHICAL ABSTRACT

- Preparation of the embossed Al current collector.
- Effect of an embossing surface for Al current collector.
- Stable cycle life at high current density using an embossed Al current collector.

A facile strategy is introduced to fabricate an embossed Al current collector surface for rechargeable batteries.



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ABSTRACT

Al foil carefully prepared via an anodization process followed by chromate-phosphate treatment is evaluated for use as a current collector for a cathode powder prepared by the conventional electrode coating method. The formation of an embossed surface on the current collector imparts hydrophilic properties, thereby enhancing the adhesion capabilities of the cathode active material. In Li-ion cells where $LiCoO_2$ is employed as the active cathode material, the embossed Al foil is an electrochemically stable current collector when in contact with other cell components during the charge–discharge processes. This also produces a notable decrease in the polarization associated with the electrical disconnection between the active material and the Al current collector, which ultimately results in a stable cycle life at high current densities.

1. Introduction

Energy storage devices are attracting increasing attention due to strengthening regulations regarding global warming, the depletion of petroleum resources, and demands for new national growth engines [1]. For example, Li-based rechargeable batteries have been widely employed in portable electronic devices because of their high energy densities, lack of memory effect, excellent cyclability, and long-term preservation. They have also been developed for applications such as electric vehicles (EVs) and large energy storage systems (ESSs) [2,3]. However, as currently commercialized Li-ion batteries will soon reach their performance limits as typical intercalation-type electrode materials, the search for alternative active materials to replace commercial layered oxides and carbonaceous materials for Li-ion batteries is an ongoing challenge, with new battery systems such as Na-ion, Mg-ion, and Li-S batteries having been proposed [4–7]. However, such battery systems based on new alternative active materials must be configured to their appropriate electrolyte compositions and separators, in

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addition to any other battery components. Furthermore, such novel battery systems are in the early stages of development, and so they require considerable research time and cost.

In addition to the development of novel core materials, battery performances can be improved by enhancing the energy or power density through a reduction in the weight or volume of the various battery components (i.e., current collectors, lead taps, and external casing) [8,9]. In particular, the development of thinner adhesive cathode and anode current collectors that serve as bridges for electron transfer to/from the laminated active material film can improve battery performances while using existing core materials. However, the application of novel current collectors to commercialized batteries is only possible when adequate mechanical properties, electrochemical/chemical stabilities, and heat dissipation capacities can be ensured, as significant amounts of resistance are generated between the laminated active material film and the current collector interface during the charge-discharge process [10-13]. Although the current collector has a direct influence on the characteristics of the cell to which a high power and/or energy density characteristic is to be imparted, no significant attention has been paid to this component, as it appears to be a conventional topic that does not give any noticeable or innovative changes to battery technologies.

We previously reported the preparation of crater-like Al foil surfaces through a simple wet chemical etching method and investigated the effect of surface morphology on its electrochemical performance as a cathode current collector [14]. By extension, this leads to the consideration of Al current collectors with embossed surfaces through an anodization synthetic process. It should be noted that Al has a distinctive property in that a porous structure can be formed on the surface by anodizing under the appropriate conditions [15,16]. More specifically, the anodization of Al produces a thin Al oxide layer along a selforganized hexagonal array of parallel pores that are aligned perpendicular to the thin film surface. Self-aligned hexagonal arrangements with well-controlled pore diameters and interpolation distances are thereby obtained by careful selection of the process conditions, including the acidic electrolyte concentration, anodizing time, anodizing potential, current density, and temperature [17]. In particular, porous anodized Al surface structures provide great potential for the production of surfaces with special wettabilities (i.e., conversion from superhydrophobic to superhydrophilic) due to controllable pore dimensions and adjustable surface chemistries [18]. Thus, we herein investigate the correlation between an Al current collector with a carefully controlled embossed surface and the electrochemical characteristics of the resulting Li-ion battery.

2. Experimental

The surface treatment procedure for preparing the embossed structure of the Al current collector is illustrated in Fig. 1. More specifically, AA1100 alloy Al foil (11 µm thickness, DONG-IL Aluminum Corp.) treated with Corona to remove the remaining grease was used as a starting material. The clean Al pieces were cut into $15 \text{ cm} \times 6 \text{ cm}$ pieces and immersed in a 0.3 M solution of oxalic acid ($C_2H_2O_4$, > 98%, Alfa Aeser) with a parallel electrode arrangement using Pt mesh as the cathode. A mild anodization process was performed in potentiostatic mode using a DC power supply (TDP-1005 B) applying constant anodization voltages of 30 V to the electrolyte while maintaining the cell temperature at 25 °C for either 5 or 10 min. The alumina layer was then removed from the Al substrate using a mixture of 4 vol% chromic oxide (CrO₃, > 99%, Sigma Aldrich) and 6 vol% phosphoric acid (H₃PO₄, 85%, Alfa Aeser) in deionized water (DI water) at 25 °C over 12 h, to give a highly ordered concave pattern on the sample surfaces. The anodized Al pieces were subsequently rinsed with DI water and dried in an oven for several hours prior to characterization.

