NANO LETTERS

pubs.acs.org/NanoLett

Stretchable, Porous, and Conductive Energy Textiles

Liangbing Hu,[†] Mauro Pasta,[†] Fabio La Mantia,[†] LiFeng Cui,[†] Sangmoo Jeong,[‡] Heather Dawn Deshazer,[†] Jang Wook Choi,[†] Seung Min Han,[†] and Yi Cui[†,*]

[†] Department of Materials Science and Engineering and [‡] Department of Electrical Engineering, Stanford University, Stanford, California 94305

ABSTRACT Recently there is strong interest in lightweight, flexible, and wearable electronics to meet the technological demands of modern society. Integrated energy storage devices of this type are a key area that is still significantly underdeveloped. Here, we describe wearable power devices using everyday textiles as the platform. With an extremely simple "dipping and drying" process using single-walled carbon nanotube (SWNT) ink, we produced highly conductive textiles with conductivity of 125 S cm^{-1} and sheet resistance less than 1 Ω/sq. Such conductive textiles show outstanding flexibility and stretchability and demonstrate strong adhesion between the SWNTs and the textiles of interest. Supercapacitors made from these conductive textiles show high areal capacitance, up to $0.48 F/cm^2$, and high specific energy. We demonstrate the loading of pseudocapacitor materials into these conductive textiles that leads to a 24-fold increase of the areal capacitance of the device. These highly conductive textiles can provide new design opportunities for wearable electronics and energy storage applications.

KEYWORDS Energy storage, textile, carbon nanotube, supercapacitor

Wearable electronics represent a developing new class of materials with an array of novel functionalities, such as flexibility, stretchability, and lightweight, which allow for many applications and designs previously impossible with traditional electronics technology. High-performance sportswear, wearable displays, new classes of portable power, and embedded health monitoring devices are some examples of these novel applications.[1−3] All these electronic applications require lightweight, wearable power conversion and storage devices. Textile is a flexible and porous material made by weaving or pressing natural or synthetic fibers, such as cotton or polyester. The ideal wearable power would incorporate textile as a component. Previously, studies have been done for integrating nanoscale materials into textiles to improve the clothing colors, antiodor function, UV protection, and human biomonitoring.[4−6] In this work, we conformally coat single-walled carbon nanotubes (SWNTs) on cellulose and polyester fibers to make porous conductors. The fabrication process is simple and scalable, similar to those widely used for dyeing fibers and fabrics in the textile industry. The SWNT coating makes these textiles highly conductive with sheet resistance less than 1Ω/sq. The conductive textiles (e-Textile) show outstanding mechanical and chemical properties. Their porous structure allows high mass loading of materials that increases the performance of energy storage devices. When conductive textiles are used as electrodes and standard textiles are used as separators, fully stretchable SCs are demonstrated, which will significantly benefit the development of wearable and stretchable electronics.[7]

"Dyeing" Textile with SWNT Ink. Textile fibers, such as cellulose or polyester, have a hierarchical structure with complicated surface morphology, functional groups such as hydroxyl groups, and high porosity.[8] For example, each cotton fiber is comprised of multiple individual cotton fibrils, which are in turn composed of multiple microfibrils bundled together. The microfibrils are made of poly-D glucose chains, usually arranged in crystalline, or partially crystalline, domains (Figure 1b). This structure allows the fibers to absorb large amounts of water, or other polar solvents, which causes the fibers to swell when placed in such solutions. The interaction of these polymeric fibers with SWNTs is facilitated by these properties, as well as several properties of the SWNTs themselves. SWNTs are mechanically flexible, which allows them to conform to the shape of the polymer fibers. Also, the interactions of SWNTs with polymer fibers have been widely studied. SWNTs have been proven to have large van der Waals interactions with many types of polymers, and SWNTs and cellulose fibers exhibit the large van de Waals forces that are demonstrated in other SWNT-polymer composites.[9−11] Furthermore, acid treated SWNTs have carboxyl functional groups on the surfaces and the ends, which can form strong hydrogen bonds with the hydroxyl groups in the cellulose fibers.[9−11] Because of the mechanical flexibility of SWNTs and the high surface area of cellulose fibers, together with the large water absorption of the fibers, surface contact between SWNTs and cellulose fibers is maximized. Upon contact, large van der Waals forces and hydrogen bonding occurs, which binds the SWNTs very tightly to the cellulose. Figure 1(b) shows a schematic of

