



Featured Letter

Nano-porous copper metal current collector for lithium ion batteries

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ABSTRACT

A low-cost and scalable ultrasonic synthetic method to produce copper metal current collector with uniform nanopores on the surface is achieved for the first time. A layer of silicon active material was sputtered on the surface of as-prepared metal current collector (vs. metal lithium) as an innovative current collecting material for lithium ion battery. The assembled battery with this current collector showed an areal capacity of 2.2 mAh/cm², comparable to that of commercially available lithium ion batteries. Due to the nano-porous structure of current collector, the silicon active material electrochemical capacity retention and cycling stability also improved significantly.

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1. Introduction

Safety and fast charging/discharging properties of lithium ion battery are highly desirable for purposes ranging from portable devices to electric vehicles. Metal films with dimensionally arrayed nanopores on the surface have been long sought-after in the quest for high-surface-area conductive current collectors [1–5] and catalytic materials [1,2]. Recently, it has been reported that electrodes with *meso*-structure produce high power batteries with ultra-fast charging/discharging property (within one minute [6]). In lithium ion battery field, porous structures can provide sufficient inner free space to absorb large volume expansions and thereby enhance the cycling stability, and are favourable for fast transport of Li⁺ to increase the rate performance. However, turning block metals into metal current collectors with dimensionally arrayed nanopores on the surface economically remains a significant challenge.

There are several well-established approaches for preparing nano-porous metal films: notably through de-alloying, recently through deposition of metals on nanostructured templates and laser etching, etc. There has been an ongoing search over the past decades, but no commercial metal nano-porous network film is available yet.

Ultrasound can generate oscillating bubbles and lead to extremely rapid heating or cooling rates, as well as pressures of up to several hundred mega-pascals in transient cavities while the bulk

conditions of the liquid solution remain at ambient temperature. In 2010, Ekaterina V. Skorb's group reported that a metal aluminium film with nanopores on the interface was obtained in pure water through ultrasound treatment using ultrasound cell disrupting equipment [2]. However, it is difficult to make nanopores on a large piece of metal through ultrasound probe impacting due to the size limitations of the probe.

In this paper, we report an ultrasonic synthetic method for producing metal current collector with uniform nanopores on every interface and their electrochemical performance as current collectors in lithium ion batteries. To our best knowledge, uniform formation of nanopores on metal surface through a combined ultrasonic and chemical etching method has not been reported. There are also no commercial lithium ion batteries adopting metal current collectors with uniform nanopores on the surface.

2. Experimental

Rolled metal copper films (10 × 10 cm², 10 μm in thickness) as raw materials were transferred to a glass beaker filled with dilute ammonia solution. Then the rolled copper films in ammonia solution were put into ultrasonic cleaner to stir for 30 min at room temperature. In this paper, the ultrasonic chemical treated copper film will be named as nano-porous copper film. After drying, morphology of copper films was observed using Scanning Electron Microscope (SEM). Energy Dispersive X-ray analyses (EDX) were obtained with an EDAX detector. X-ray diffraction (XRD) scans were acquired with a Bruker D8 Advance system. Rolled metal

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copper films and nano-porous copper films were used as substrates. Silicon layers were sputtered to metal copper films with a Denton sputtering (Discovery system) machine using DC Mode at 200 W power in Ar gas atmosphere for 30 min.

Electrochemical performance of metal copper films and nano-porous copper films (in an area of $1 \times 1 \text{ cm}^2$) as current collectors was investigated through a CR2032 coin cell (half-cell) with sputtering silicon as the active cathode material, metal lithium as the reference electrode and counter electrode, and Celgard film as separator. The electrolyte was 1.0 M LiPF_6 in ethylene carbonate and diethyl carbonate with volume ratio of 1/1. To compare electrodes properties, electrochemical performance was evaluated using Neware battery tester at room temperature.

3. Results and discussion

3.1. Preparation of nano-porous copper metal current collector from metal copper film

Usually defects on metal copper surface and subsurface can occur in various forms, e.g., folds, splashes, cracks, inclusions, and voids. The voids are caused by gas porosity, shrinkage porosity, and shrinkage cavities when metal copper is manufactured. In the atmosphere, the surface of copper oxidizes over the course of years to become a mixture of green basic salts, the patina, consisting mainly of the basic sulphate, with some basic carbonate.

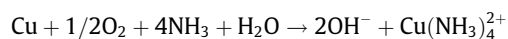
Fig. 1 shows the surface morphology as-received and after ultrasonic chemical treatment of rolled metal copper films. The as-received rolled metal copper surface (Fig. 1a and b) is smooth and flat visually. After ultrasonic chemical process treatment, uniform nano-holes appear on the surface of rolled metal copper film as shown in Fig. 1c and d. These nanopores on the surface are very

similar to dimensionally arrayed nanopores prepared by nanostructured polymer templates method [5]. The size of nano-holes might increase with processing time and ammonia solution concentration. The details of copper surface morphology reveal the sizes of nano-hole varying from 80 to 800 nm as shown in Fig. 1c and 1d.

