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Thin, Flexible Secondary Li-Ion Paper Batteries

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ABSTRACT There is a strong interest in thin, flexible energy storage devices to meet modern society needs for applications such as interactive packaging, radio frequency sensing, and consumer products. In this article, we report a new structure of thin, flexible Li-ion batteries using paper as separators and free-standing carbon nanotube thin films as both current collectors. The current collectors and Li-ion battery materials are integrated onto a single sheet of paper through a lamination process. The paper functions as both a mechanical substrate and separator membrane with lower impedance than commercial separators. The CNT film functions as a current collector for both the anode and the cathode with a low sheet resistance (~5 Ohm/sq), lightweight (~0.2 mg/cm^2), and excellent flexibility. After packaging, the rechargeable Li-ion paper battery, despite being thin (~300 μm), exhibits robust mechanical flexibility (capable of bending down to <6 mm) and a high energy density (108 mWh/g).

KEYWORDS: paper batteries · lamination · free-standing thin film · flexible

Integration of electronics onto existing, widely used paper could bring unprecedented opportunities for consumer electronics.[1−3] These devices can be paper-thin, flexible, lightweight and manufactured by a low cost, roll-to-roll printing process. Power sources are needed for the operation of the paper electronics, and ideally, a power source directly integrated onto paper would be preferred for easy system integrations. On the other hand, secondary Li-ion batteries are key components in portable electronics due to their high power and energy density and long cycle life.[4] In these devices, metal strips, mainly copper (~10 mg/cm^2) and aluminum (5 mg/cm^2), are used as current collectors. Recently, solution-processed carbon nanotube (CNT) thin films have been widely studied and applied as electrodes for optoelectronics due to their high conductivity and flexibility.[3,5] CNT thin films on plastic substrates have been explored as current collectors for supercapacitors.[6] We recently demonstrated that paper coated with CNTs or silver nanowires can be used to replace heavy metals in supercapacitors and Li-ion batteries.[7] The CNT films on substrate function effectively as current collectors and enable some new properties for devices.

In this paper, we integrated all of the components of a Li-ion battery into a single sheet of paper with a simple lamination process. Free-standing, lightweight CNT thin films (~0.2 mg/cm^2) were used as current collectors for both the anode and cathode and were integrated with battery electrode materials through a simple coating and peeling process. The double layer films were laminated onto commercial paper, and the paper functions as both the mechanical support and Li-ion battery membrane. Due to the intrinsic porous structure of the paper, it functions effectively as both a separator with lower impedance than commercial separators and has good cyclability (no degradation of Li-ion battery after 300 cycles of recharging). After polymer sealing, the secondary Li-ion battery is thin (<300 μm), mechanically flexible, and has a high energy density. Such flexible secondary batteries will meet many application needs in applications such as interactive packaging, radio frequency sensing, and electronic paper.

CNT thin films were coated onto stainless steel (SS) substrates with a solution-based process. Aqueous CNT ink was prepared with 10% by weight sodium dodecylbenzenesulfonate (SDBS) as the surfactant.[8] The concentration of CNT is 1.7 mg/mL. The CNT ink was applied to the SS substrate with a doctor blade method.[9] A dried film with a thickness of ~2.0 μm was formed after drying the CNT ink on the SS substrate at 80 °C for 5 min. Slurries of battery materials, $Li_4Ti_5O_{12}$ (LTO) (Süd Chemie) and $LiCoO_2$ (LCO) (Predmaterials & LICO), were prepared by mixing 70 wt % active