---

* To whom correspondence should be addressed. E-mail: yicui@stanford.edu.

Received for review: 11/26/2009
Published on Web: 01/05/2010

DOI: 10.1021/nl903949m | *Nano Lett.* **2010,** *10,* 708-714

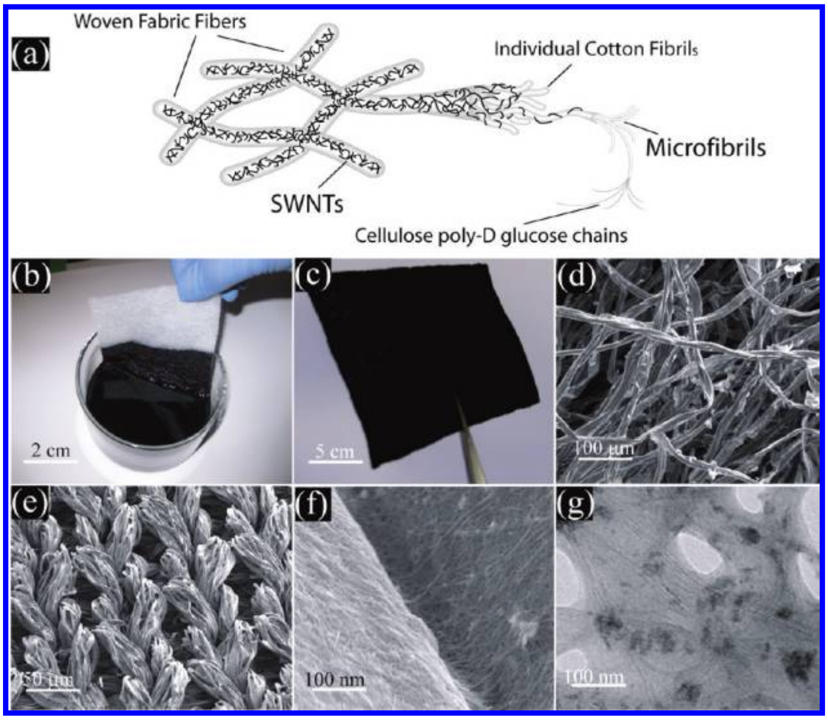

FIGURE 1. Porous textile conductor fabrication. (a) Schematic of SWNTs wrapping around cellulose fibers to form a 3D porous structure. (b) Conductive textiles are fabricated by dipping textile into an aqueous SWNT ink followed by drying in oven at 120 °C for 10 min. (c) A thin, 10 cm × 10 cm textile conductor based on a fabric sheet with 100% cotton and R_s of 4 Ω/sq. (d) SEM image of coated cotton reveals the macroporous structure of the cotton sheet coated with SWNTs on the cotton fiber surface. (e) SEM image of fabric sheet coated with SWNTs on the fabric fiber surface. (f) High-magnification SEM image shows the conformal coating of SWNT covering and bridging between the fabric fibers. (g) TEM image of SWNTs on cotton fibers.

