Electronic Supplementary Information

Entrapping electrode materials within ultrathin carbon nanotube network for flexible thin film lithium ion batteries

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Estimation of CNT wt% in the thin film electrode:

LFP or LTO powders were spray-painted on a 1×1 cross-stacked CNT film and this procedure was repeated for 5 times to make each sample. Another 1×1 CNT film covered the top surface. Thus, each electrode contained 12 (2×6) single-layer CNT films. The size of each sample was 3 cm × 3 cm, equal to a total area of 9 cm². The areal density of single-layered CNT film was 0.002 mg cm⁻² according to Ref. 1. Thus, the mass of CNT in each sample could be calculated as 0.216 mg. In this work, total spray-painting time for LFP/CNT and LTO/CNT was 20 minutes and 10 minutes, respectively. These conditions resulted in an average mass of 4.62 mg for LFP/CNT and LTO/CNT was 4.62 % and 4.67 %, respectively.



Fig. S1 (a) Dependence of total mass loading of LFP or LTO powders in CNT films on painting time. (b) & (c) Photographs of LFP/CNT and LTO/CNT samples, which are suspended on square Teflon frames, prepared at conditions circled in (a).



Fig. S2 SEM images of thin film electrodes prepared with the spray painting method. (a) & (b) Top view of LTO/CNT electrode in high and low magnifications, respectively. LTO particles were embedded in the CNT film and entrapped by a bunch of CNT yarns (b). (c) Cross-sectional SEM image. The sample was cut by a razor.



Fig. S3 CV profiles of (a) LFP/CNT and (b) LTO/CNT at 0.5 mV s⁻¹ scan rate.



Fig. S4 Thickness of an LFP/LTO thin film battery measured in cross-sectional SEM image. The carbon tape was presented on the left as a reference.



Fig. S5 CV profiles of LFP/LTO full battery prepared with the spray-painting method at various scan rates.



Fig. S6 Cycle performance measured at 100 μ A cm⁻² current density for a 180° bent battery.



Fig. S7 Post-cycle microstructure images by SEM for LFP/CNT (a and b) and LTO/CNT (c and d) electrodes.



Fig. S8 (a) Illustration of connecting 3 LFP/LTO batteries in parallel. The parallel configuration was arranged by point-welding Ni tab (not drawn) to all CNT papers attached to LFP electrodes for the positive pole, and to those attached to LTO electrodes for the negative pole. (b) Areal specific capacity measured at 1000 μ A cm⁻².

1 K. L. Jiang, J. P. Wang, Q. Q. Li, L. Liu, C. H. Liu and S. S. Fan, *Adv. Mater.*, 2011, **23**, 1154