ADVANCED ENERGY MATERIALS

Supporting Information

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Highly Conductive, Light Weight, Robust, Corrosion-Resistant, Scalable, All-Fiber Based Current Collectors for Aqueous Acidic Batteries

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Key Words: Aqueous Batteries, Carbon nanotube, Cellulose nanofiber, Light weight current collector, Corrosion free

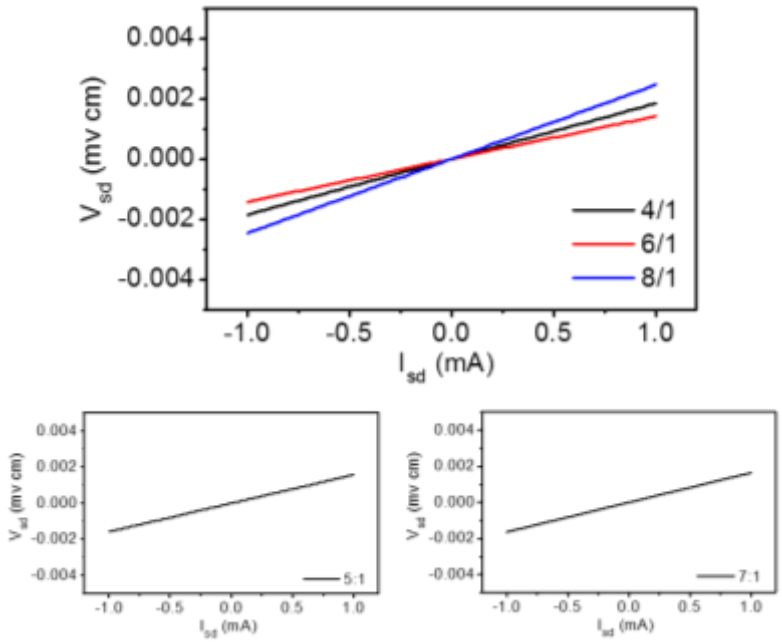

Figure S1. I-V curves of CNT-CNF films with different CNT/CNF weight ratios.

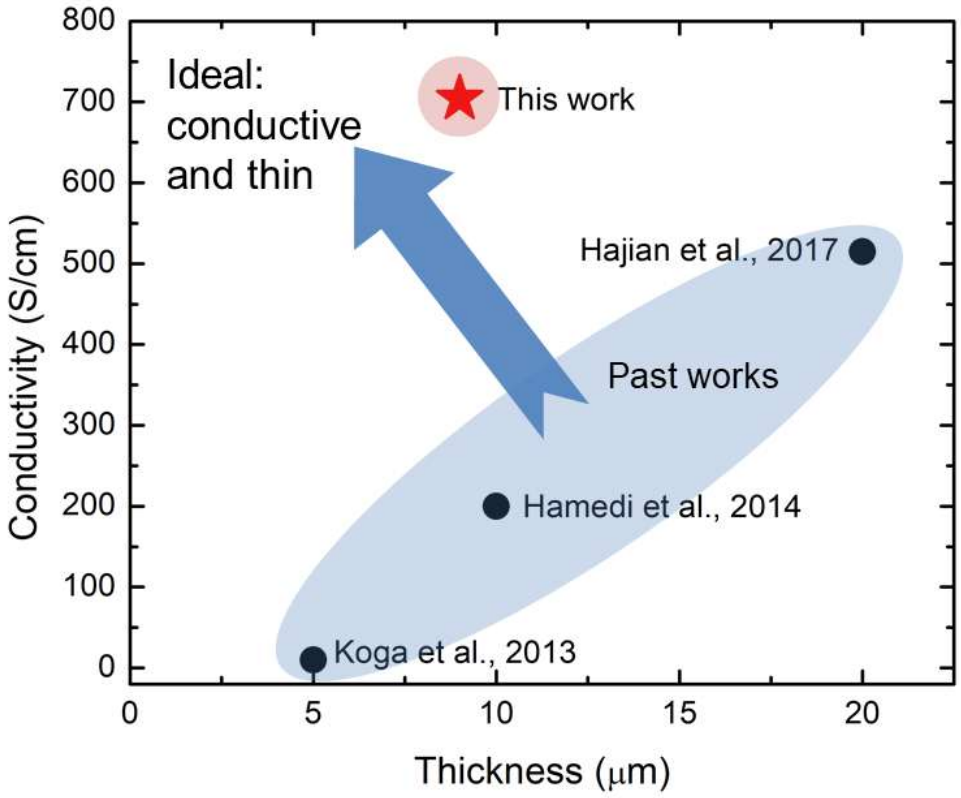

Figure S2. Literature DC conductivities of CNT-CNF films with varying thickness.