SWNTs wrapping around cellulose fibers to create a 3D porous structure. To realize such structure, a ink with well-dispersed SWNTs is prepared by dispersing 1.6 mg/mL laser ablation tubes in water with 10 mg/mL sodium dodecylbenzenesulfonate (SDBS) as surfactant.[12] Then a textile is dipped into the black SWNT ink. Because of the strong absorption, the textile is quickly coated by the SWNT ink. The textile with SWNT ink is subsequently dried in oven at 120 °C for 10 min to remove water. A highly conductive textile is achieved through this simple dipping and drying process. Textiles investigated in this study include cotton sheet (Figure 1a) and woven polyester fabric (see Supporting Information). Figure 1c shows a conductive fabric with sheet resistance (R_s) of ~4 Ω/sq. These highly conductive textiles retain their texture and structure after SWNT coating and feel the same as the original material. This fabrication process can be easily applied to other ink made of nanostructured materials and scaled up with roll-to-roll techniques using slot-die or curtain coating processes.[13] SEM images of Figure 1d,e reveal the macroporous structures of a cotton sheet and a fabric, respectively. Conformal coating of SWNTs onto the fibers was observed for both the cotton and the fabric (Figure 1f). This conformal coating is a result of the mechanical flexibility of individual SWNTs and the strong binding energy between SWNTs and the cotton fibers[14] and accounts for the high conductivity of the textile. Previous studies have shown that SWNT films have microscale porosity, which is required to maximize the electrical double layer capacitance.[15] The microporous structure on top of the macroporous textile leads to what we call a double porous structure. Such double porous structures facilitate the easy access of electrolyte ions to the SWNTs, which is essential for high power SC applications. TEM images taken on SWNT-cotton fiber hybrids (Figure 1g) show the SWNTs are well bonded to the fiber and forming cross-linked networks, which provide conducting pathways.

Such porous textile conductors demonstrate excellent electrical, mechanical, and chemical resistance performance. Figure 2a shows that the conductance increases as the dipping number in SWNT ink increases. The R_s difference for cotton and fabric with different dipping number is due to the SWNT ink absorption difference for cotton and fabric per area (Supporting Information, Figure S1). Soaking the conductive cotton sheets in 4 M nitric acid for 30 min followed by blowing dry can wash away the surfactant molecules and induce hole doping,[16-18] which resulted in decrease of their R_s by approximately three times. The thickness of cotton sheets decreases from ~2 mm to ~80 μm with mechanical pressing, which changes the electrical conductivity from 5 to ~125 S/cm. Conductive textiles with a large range of conductance could be achieved by tuning the SWNT ink concentration and dipping number.

The conductive textiles show outstanding mechanical properties, that is, strong binding between SWNTs to the