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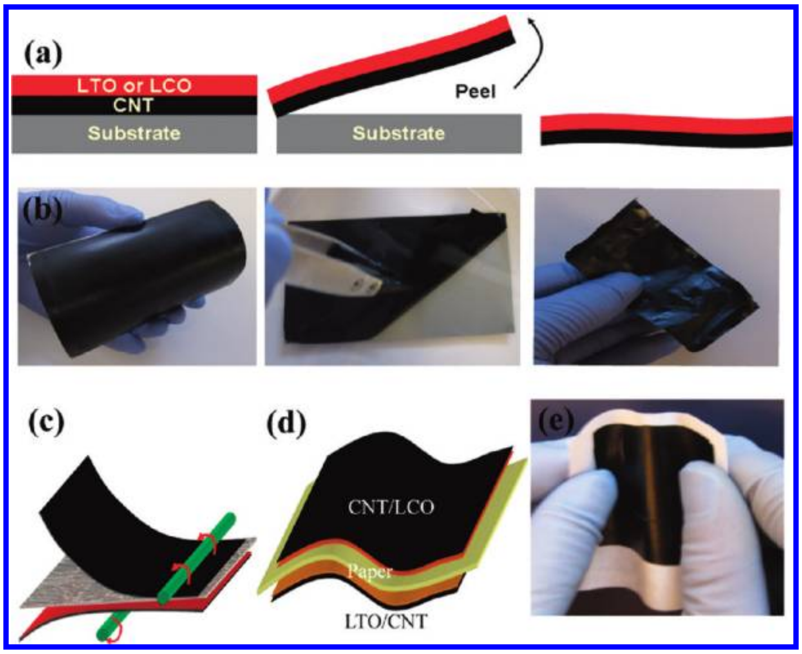

Figure 1. (a) Schematic of fabrication process for free-standing LCO/CNT or LTO/CNT double layer thin films. The CNT film is doctor-bladed onto the SS substrate and dried. An LTO or LTO slurry is then doctor-blade-coated on top of CNT film and dried. The whole substrate is immersed into DI water, and the double layer of LTO/CNT or LCO/CNT can be easily peeled off due to the poor adhesion of CNTs to the SS substrate. (b) (Left) 5 in. × 5 in. LTO/CNT double layer film coated on SS substrate; (middle) the double layer film can be easily separated from the SS substrate in DI water; (right) the final free-standing film after drying. (c) Schematic of the lamination process: the free-standing film is laminated on paper with a rod and a thin layer of wet PVDF on paper. (d) Schematic of the final paper Li-ion battery device structure, with both LCO/CNT and LTO/CNT laminated on both sides of the paper substrate. The paper is used as both the separator and the substrate. (e) Picture of the Li-ion paper battery before encapsulation for measurement.

materials, 20 wt % Super P carbon, and 10 wt % polyvinylidene fluoride (PVDF) binder (Kynar 2801) in *N*-methyl-2-pyrrolidone (NMP). The battery slurries were applied to CNT/SS with the same doctor blade method. The slurries were dried at 100 °C for 0.5 h. The battery electrode material on the CNT film forms a double layer film, where CNT films function as the current collectors. As shown in Figure 1a, the double layer LCO/CNT or LTO/CNT film was lifted off by immersing the SS in DI water followed by peeling with tweezers. Figure 1b shows a LTO/CNT film with a size of 7.5 cm × 12.5 cm on a SS substrate (left) being peeled off in water (middle) and in a free-standing form (right). Previously, CNT thin films have been coated mainly on plastic substrate for use as transparent electrodes in various device applications, including solar cells and light-emitting diodes.[3,5,10,11] In this study, we found that CNTs have weaker interaction with metal substrates when compared with plastic or paper substrates, which allows us to fabricate free-standing films with integrated current collector and battery electrodes. The double layer films obtained with this method are lightweight, with ~0.2 mg/cm^2 CNT and ~2 −10 mg/cm^2 electrode material. The free-standing double layer film shows a low sheet resistance (~5 Ohm/sq) and excellent flexibility, without any change in morphology or conductivity after bending down to 6 mm (Mandrel). Due to the excellent mechanical integrity of the double layer film and the loose interaction between the CNT film and SS, peeling off the double layer film from the SS is highly reproducible.

After integrating the battery electrode materials on the lightweight CNT current collectors, a lamination process was used to fabricate the Li-ion paper batteries on paper. A solution of polyvinylidene fluoride (PVDF) polymer was Mayer-rod-coated on the paper substrate with an effective thickness of 10 μm. The wet PVDF functions as a glue to stick the double layer films on paper. The concentration of PVDF in *N*-methyl-2-pyrrolidone (NMP) was 10% by weight. As shown in Figure 1c, the double layer films were laminated on the paper while the PVDF/NMP was still wet. During this process, a metal rod rolls over the films to remove air bubbles trapped between films and the paper separator. After laminating LTO/CNT on one side of the paper, the same process was used to put LCO/CNT on the opposite side of the paper to complete the Li-ion battery fabrication. Figure 1d,e shows the scheme and a final device of the Li-ion paper battery prior to encapsulation and cell testing. Although a paper-like membrane has been used as the separator for other energy storage systems including supercapacitors, it is the first demonstration of the use of commercial paper in Li-ion batteries,[12] where paper is used as both separator and mechanical support.