The current associated with the Q-HQ region as well as the area under the CV curves increased with time for the 1.7 V hold, with a very slight change after 12 h of potential holding. During the 48 h potential hold the total peak height increased by a factor of 2.9, compared to an increase of 4.8 times for commercially available active carbon cloth (ACC) current collector.

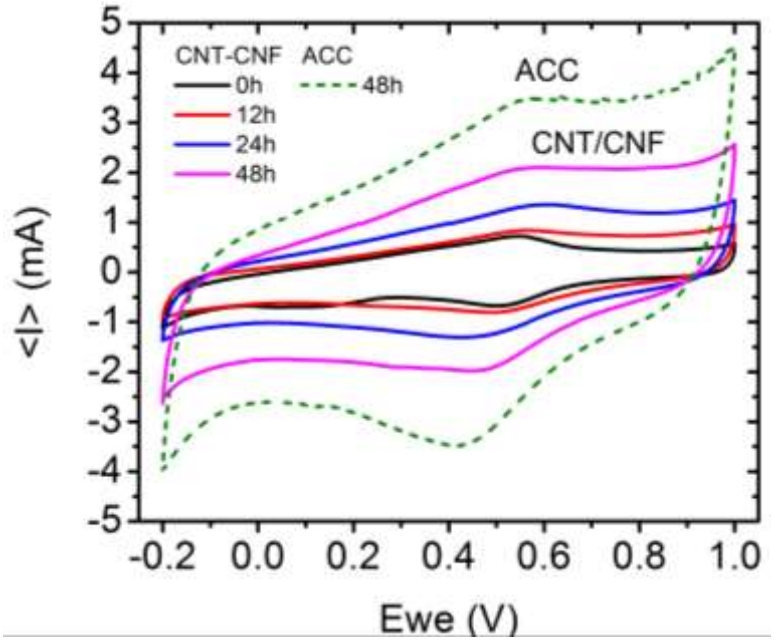

Figure S3. CV curves recorded after holding a CNT-CNF film at 1.7 V for 0 h, 12 h, 24 h, and 48 h in 5.0 M H_2SO_4, scan rate: 0.5 mV/s. Control: CV curves of ACC film at 1.7 V for 48 h in the same testing conditions.

Application of the CNT-CNF film (CNT/CNF weight ratio is 6/1) as a current collector in non-aqueous electrolytes such as in organic Li-ion based systems is also demonstrated. $LiFePO_4$ (LFP) particles were mixed with carbon black, PVDF binder in NMP solvent to make a slurry, which was coated onto CNT-CNF by doctor blade method. The mass ratio of LFP/carbon black/binder is 8/1/1. SEM and EDX images in Figure S4 confirmed that LFP electrode were coated well on the CNT-CNF current collector.

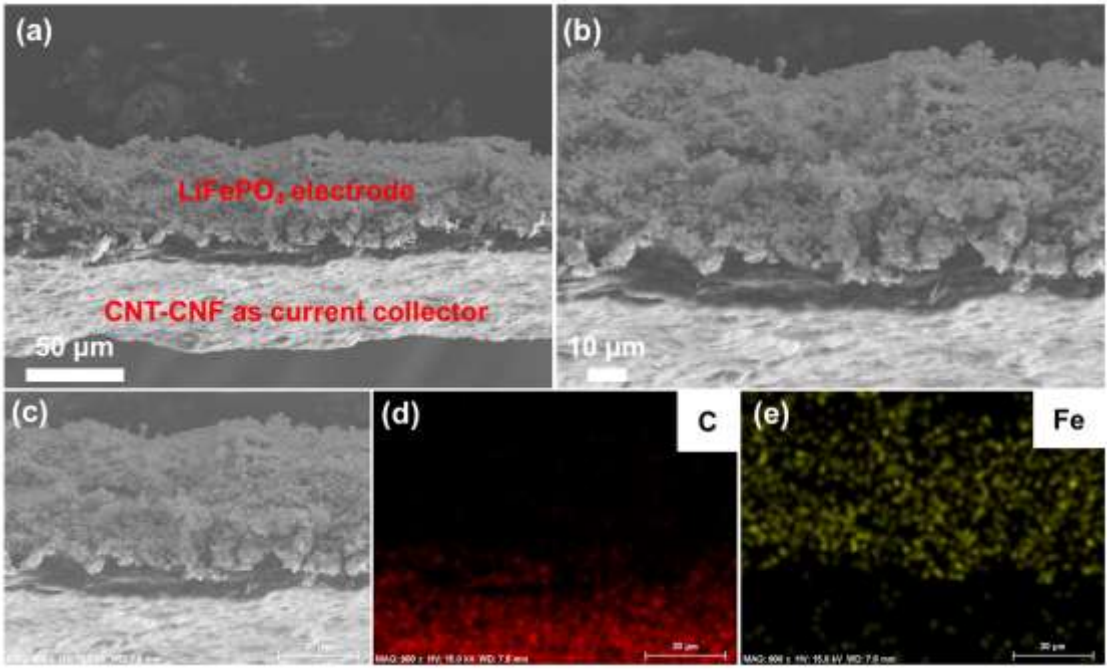

Figure S4. (a, b) SEM and (c-e) EDX images of LFP electrode coating on CNT-CNF film.