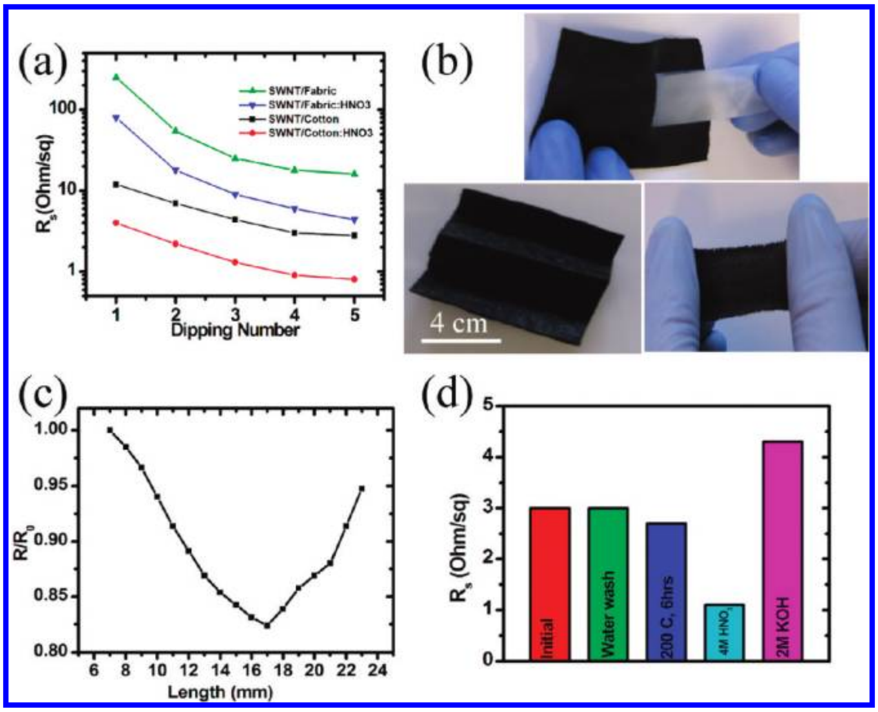

FIGURE 2. Properties of textile conductors. (a) Sheet resistance of fabric and cotton sheet after SWNT coating, which shows the same values on both faces for either fabric or cotton. The sheet resistances decrease by a factor of approximately 3 after HNO_3 treatment. (b) Excellent mechanical properties of conductive textile, that is, strong adhesion between SWNTs and textile (passing the scotch tape test), foldable, and stretchable. (c) The SWNT-coated textiles show unusual stretching properties. The film sheet resistance decreases as the SWNT/fabric is stretched up to 240% of its initial length, after which the resistance starts to increase. (d) SWNT/cotton is resistant to water washing, thermal treatment at 200 C for 6 h, 4 M HNO_3 acid, and 2 M KOH.

textile, foldability, and stretchability. The mechanical adhesion tests for SWNTs to cotton by the standard tape test and by washing in water show no visible nanotubes in the solution, or on the tape, and no conductivity degradation of the sample. In the water-washing test, the conductive cotton is soaked in water, then squeezed and wrung out. SWNTs stick well to the cotton without peeling or dissociating and precipitating in water. (Supporting Information, Figure S3b). These tests suggest that SWNTs adhere very strongly to the cotton fibers, which is critical for wearable electronic and power devices. Such strong binding may be due to the following reasons: (1) Large van der Waals forces and hydrogen bonding exist between SWNTs and the textile fibers.[14] (2) The flexibility of SWNTs allow them to be conformally adhered to the surface of cotton fibers which maximize the surface contact area between SWNTs and textile fibers.[19] The superior mechanical adhesion of SWNTs on cotton is essential for high-speed roll-to-roll fabrication and energy storage device stability. To test the stretchability, the resistance of a fabric sample with a dimension of 2.5 cm × 7 cm e-Fabric is monitored as it is being loaded in tension using a tensile tester. More detailed information can be found in the Supporting Information. In contrast to stretchable conductors reported by others[7,20,21] in which the conductivity decreases with stretching, as our SWNT-fabric is stretched, the conductance increases. It is remarkable that the conductance keeps increasing until the strain reaches a value of 140%, that is, the fabric is stretched to 2.4 times of its original length. This increase in conductance is due to the improvement of the mechanical contacts between fabric fibers, which leads to the better electrical contacts for SWNTs. As the strain increases further, the conductance starts to decrease, which is likely due to severe inhomogeneous deformation at large strains, as well as the reduced cross sectional area. Such stretchability of conductive textile could enable various stretchable electronic devices.[20] Fibers in cotton and fabric, as well as SWNTs, are known to be resistant to acid, base, and organic solvents.[22] Therefore, conductive textiles should also show the same chemical resistance. Figure 2d shows the R_s for e-Cotton after being exposed to water washing, thermal treatment at 200 °C for 6 h, 4 M HNO_3 acid for 30 min, and 2 M KOH for 30 min, respectively. Only 2 M KOH slightly increases the R_s of conductive cotton, while the others actually decrease the R_s possibly due to removing the surfactant or doping the nanotubes.[16−18] The chemical stability of the e-Textile allows a wide range of operations of energy storage devices when the porous textile conductor is used.

