Supporting Information

Polyimide-wrapped carbon nanotube electrodes for long cycle Li-air batteries

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Experimental Details

Preparation of PI-coated CNTs: The formation of a polyimide (PI) coating layer on the surface of CNTs was achieved by following a previously reported method [1]. For this, a polyamic acid solution was first prepared under a nitrogen atmosphere using dimethyl acetamide (DMAc) as a solvent to give a PMDA/ODA molar ratio of 1.00/1.01. Commercial CNTs (CNT-90, Hanwha chemical) were then soaked in this 1.0 wt% polyamic acid solution for 20 min at room temperature while stirring. The CNTs were then filtered from the solution and dried at 85 °C for 8h. To convert the polyamic acid coating the CNTs into PI, the CNTs were thermally treated under a nitrogen atmosphere via a stepwise imidization process (60 °C for 30 min \rightarrow 120 °C for 30 min \rightarrow 200 °C for 60 min \rightarrow 300 °C for 60 min \rightarrow 400 °C for 10 min). The microstructure of the pristine and PI-coated CNTs were observed using transmission electron microscopy (TEM, AP tech TECNAI G2 F30 S-Twin), and the surface composition of the CNTs was investigated by energy-dispersive spectroscopy (EDS).

Preparation of air electrode: An air electrode was prepared by mixing 90 wt% of CNTs

(pristine and PI coated) with 10 wt% of binder (PVDF) to give an electrode loading weight of $0.5 \text{ mg} \pm 0.05 \text{ mg}.$

Electrochemical testing: The electrochemical performance of the electrodes was examined using a modified Swagelok cell consisting of an air electrode, a metallic Li anode, a Whatman glass filter separator and an electrolyte made from 1 M of LiTFSI in TEGDME. The cells were assembled in an Ar-filled glove box and were subjected to galvanostatic cycling using a charge–discharge system. All experiments were conducted under an ambientpressure O_2 atmosphere.

Characterization of the electrode: X-ray diffraction (XRD, Philips) was used to analyze the reaction products formed on the surface of the electrodes during their initial cycles. Impedance measurements were carried out using an electrochemical workstation (AMETEK, VersaSTAT 3) by applying an AC voltage with an amplitude of 5 mV over a frequency range of 0.1 Hz to 100 KHz. Scanning electron microscopy (SEM, AP tech TECNAI G2 F30 S-Twin) was employed to observe the surface morphology of the electrode during cycling, and Fourier transform infrared spectroscopy (FTIR) spectra were also collected from the electrodes using a FTIR-4200 (JASCO) to ascertain the reaction products accumulated during cycling.

Reference

1. J.-H. Cho, J.-H. Park, M.-H. Lee, H.-K. Song and S.-Y. Lee, *Energy Environ. Sci.*, 2012, **5**, 7124

Figure S1





Voltage (V)

3.5

3.0

2.5

2.0

300

600

Capacity (mAh/g _{electrode})

3.5

3.0

2.5

2.0

@ 65th cycle

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Pristine electrode

1200

1500

PI electrode

(S1) (a) Discharge-charge profiles of a during cycling; pristine electrode (b) discharge-charge profiles of a PI electrode during cycling; (c) 30th discharge-charge profiles of both electrodes; (d) 65th dischargecharge profiles of both electrodes.

Figure S2



(S2) FTIR spectra of pristine and PI-coated electrodes after 50 cycles. (a) 500-700 cm⁻¹; (b) 1400-1700 cm⁻¹.

Figure S3



(S3) Nyquist plots of cells employing pristine and PI electrodes obtained (a) before testing– and (b) after 50 cycles (charged state).