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# Single wall carbon nanotube paper as anode for lithium-ion battery

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**Abstract**

"Free-standing" single wall carbon nanotube (SWNT) papers have been synthesised by simple filtration method via positive pressure. A conventional SWNT slurry coated electrode was fabricated to compare with the SWNT papers. The results show that the capacity of the "Free-standing" electrode was slightly lower than that of the conventional electrode, but the "Free-standing" electrode was produced without any binder, and metal substrate, so that the weight of electrode was reduced significantly. On the other hand, the procedures for SWNT electrode preparation were simplified, so the cost of the manufacturing could be reduced.


*Keywords:* Single wall carbon nanotubes; Free-standing electrode; Lithium-ion battery; Impedance; Capacity

## 1. Introduction

Recent developments in the field of portable equipment have been the main driving force behind the search for batteries with high energy density and form flexibility [1,2]. It is well known that lithium batteries have the highest energy density of all the commercialised rechargeable batteries [2–4]. They typically consist of a positive electrode and a negative electrode spaced by a separator, an electrolyte, a case and feed-through pins, respectively, connected to the electrodes and extending externally from the case. Each electrode is formed from a metal substrate that is coated with a mixture of an active material, an electrical conductor, a binder and a solvent. The negative electrode is formed from a copper substrate with graphite as the active material. The positive electrode is typically formed from an aluminium substrate with lithium cobalt dioxide as the active material.

Carbon nanostructures are of tremendous interest [5,6], from both a fundamental and an applied perspective. The applications investigated include using carbon nanotube as an anode for lithium-ion batteries [7,8]. However, limited research work [2] has been done on the fabrication and evaluation of a "Free-standing" carbon nanotube paper electrode without any electrode substrate that is produced with a simple filtration method via positive pressure. Note that the manufacturing process for the "Free-standing" electrode is considerably simplified compared with that of the conventional electrode in which a metal substrate is coated with a mixture of an active material, an electrical conductor, a binder and a solvent.

In this work, we aim to develop a unique and simple electrode making technique with a view to improving specific energy density and simplify manufacturing procedures for lithium-ion batteries, and consequently reducing the manufacturing cost.

## 2. Experimental

### 2.1. Preparation of single wall carbon nanotube paper

A standard SWNT/Triton X-100 dispersion was prepared via addition of 50 mg of high purity (95%, with remainder, ionic iron (Fe) impurities from catalyst source) single-walled carbon nanotubes (SWNTs) (Lot #PO257) supplied by Car-

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bon Nanotechnologies Incorporated (CNI), USA, and 0.5 g of a surfactant, Triton X-100, into 50 ml of Milli-Q water followed by ultrasonication for 2 h. A polyvinylidene fluoride (PVDF) membrane with pore size of 0.22 μm was cut into a 4-cm disk to fit the filtration cell. The membrane acts as a filter paper, and the membrane was wetted in 50:50 (v/v) Milli-Q water to ethanol solution for 30 min. Passing the prepared SWNT suspension through the wetted PVDF filter in a filtration cell under a positive nitrogen gas pressure of 400 kPa produced a "Free-standing" mat of entangled SWNT ropes, and known as "bucky paper". Subsequently, the resultant SWNT mat was washed with 200 ml of Milli-Q water followed by 100 ml of methanol, where methanol was used to remove any residual surfactant. Finally, the SWNT mat was peeled off from the PVDF filter after letting it dry overnight in a vacuum oven. Meanwhile, for the SWNTs paper containing carbon black (CB), the dispersion was prepared by substituting 10 wt.% of the SWNTs with carbon black. Then, the same procedures were as applied in the fabrication of SWNT paper were repeated to produce the composite paper of SWNTs with carbon black. The thickness of the SWNT papers was controlled by the concentration of the SWNT suspension.

### 2.2. Sample characterization

Single wall carbon nanotubes were observed under a transmission electron microscope (TEM, JEOL 2010). Single wall carbon nanotubes powder was characterised by X-ray diffraction (XRD) on a Philips PW1730 diffractometer with Cu Kα radiation. Raman spectroscopy was used to monitor the variations of the precursors and SWNT papers using a JOBIN YVON HR800 Confocal Raman system.

