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High-performance lithium-ion batteries with $1.5 \mu m$ thin copper nanowire foil as a current collector

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- \bullet Cu NW foil as thin as ~1.5 µm used as a lithium ion battery current collector.
- Graphite-CuNW foil anode exhibits electrochemical performance.
- 3 mAh/cm² full cell exhibits 83.6% capacity retention for 600 cycles.
- 700 mA pouch-type battery with graphite-Cu NWs foil anodes is fabricated.

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1. Introduction

With the advances in technology, the escalation of dependence on cordless power source is greatly considered. Rechargeable lithium-ion batteries (LIBs) have become the most universal energy storage system in modern portable electronics and full/hybrid

GRAPHICAL ABSTRACT highlights graphical abstract

ABSTRACT abstract

Cu Foil, a thin sheet of Cu, is the common anode current collector in commercial lithium ion batteries (LIBs) which accounts for ~ 10 wt% of the total cell weight. However, thickness reduction of LIB-based Cu foils below $6 \mu m$ has been limited by the incapability of conventional rolling annealing or electrodeposition process. We here report a new type of Cu foil, so called Cu nanowire foil (CuNW foil), for use as an LIB anode current collector. We fabricate Cu NW foils by rolling press Cu nanowire fabric to reduce the thickness down to ~1.5 μ m with an areal weight down to ~1.2 mg cm⁻² and a density approximately 96% to that of bulk Cu. The rough surface and porous structure of CuNW foil enable better wetting and adhering properties of graphite slurry on foil. In full cell examination, a cell of a areal capacity of 3 mAh cm⁻² exhibits 83.6% capacity retention for 600 cycles at 0.6 C that meets the standard specification of most commercial LIBs. As a proof-of-concept of demonstration, we fabricate a 700 mA pouchtype battery implemented with graphite-Cu NWs foil anodes to serve as energy supply to operate electronic devices.

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electric vehicles due to their high energy density, high working voltage, and long cycle life $[1-7]$ $[1-7]$ $[1-7]$. A LIB is an electrochemical system consisting of anode, cathode, separator, current collector, and electrolyte. To accommodate the tendency of the electronic devices that requires lighter weight and longer battery life, a major effort has been made toward increasing energy density associated with a light-weighting design of batteries [\[8\]](#page--1-0). The battery energy density could be increased by strategies such as introduction of high capacity electrode materials, increase of the contact area of active Email address hutun@che pthy odu by (H V Tupp)
Email address hutun@che pthy odu by (H V Tupp) materials/electrolyte interface, and reduction of the weight of

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battery components $[9-18]$ $[9-18]$ $[9-18]$.

Cu foil is the common anode current collector in standard LIBs which accounts for approximately 10 wt% of a cell's weight (based on a foil with a ~9 mg cm^{-2} in areal density and ~10 μ m in thickness) [\[19\]](#page--1-0). The use of thinner and lighter weight of Cu foil collector can directly increase the energy and volume density of LIBs. Conventional methods to fabricate commercial LIB Cu foils are mainly rolling-annealing (RA) and electrodeposition (ED) process [\[20\].](#page--1-0) However, both methods come at the challenges regarding the issues of the cost, strength and quality control. RA-Cu foil (Fig. 1) is made by annealing the Cu ingots and then rolled to thin, flat Cu sheets. The disadvantages of the RA Cu foils are the available thickness limited to $6 \mu m$ associated with the increased cost of fabrication process. The ED Cu foils $(Fig. 1)$ are fabricated by electroplating the Cu onto a rotating drum from electrolytic solution. The cost of ED foils is lower than RA foils, but the mechanical strength is weaker than RA foils. In addition, prior to usage, both the surfaces of ED- and RA-foils need further treatments to obtain chemically passivated rough surfaces for enhanced slurry adhesion and oxidation prevention [\[20\]](#page--1-0).