The cross section of the laminated Li-ion paper battery, with the CNT/LTO/paper/LCO/CNT structure, was examined with SEM. Figure 2a reveals the surface morphology of Xerox paper, with large fibers (~20 μm diameter) and surface roughness (peak to valley is ~10 μm). Xerox paper lacks microsize holes, which makes it an excellent separator for Li-ion batteries with the laminated electrode films. We tried coating battery electrode materials with the same slurries directly onto either side of Xerox paper, and we found occasional shorting of the device due to the leakage of battery electrode materials through paper. The lamination process provides an efficient approach for solving the leakage problem by using Xerox paper as a separator because the battery electrode forms a solid film and is integrated with the CNT film. An SEM image at low magnification reveals that LTO/CNT and LCO/CNT form a continuous, solid film (see Supporting Information). Figure 2b shows the cross section of the LTO/CNT double layer on top of Xerox paper separator. The SEM reveals the continuous morphology of CNT thin films

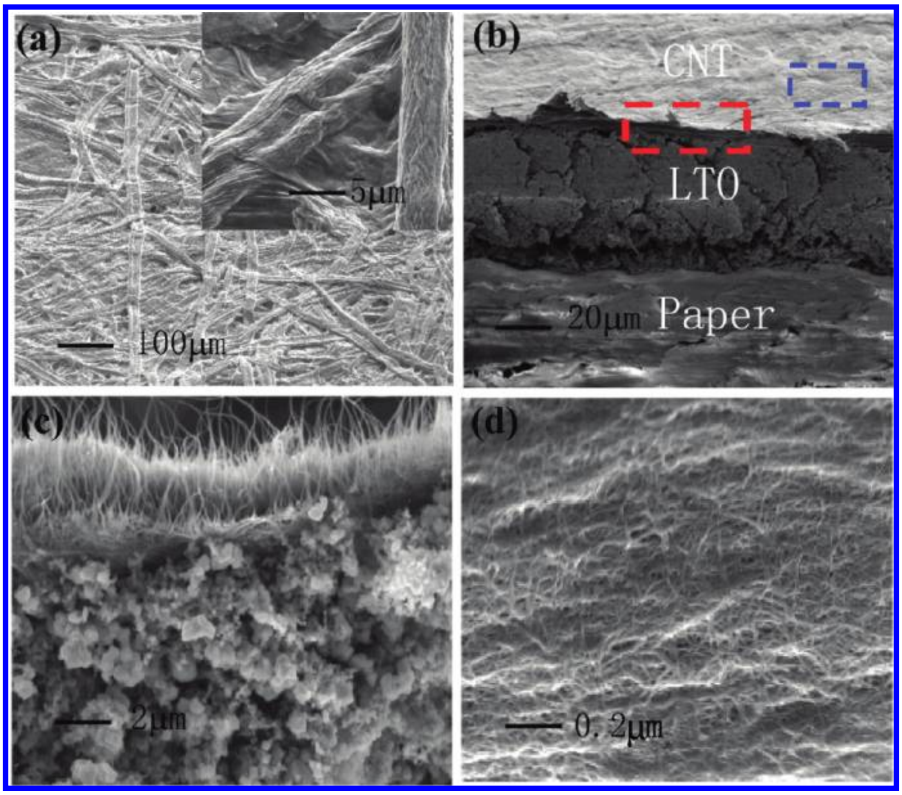

Figure 2. (a) SEM of porous, rough Xerox paper as Li-ion battery separator. Inset shows the zoomed-in image of the surface morphology of the paper substrate. (b) SEM image of the cross section for the laminated paper Li-ion battery. The layers thicknesses are CNT ~2 μm, LTO ~30 μm, paper ~100 μm, LCO ~30 μm, and CNT ~2 μm. (c) Zoomed-in image of the cross section of the CNT/LTO double layer, as in the red box in (b). (d) SEM image of the CNT film as a current collector, as in the blue box in (b).