Resistance measurements of the SWNT papers were performed on long strips using the ASTM four-probe technique. A dc current of 0.5 mA was applied across the two electrodes using an EG&G PAR 363 and the voltage drop across the two inner electrodes was measured using a HP multimeter (model 34401A).

Morphologies of the "Free-standing" SWNT electrodes before and after cycling were examined using a Leica Model Stereoscan 440 scanning electron microscope.

### 2.3. Electrochemical measurements

The prepared "Free-standing" SWNT papers were cut to 1 $cm^2$ and used as electrode directly. In order to compare the performance of "Free-standing" SWNTs with conventional slurry coated SWNT electrodes, the SWNT electrodes were made by dispersing 90 wt.% SWNT powder and 10 wt.% PVDF binder in dimethyl phthalate solvent to form homogeneous slurries. The slurries were spread on Ni foam substrates. The coated electrodes were dried in a vacuum oven at 100 °C for 20 h and then pressed.

The electrochemical experiments were carried out using coin cells. CR 2032 coin-type cells were assembled in an Ar-filled glove box (Mbraun, Unilab, Germany) by stacking a porous polypropylene separator containing liquid electrolyte between the SWNT electrodes and a lithium foil counter electrode. The electrolyte was 1 M $LiPF_6$ in a mixture of ethylene carbonate (EC) and dimethyl carbonate (DMC) (1:1 by volume, provided by Merck KgaA, Germany). The cells were galvanostatically charged and discharged in the range of 0.0–2.0 V at a current density of 0.08 mA $cm^{-2}$.

The impedance was measured with an EG&G Model 6310 Electrochemical Impedance Analyzer (Princeton Applied Research) run by Model 398 software within a frequency sweep range of 100.00 kHz–0.01 Hz.

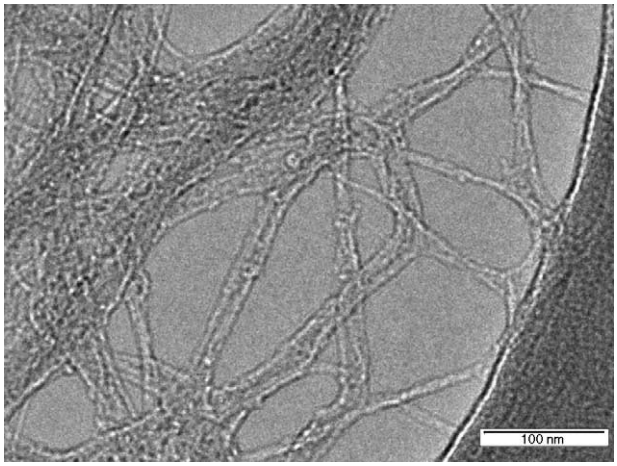


Fig. 1. TEM image of SWNTs powder.

## 3. Results and discussion

The TEM image of the single wall carbon nanotube powder in Fig. 1 shows a web of hollow tubes with outer diameter 10–30 nm. The XRD pattern of the SWNTs used as the precursor of carbon nanopaper is shown in Fig. 2. The broad

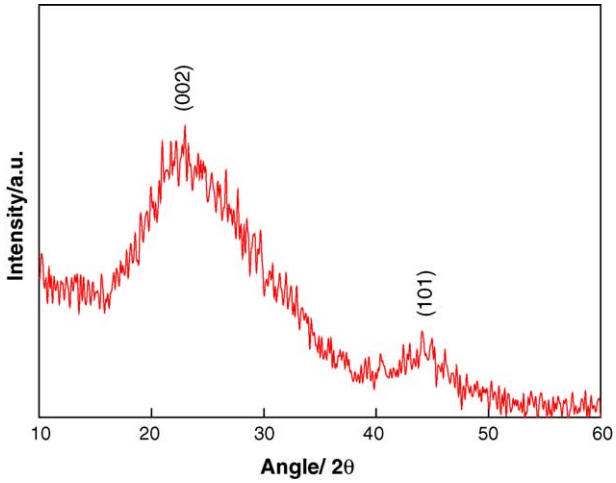


Fig. 2. X-ray diffraction pattern of single wall carbon nanotube powder.