Porous Textile Conductor for Energy Storage. Porous textile conductor with SWNT coating was tested as both active charge storage electrodes and current collectors in SCs. Figure 3a shows the schematic drawing of the SC tested, where 1 M $LiPF_6$ electrolyte is used. The porous structure of conductive cotton facilitates the access of electrolyte to SC

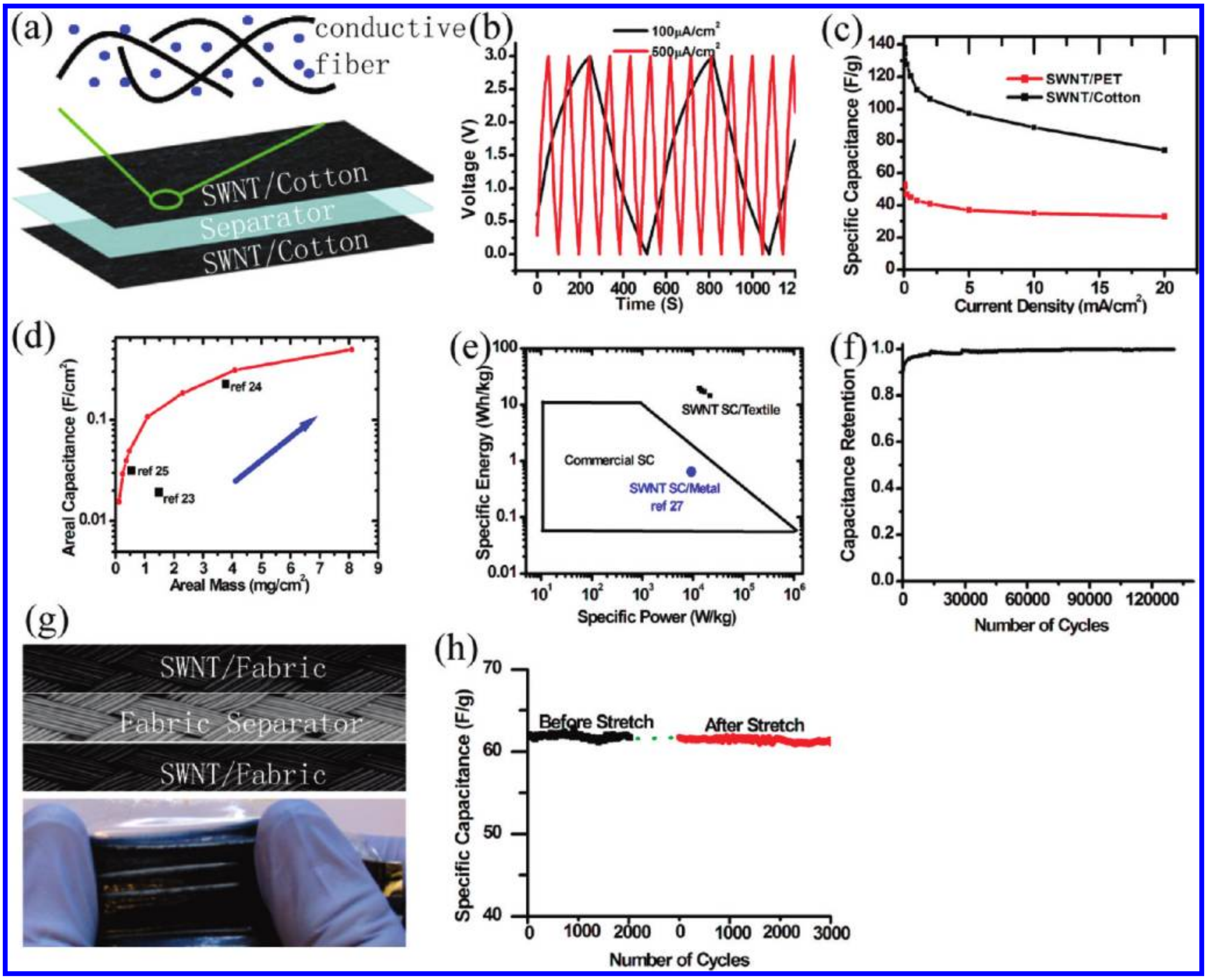

FIGURE 3. Organic SC with porous textile conductor. (a) SC structure with porous textile conductors as electrodes and current collectors. The porous structure facilitates the accessibility of electrolyte. (b) SC performance comparison between SWNTs on PET and SWNTs on cotton. (c) Areal capacitance increases with areal mass loading of SWNTs. Comparison with previous studies shows that our porous conductors allow the highest mass loading and highest areal capacitance. The current used is 200 μA/cm^2. (d) Ragone plot of commercial SCs, SWNT SC on metal substrates, and SWNT SC on porous conductors including all the weight. (e) Cycling stability of a SC with porous textile conductor. (f) The schematic drawing of the stretchable SCs with SWNT/fabric as electrodes and with stretchable fabric as the separator (top). A SC under 120% strain (bottom). (g) The specific capacity for a strechable SC before and after stretching to 120% strain for 100 cycles. The current density is 1 mA/cm^2.