Figure S5a gives the charge/discharge curves of the LFP electrode using a current rate of 1C, which exhibited a typical redox couple by the flat plateaus with a small voltage gap. This indicates the good kinetics by using CNT-CNF as the current collector. Moreover, the cycling performance is stable over about 140 cycles.

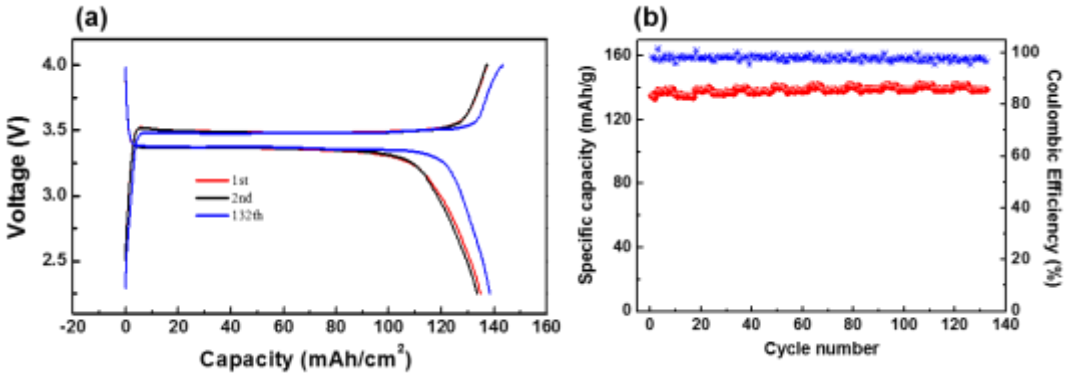

Figure S5. (a) Charge/discharge curves and (b) cycling performance of LFP//Li metal cell in 1.0 M $LiPF_6$-EC-DEC electrolyte, where LFP cathode was coated on CNT-CNF current collector.

In addition to testing in acidic electrolyte as reported in the main text, the stability of CNT-CNF composite in alkaline electrolyte (5M KOH) was tested. However, the CNT-CNF is chemically unstable against the alkaline electrolyte. The CV below shows the instability under this condition. After 12 h potential holding the films became very mechanically weak and were unusable (Figure S6).

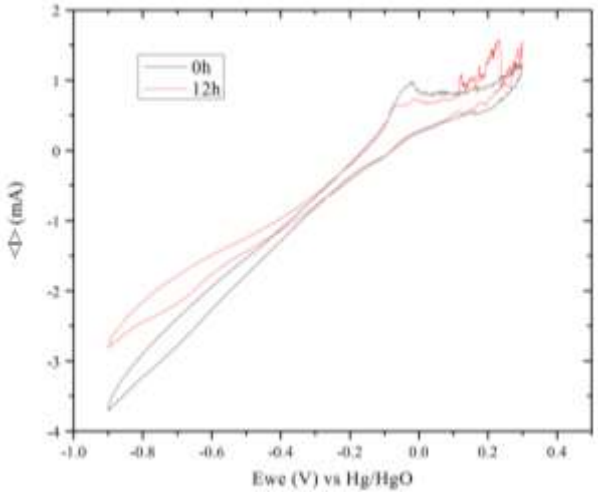

Figure S6. CV curve of CNT-CNF film in 5M KOH electrolyte system.

Recently, Suo et al. reported a new high concentrated aqueous electrolyte (21.0 M LiTFSI in water), which exhibited a wide potential window (Ref. 9). Here we also tested the stability of CNT-CNF film in such a highly concentrated electrolyte. The stability window is 2.7 V, as shown in the figure below (Figure S7).