The compositions of these films are shown in Fig. 2a and b respectively. EDX analysis does not reveal any obvious compositional difference (with more than 97 at.% of copper and less than 3 at.% of oxygen) between the copper films as-received and after ultrasonic chemical treatment. Thus ultrasonic chemical treatment process does not change the composition of copper film. The nano-porous copper film looks darker than the metal copper film due to the induced nano-porous structure. As shown in Fig. 2c, Brunauer–Emmett–Teller (BET) tests show that adsorption cumulative surface area of nano-porous copper film is obviously larger than that of rolled metal copper film ($0.499 \text{ m}^2/\text{g}$ vs $3.896 \text{ m}^2/\text{g}$).

Metal aluminium films with nanopores on parts of the interface as reported by the Ekaterina V. Skorb group are formed in purified water through ultrasound probe. Our method differs greatly from that of the Ekaterina V. Skorb group in reaction mechanisms [1,2] and the interface result. Through our method, nanopores were generated on every interface of copper film. They are not as the same as nanopores created using the probe cup-horn system from which ultrasound is attenuated rapidly, and only a few centimeters are sonochemically active [1,2].

The reaction mechanism of our method is as follows:



Because of low solubility of oxygen gas in water, the rate of dissolution is proportional to the surface area of the copper, and, at low oxygen pressures, is mainly determined by the transport step

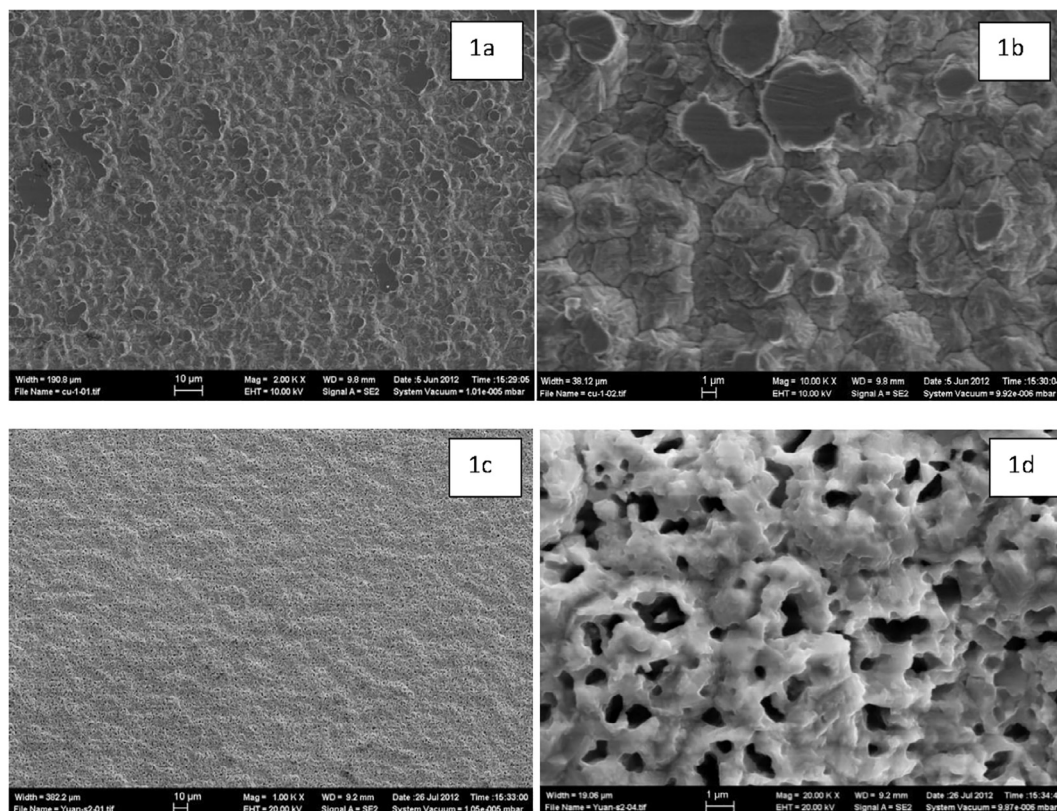


Fig. 1. SEM images of as received rolled metal copper film at low magnification (1a) and high magnification (1b); SEM images of rolled metal nano-porous copper film with ultrasonic chemical treatment at low magnification (1c) and high magnification (1d).

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