Five specimens were prepared and analyzed for each sample of Al pieces. For the tensile strength measurements, Al samples measuring

10 cm \times 1.27 cm were tested on a universal testing machine (UTM, Shimadzu AG-X), according to the standard procedure described in ISO 6892. The foil thickness was measured using a Dektak 150 Alpha Step Surface Profiler. Phase analysis was performed using a D8-Bruker X-ray diffractometer (XRD) with Cu K α radiation. The morphologies and microstructures of the samples were examined by scanning electron microscopy (SEM, Tescan Mira LM), while the water contact angles were measured using a Phoenix 300 apparatus, and the reported values were averages calculated from five measurements at different parts of the surface. The surface roughness was observed by atomic force microscopy (AFM, Shimadzu SPM-9700) in dynamic mode.

The cathodes were prepared with active material (LiCoO₂), carbon black (super P), and polyvinylidene fluoride (PVDF) binder mixed in Nmethylpyrrolidone (NMP) at a weight ratio of 8:1:1. In order to exclude the electrochemical performances in the current collector according to the characteristics of the cathode material, we used the most commonly attested LiCoO₂ as cathode active material. The mixture was coated on the anodized Al foils, and drying at 120 °C for 2 h under vacuum. Cointype battery-test cells (CR2032) were assembled in an argon-filled glove box, using Celgard 2400 polypropylene separator, Li metal anode, and 1 M LiPF₆ in ethylene carbonate (EC)/ethyl-methyl carbonate (EMC) (1:2 v/v) as the electrolyte (Panax Etec). The charge-discharge experiments were performed galvanostatically at a constant current density of 30 mA g^{-1} (with regard to the active material) in the voltage range of 3.5–4.5 V vs. Li⁺/Li. Electrochemical Impedance Spectroscopy (EIS) was carried out with ZIVE SP2 equipment by applying a 10 mV amplitude signal in the frequency range from 10 kHz to 0.01 Hz. Then, active material content of $\sim 2 \text{ mg}$ served as the working electrode and lithium foil served as the counter and reference electrodes. The impedance response was measured after the 20th cycles, and the data were fitted with the ZMAN software.

3. Results and discussion

Electrode coating involves the coating of a prepared slurry on a metal current collector with a predetermined pattern and thickness using a coater head, followed by subsequent drying. This process consists of unwinding, coating, drying, density measurement, and winding processes. As such, physical properties such as the tensile strength of the current collector are essential in electrode manufacturing processes that employ roll-to-roll coating. Fig. 2a shows the tensile strengths of the prepared Al foils following surface treatment. More specifically, sample 1 prepared using chromate-phosphate treatment after anodization at 30 V for 5 min exhibited a tensile strength of \sim 235 MPa, which was lower than that of the pristine Al foil (i.e., 245 MPa). In addition, increasing the Al anodization time to 10 min to give sample 2 further lowered the tensile strength to \sim 227 MPa. We expected that the tensile strength gradually reduced as the anodization time increased as Al atoms broke away from the surface to form the embossed structure. However, despite this lowering of the tensile strength, which has a detrimental effect on the electrode coating process, an acceptable value of \sim 227 MPa was obtained. A similar reduction in tensile strength was also observed upon lowering the anodization voltage, as shown in Fig. S1. Furthermore, the variations in thickness of the surface-treated Al samples are given in Fig. 2b, where all thicknesses were lower than that of the pristine Al foil (i.e., 11 µm), and decreased upon increasing the anodization voltage and time, as in the case of the tensile strength (see Fig. S1).

With the aim of determining the crystal structures of the surface treated Al samples, XRD was employed, and the results are outlined in Fig. 3. For each sample, all reflections were indexed based on the cubic Al structure ($Fm\bar{3}m$, JCPDS no. 89-2837). In the case of the surface-treated samples, no peaks corresponding to an aluminum oxide surface impurity were observed, and the intensities of all Al peaks were slightly decreased. This indicates that the anodized aluminum oxide (AAO) formed through the Al anodization process was completely removed by

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