Recently, nanostructure-based current collectors have been reported, including metal foam $[21-23]$ $[21-23]$ $[21-23]$, metal array $[24,25]$, graphene sheet $[26,27]$, CNT network $[28-30]$ $[28-30]$ $[28-30]$ and Cu based nanomaterials (nanoparticles [\[31\]](#page--1-0), nanorods [\[32,33\],](#page--1-0) and nanowires (CuNWs) $[34-38]$ $[34-38]$ $[34-38]$). Nanostructured current collectors can enhance the adhesion of active materials and the penetrability of electrolyte, and release the strain arisen from the volume change during the lithiation/delithiation process [\[24,36\]](#page--1-0). However, much of uncertainty about their applicability for practical uses have been raised by the following points: (1) the resistivity of nanostructure current collector is too high [\[26\];](#page--1-0) (2) the complicated manufacturing process increases the scale-up difficulty of products; (3) the weight loading in active materials is very low due to incompatibility to conventional slurry-deposited process, resulting in small amount of energy stored in electrodes for practical applications; (4) most electrodes with nanostructures-based current collectors have low practical volume capacity due to their porous structures and low volume density.

Herein, ultra-thin, flexible, light-weight and area-scalable CuNWs foils are fabricated as current collectors for LIBs. The thickness-tunable and density-uniform CuNW foils were made by rolling press drop-casted CuNW fabric (Fig. 1). The anodes made by depositing graphite slurry on CuNW foil current collector (graphite-CuNW foil anode) were assembled to half and full cells to examine the electrochemical performances. The graphite-CuNW foil anode exhibits excellent electrochemical performance during the charge/discharge cycling, even at high loading amount in

Hot rolling

Rolling-annealing

 (a)

 (b)

graphite. The full cell with an areal capacity of 3 mAh g^{-1} remains 80% of the capacity retention over a 600 cycles-period. Moreover, CuNW foils are area-scalable via the foil-making technology. Largearea CuNW foils could be implemented into pouch type full batteries to offer large output capacity. We demonstrate batteries using graphite-CuNW foil anodes can output current > 100 mA h to provide sufficient electricity to power various electronic devices requiring various working conditions, such as light up LED arrays, charge smart phones, power cordless screwdrivers and drone to demonstrate the multiusability in different requirements.

2. Experimental section

2.1. Materials

Copper chloride (CuCl, 99.9%) and oleylamine (70%) were purchased from Sigma-Aldrich. Hexane (ACS) was purchased from Marcon. Graphite powder was purchased from Long Time Technology. Lithium Hexafluorophosphate (LiPF6), electrolyte (1 M LiPF6 in ethylene carbonate (EC): dimethyl carbonate (DMC) 1:1) (v/v), Nmethyl-2-ptrrolidone (NMP), poly(vinylidone difluoride) (PVDF), super p carbon black, copper metal foil, lithium metal foil, celgard membrane, coin-type cell CR2032 and laminated aluminum film were purchased from Shining Energy. Commercial Li(NiCoMn) $O₂$ cathode electrodes was purchased from Vista Advance Technology. All the chemicals were used without further purification.

2.2. Synthesis of oleylamine-coated copper nanowires

CuCl (1.2 g) and OLA (100 ml) were added to a 100 ml three-neck flask in glovebox. After connected to schlenk line, the solution in flask was stirred magnetically and purged with argon. Until whole of CuCl powder was dissolved, the solution was heated to 110 \degree C to remove the residual oxygen and water. The color of solution changed from green to yellow after 60 min of heating, and then the temperature was risen to 260 \degree C and held for 120 min. During this process, the color of solution slowly changed from yellow to reddish. Afterward, the reaction system was cooled to room temperature by water quenching. The reaction mixture was dispersed in hexane and centrifuged at 8000 rpm 5 min for several times until the suspension become clear, and the CuNWs precipitate was collected and dried. The CuNWs after purification were stored in a vial under argon conditions to prevent oxidation.

2.3. Fabrication of CuNW fabric and CuNW foil

 9 mg cm $^{-2}$

1.2 mg cm^{-2}

The CuNW fabric was prepared by drop casting method.

 \prod 10 μ m

 $\pm 1.5 \mu m$

Surface

Electrodeposition

Fig. 1. Schematic of the process of (a) conventional Cu foil fabrication process using rolling-annealing or electrodeposition methods and (b) CuNW foil fabrication process using a rolling press method.

Commercial Cu foil

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