with thicknesses of ~2 μm. The composite LTO electrode film is densely packed with a thickness of ~30 μm. The thickness of Xerox paper used in this study is ~100 μm. The porous morphology of paper allows the electrolyte to diffuse efficiently into it, which allows the paper to be used effectively as a separator. Figure 2c reveals the zoomed-in image of the interface between LTO and CNT as in Figure 2b, which shows no CNT penetration into the LTO layer. CNT thin films form continuous mechanical supports and serve as electrical current collectors for the electrodes. The sheet resistance of the CNT thin film is measured with a four-point probe and is ~5 Ohm/sq, and it can be further decreased with acid doping such as with HNO_3 or $SOCl_2$.[13] A similar double layer resulting from the integration of the cathode material, LCO, on top of CNT film was observed, as well (see Supporting Information). Figure 2d shows the surface of a highly conductive CNT film as a current collector.

To evaluate the performance of paper as an effective separator membrane for Li-ion batteries, its stability in the electrolyte and the effect of the impurities, mainly OH groups, in a large voltage range with respect to Li metal were tested. Pouch cells were fabricated with CNT films as cathodes, Li-metal as anodes, and Xerox paper as the separators (see Supporting Information). The cells were cycled with 50 μA/cm^2 current densities between 1 and 4.3 V (Figure 3a). The charge and discharge capacities are minimal, ~0.01 mAh/cm^2, which shows that the irreversible capacities from both the paper separator and the CNT film are negligible (<0.001 mAh/cm^2). It has been reported that CNT thin films have been used as anodes for Li-ion batteries due to their large surface areas, but they show large irreversible capacities and low coulombic efficiencies for the first cycle when cycled below 1 V *vs* Li/Li^+.[14] Due to the small mass loading of the CNTs, ~0.2 mg/cm^2, and the operating voltages of LTO and LCO (above 1 V), the irreversible capacities from CNTs are negligible. Furthermore, paper shows low resistivity in the electrolyte. Impedance spectroscopy was used to obtain information on the resistivity of the solution in the paper. Coin cells with LTO *versus* Li metal were made, and the Nyquist plot at open circuit conditions is reported (Figure 3b). The high frequency intercept of the impedance spectrum with the *x*-axis represents the resistance of the solution in the pores of the separator, R_{SL}, as evidenced in the plot. In the inset of Figure 3b, the value of R_{SL} for different thicknesses of the separator is reported. The value of R_{SL} is given by the following expression:

$$R_{SL} = \frac{\rho_S L}{A}\left(\frac{\tau}{f}\right) \qquad (1)$$

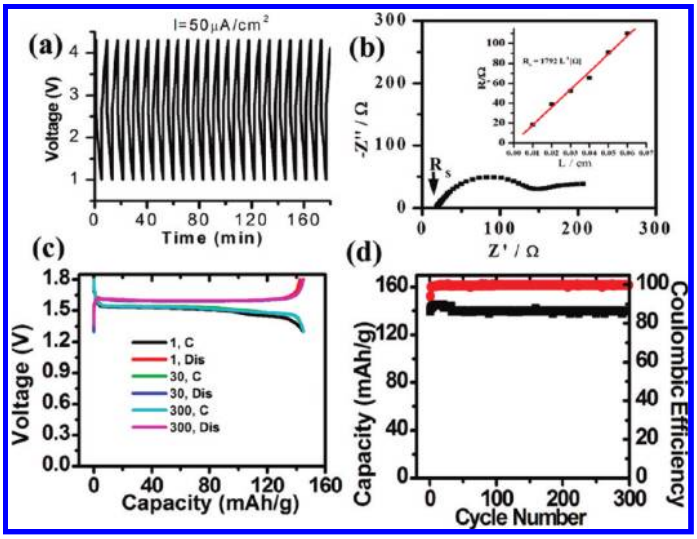

Figure 3. (a) Galvanostatic charging/discharging curves of CNT/paper *vs* a Li metal anode to show the negligible capacitance between 1 and 4.3 V. (b) Impedance of Xerox paper as a separator in a Li-ion battery test where LTO is the anode and Li metal is the cathode. The inset shows the impedance with different layers of paper. (c) Galvanostatic charging/discharging curves of the LTO anode (1.3−1.7 V) half cells with conductive paper current collectors. The mass of the LTO electrode is 1.8 mg. The current rate is C/5. (d) Cycling performance of LTO nanopowder (C/5, 0.063 mA) half cells.