electrode materials, that is, SWNTs. We tested the SC performance under galvanostatic cycling with a variety of currents (Figure 3b). The linear voltage–time profile confirms the charging and discharging of the SCs. To evaluate the advantage of using porous textiles over flat plastic substrates, SCs were assembled and tested with a SWNT loading of 0.24 mg/cm^2 on a cotton sheet and on a polyethylene terephthalate (PET) substrate. The specific capacitance of SC with porous textiles is around 2–3 times better than that with PET substrates in the range of current density 20 μA/cm^2 to 20 mA/cm^2 (Figure 3c). Values of 140 F/g at 20 μA/cm^2 and 80 F/g at 20 mA/cm^2 are achieved in our conducting textiles, which are comparable to the highest specific capacitance achieved with SWNTs as electrode material.[15] The higher SC capacitance with SWNTs on textile is likely due to its better ion access in the porous structure. We also use the porous structure to load SWNTs with high mass in our SCs, up to 8 mg/cm^2. The areal capacitance increases with the areal mass (Figure 3d).[23,24] Compared with the reported values using SWNT electrodes in literature, our SCs reached much higher areal mass loading and areal capacitance with respect to the geometric area of the device. Our devices achieve an areal capacitance equal to 0.48 F/cm^2, the highest value reported for SWNTs and close to commercial values.[23−25]

Gravimetric energy density and power are important parameters for SCs in large-scale energy storage. Heavy metal strips are widely used as current collectors in com-