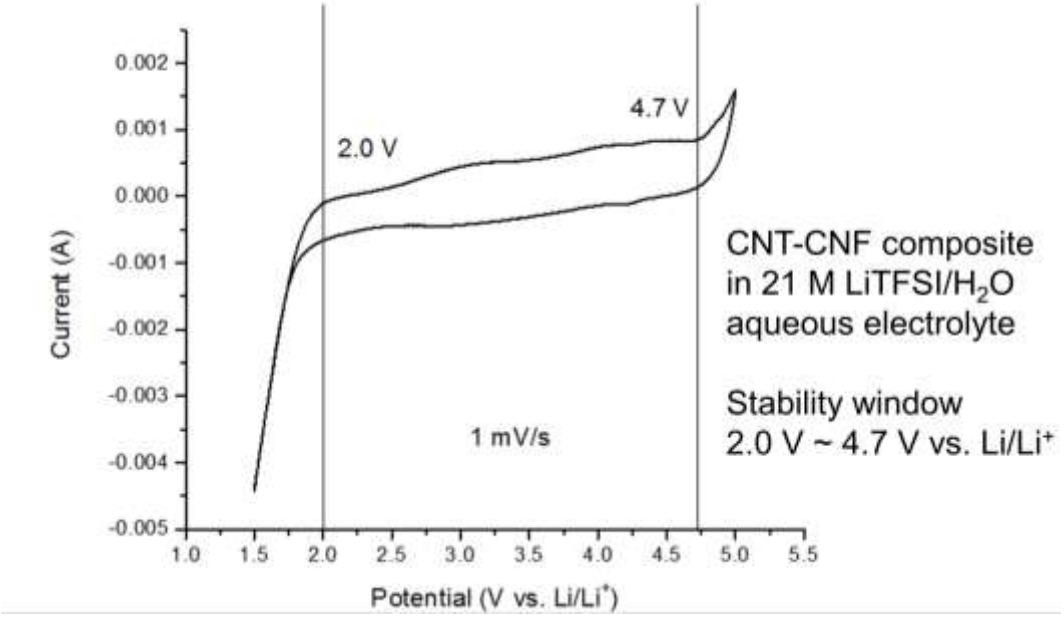

Figure S7. CV curve of CNT-CNF film in 21 M LiTFSI in water electrolyte system.

Figure S8 shows initial attempts at fabrication of lead and lead dioxide electrodes on CNT-CNF and ACC current collectors. After galvanostatic cycling with potential limitation (GCPL) for 10 cycles, the CNT-CNF current collector remained mechanically intact, showing cycling stability in our initial efforts to fabricate a full cell lead-acid battery.

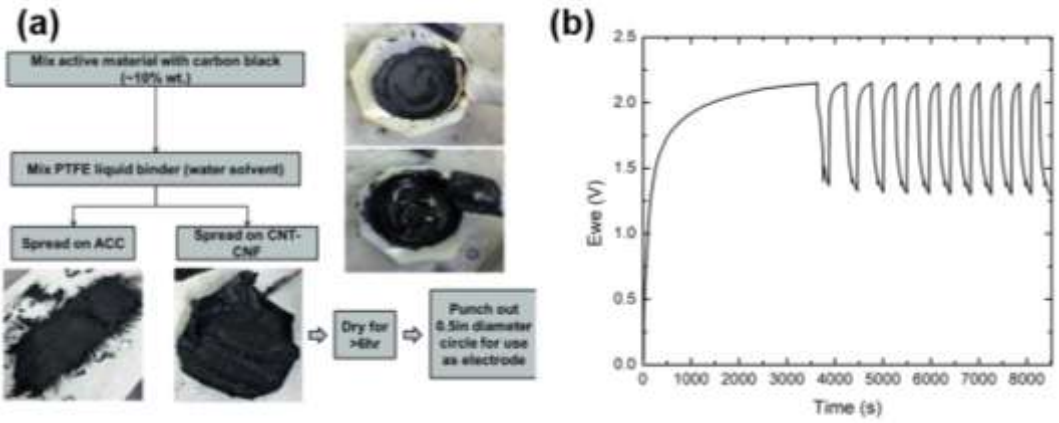

Figure S8. (a) Flow chart of lead/lead dioxide electrode fabrication on CNT-CNF and ACC current collectors. (b) Galvanostatic cycling of lead-acid full cell with CNT-CNF current collectors with potential limitations from 1.3 V to 2.15 V.

Cost Analysis

A cost analysis for the fabrication of CNT-CNF current collectors, specifically the associated materials cost, is performed. Materials required for synthesizing carbon nanotubes (CNT) include: (1) catalysts for CNT growth; (2) acids for CNT purification; (3) carbon resources. Currently, we can get a large number of CNT from market and the price can be less than 100 $/kg from Alibaba. Another component, cellulose nanofiber (CNF) or TEMPO-treated cellulose nanofiber (CNF) can also be purchased from the market at a price of about 4-6 $/kg from Alibaba.

The material cost of the CNT-CNF current collector is estimated to be around 1.0 $/m^2. The processing cost is then estimated. The price of roll-to-roll coating is estimated to be 500 $/hour. The coater width can be 72-inches wide, so the resulting coating layer cost is ~0.027 $/m^2, which is small compared to the raw materials cost.

The gross total cost of CNT-CNF is about 1.027 $/m^2. This is quite low compared to the cost of common heavy metal current collectors such as nickel foam, titanium foil, and stainless steel, which can be upwards of 10 $/m^2.