where ρ_S is the resistivity of the electrolyte (~100 $\Omega \cdot$ cm for the standard EC/DEC solution),[15] L is the thickness of the separator, A is transversal area (the area perpendicular to the axis of the electrode), τ is tortuosity (the ratio between the path length of the ions and the thickness of the electrode), and f is pore fraction (the ratio between the pore volume and the total geometrical volume of the electrode). The ratio τ/f is important for the separator, which indicates how easy it is for the electrolyte to penetrate through. The value of the ratio between the tortuosity and the pore fraction for the paper is $\tau/f \approx 9.1$, while it is $\tau/f \approx 28.8$ for the standard separator. This fact is significant because it demonstrates that the paper will show a better conductivity than the standard separator at the same thickness. The cheap, commercial paper functions as an effective replacement for a standard separator membrane and can serve as well as a mechanical support with similar impedance and a smaller ratio between the tortuosity and the pore fraction.

To test the feasibility of using Xerox paper as the separator in Li-ion batteries with the lamination process, half cells were made with CNT/LTO or CNT/LCO with lithium foil as a counter electrode (see Supporting Information). Voltage profiles closely match those with metal current collectors according to previous work, and no apparent voltage drop was observed.[16−18] Figure 3c shows the voltage profile for a half cell of CNT/LTO, and no apparent voltage drop was observed when the voltage profiles for first, 30th, and 300th cycles were compared. The cycling performance of these conductive paper-supported electrodes is shown in Figure 3d. The CNT/LTO electrodes achieved initial discharge capacities of 147 mAh/g and exhibited a capacity retention of 95% after 300 cycles at C/5. These values are close to those obtained for metal collector-based Li-ion batteries.[16−18] The coulombic efficiencies for the CNT/LTO half cells are generally over 99.0%. We also observed an increase in the coulombic efficiencies and discharge capacities over the first few cycles. Our recent work also shows that paper is stable in the electrolyte solution for eight months in Li-ion battery test, where the same electrolyte was used as in this study.

Full cells with integrated current collectors and battery electrodes onto a single sheet or paper are fabricated with the same lamination process. Previously, Friend *et al.* reported two-layer polymer diodes fabricated by lamination followed by annealing.[19] Yang *et al.* has demonstrated stacked plastic solar cells with an electronic glue-based lamination process with interface modification.[20] The laminated Li-ion paper battery has the structure illustrated in Figure 1d (see Supporting Information, as well). After the CNT/LCO and CNT/LTO films were laminated onto the two sides of Xerox paper, the whole device was sealed with 10 μm PDMS (see Supporting Information) in an Ar-filled glovebox using $LiPF_6$ in EC/DEC electrolyte. The Li-ion paper battery is thin, ~300 μm in total. The anode and cathode mass loadings are 7.2 and 7.4 mg/cm^2, respectively. The assembled paper battery was taken outside of the glovebox for battery testing. As shown in Figure 4a, the paper battery is able to light up a red LED continuously for 10 min without fading. Due to the small thickness and the great flexibilities of current collectors using CNT thin films, the whole device shows excellent flexibility (Figure 4b). No failure was observed for the paper battery after manually bending the device down to 6 mm for 50 times (see Supporting Information also). Figure 4c shows the first cycle voltage profile of the Li-ion paper battery sealed with a transparent bag, where the thickness of the plastic is ~10 μm. The cycling performance of the stacked cells is shown in the inset of Figure 4d. The first coulombic efficiency is 85%, slightly lower than that of a typical Li-ion battery with LCO and LTO electrodes. After the first cycle, the coulombic efficiency is 94−97%. The discharge retention is 93% after 20 cycles. For practical applications, especially for large-scale energy storage, good self-discharge performance is crucial. The voltage was monitored after the battery was charged to 2.7 V for 5 min at a C/10 rate and disconnected. As shown in the inset in Figure 4d, the voltage drops about 2% instantly, which is due to the IR drop after switching off the current. After that, a 5.4 mV voltage drop was observed for the full cell after 350 h. This is equivalent to <0.04% self-discharge if the Li-ion paper battery is fully charged after a month. The self-discharge performance could be further improved through device fabrication process modifications such as better sealing, longer vacuum baking times, and lower moisture levels by using standard dry rooms.

There is a great need for development of lightweight, thin, and flexible batteries for portable electronic applications with low power consumption, <1.0 mW. Ajayan *et al.* developed flexible batteries and supercapacitors based on nanocomposite paper in 2007; Mihranyan *et al.* developed ultrafast all-polymer paper-based batteries in 2009; and we explored conductive paper for energy storage recently.[7,21,22] Enfucell Inc. and Blue Spark Inc. have recently developed a flexible and soft battery by using a printing method on plastic substrates. The Li-ion paper battery developed in this article has advantages in various aspects. In Ajayan's nanocomposite-based battery, Li metal was used as one electrode and is neither thin nor flexible. The polymer battery developed by Mihranyan *et al.* performs as a mixed battery and capacitor, which shows a nonflat discharge curve and has a large thickness (~2 mm). The soft batteries from Enfucell and Blue Spark are made on plastic substrates, not paper, and are not rechargeable. Figure 4e and Table 1 in Supporting Information show the comparison of our flexible, thin paper battery with theirs. The blue arrow indicates the improvement direction for flexible storage devices. Our paper battery is rechargeable and has a higher energy density, 108 mWh/g, based on the total mass of the device, and it is much thinner (~300 μm). Currently, we are using carbon CNTs with a price of ~\$200/g. The CNT weight in our device is less than 0.2 mg/cm^2, which is ~\$0.02/cm^2. Therefore, the CNT cost is negligible. Due to the porous structure of CNT thin film as current collector, the sealing of the paper thin film battery will be more challenging. One method for increasing the total energy for the Li-ion paper battery is through stacking layer upon layer, as in Figure 4f, where conductive CNT films function as current collectors, and extended metal strips at the edge serve as connections to the external circuit. To demonstrate the feasibility of the stacking of the paper battery, we have fabricated a cell with 9 layers stacked in parallel (see Supporting Information). The individual cells are separated by 10 μm plastic. The stacked cells in parallel are sealed within a transparent plastic bag. The cells were enclosed and sealed inside the transparent plastic bag in an Ar-filled glovebox with an Al strip on the cathode side and a Cu strip on the anode side extending out for outside electrical contact (Figure 4f, right). In this way, the multiple cells are connected in parallel. The stacked cells were tested and showed similar performance to individual cells, where the total current is equal to the sum of the individual cells.

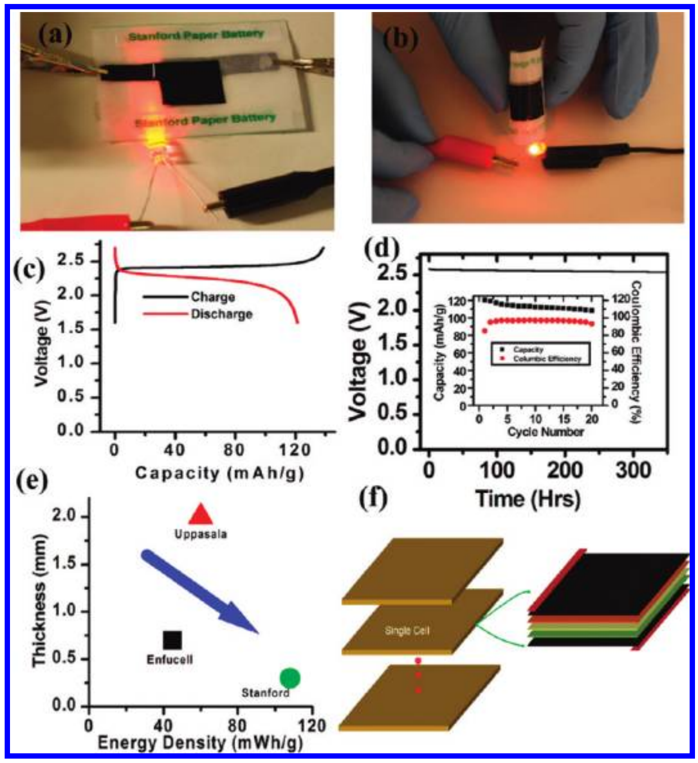

Figure 4. (a) Lighting a red LED with a Li-ion paper battery which is encapsulated with ~10 μm PDMS. (b) Flexible Li-ion paper batteries light an LED device. (c) Galvanostatic charging/discharging curves of a laminated LTO–LCO paper batteries, a structure as in Figure 1d. (d) Self-discharge behavior of a full cell after being charged to 2.6 V. The initial drop is due to the IR drop after turning off the charging current. Inset: cycling performance of LTO–LCO full cells. (e) Comparison of our paper Li-ion battery with a polymer paper battery. The green arrow indicates the target of the paper battery. (f) Schematic for stacked cells separated by 10 μm plastic paper. An individual cell is made with laminated LTO/CNT and LCO/CNT on either side of a piece of Xerox paper. A small piece of Cu is connected on the LTO/CNT side and Al on the LCO/CNT side.

Figure S8 (Supporting Information) shows the operation of the stacked Li-ion paper battery. Since the device scale is small and the sheet resistance of CNT film is ~5 Ohm/sq, the sheet resistance effect on voltage drop is small. This concept can be applied to a multiwalled CNT with enough film thickness for high surface conductance.

In conclusion, we have demonstrated a Li-ion battery integrated onto a single sheet of paper through a simple lamination process. The paper substrate functions as both the substrate and the separator, and highly conductive CNT films function as current collectors for both the anode and the cathode. Such rechargeable energy storage devices are thin, flexible, and lightweight, which are excellent properties for various applications where embedded power devices are needed, such as RFID tags, functional packaging, and new disposable applications.

METHODS

Free Standing LTO/CNT Double Layer Films: Aqueous CNT ink was prepared with 10% by weight sodium dodecylbenzenesulfonate (SDBS) as surfactant based on commercial, arc-discharged CNTs (Carbon Solutions, Inc.). The CNT ink was then blade-coated onto a SS substrate. The SS substrate was used as received. Slurries of battery materials, $Li_4Ti_5O_{12}$ (LTO) (Süd Chemie) and $LiCoO_2$ (LCO) (Pred Materials & LICO), were prepared by mixing 70 wt

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% active materials, 20 wt % Super P carbon, and 10 wt % polyvinylidene fluoride (PVDF) binder in *N*-methyl-2-pyrrolidone (NMP) as the solvent. The slurries were stirred overnight at room temperature. Afterward, the slurries with a thickness of ~125 μm were blade-coated on top of CNT films on SS substrates and dried at 100 °C for 1 h. The double layer LTO/CNT or LCO/CNT films were formed on SS substrates. To delaminate the double layer films, the SS was immersed into a beaker with DI water. After gentle shaking of the beaker, the double layer films easily delaminated from the SS substrate.

Fabrication and Test of Li-Ion Batteries: For half cell tests of LTO/CNT and LCO/CNT, coin cells were fabricated. Lithium metal foil (Alfa Aesar) was used as the counter electrode in each case. Xerox paper was used as the separator. Lithium metal and freestanding LTO/CNT or LCO/CNT films were punched into round shapes. The parts for coin cell assembly were purchased from MTI Corporation (Richmond, CA). A 1 M solution of $LiPF_6$ in EC/DEC (1:1 vol/vol; Ferro) was used as the electrolyte. The charge/discharge cycles were performed at different rates at room temperature. The devices were assembled in an argon-filled glovebox with oxygen and water contents below 1 and 0.1 ppm, respectively. The Li-ion battery tests were performed by either a Bio-Logic VMP3 battery tester or an MTI battery analyzer.

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Supporting Information Available: Additional figures and experimental details. This material is available free of charge *via* the Internet at http://pubs.acs.org.

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