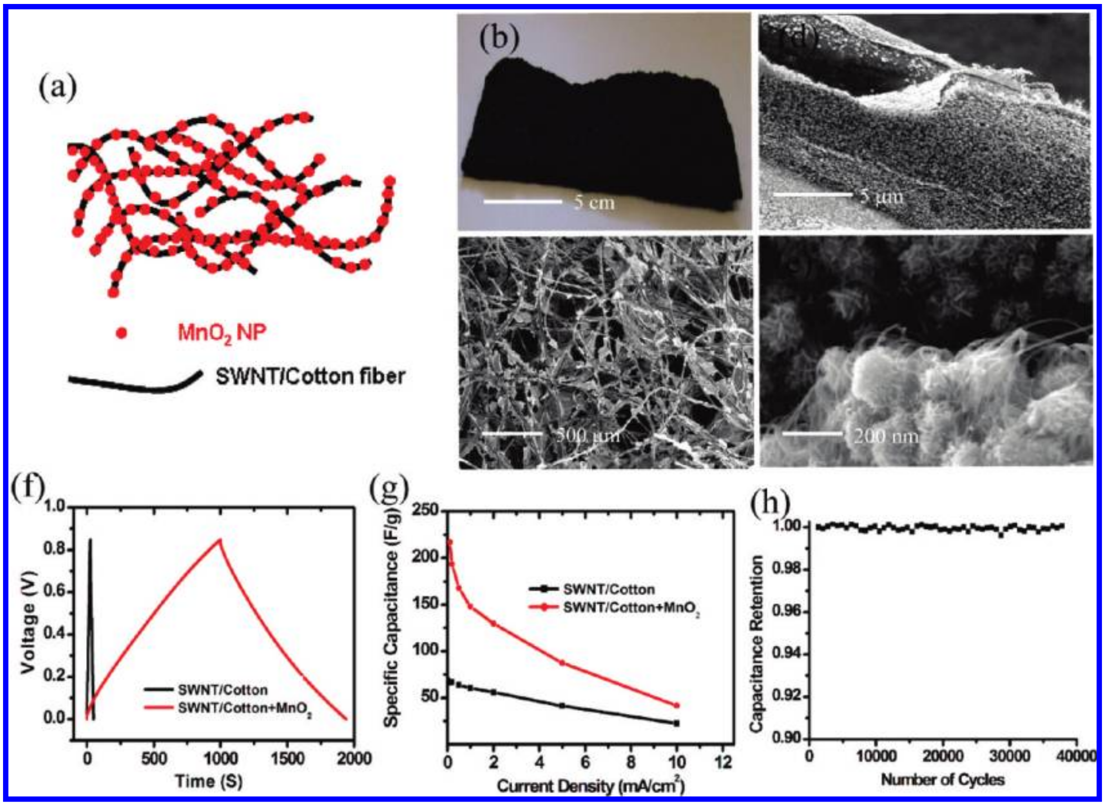

FIGURE 4. Loading pseudocapacitor or battery materials in porous conductor. (a) Schematic drawing of electrodeposition of MnO_2 onto the SWNT coated textile fibers. Due to the porous structure, the MnO_2 particles are coated on all the textile fibers including those in the interior of the textile. (b) A photo of MnO_2-coated SWNT/Cotton. (c) SEM of a top view of conductive textile after MnO_2 coating. (d) SEM of cotton fibers inside the textile after peeling the fiber layers apart, which shows that the MnO_2 nanoparticles coated the fibers in the interior of the textile, not just the surface layers. (e) High-magnification SEM image showing the flower structure of MnO_2 particles on SWNTs. (f) Charge−discharge of aqueous SC with SWNT/cotton electrodes and 2 M Li_2SO_4 as the electrolyte with current of 20 μA/cm^2. The areal capacitance increases by 24-fold after MnO_2 deposition. (g) Specific capacitance of SWNT/cotton with and without MnO_2 for different discharge current densities. (h) Cycling stability of a SC with SWNT−MnO_2 nanoparticles and porous textile conductor.

mercial SCs, with lightweight plastic and paper substrates emerging as their replacements.[15,23,24,26] The reported energy density and power density for SWNT electrodes in literature normally only includes the mass of SWNT, which is only ~4−10% of the total weight of the device.[23] Because of the lightweight of cotton at ~12 mg/cm^2, and the high mass loading of SWNTs allowed by the porous structure of cotton, the weight percentage of SWNTs in our full device is up to ~30%. In the Ragone plot (Figure 3e), the mass of electrode materials (~16 mg/cm^2), the cotton (~24 mg/cm^2), electrolyte (~6 mg/cm^2), and separator (~2 mg/cm^2) are all included in our complete SC device. The reference data indicated by the blue dot is by Niu et al.,[27] which is also based on the total mass of the full SC. Our SCs have >10 times higher specific energy than both SCs on metals and commercial SCs at the same specific power. For example, the specific energy reaches a high value of 20 Wh/kg at a specific power of 10 kW/kg. The specific power can be further improved by reducing the overall resistance down to ~0.1 Ω[28] through means of coating the textiles with even more conductive inks, such as inks that incorporate small amount of metal nanowires. Our textile SC devices show also extremely good cycling stability with only 2% variation and change in capacitance over a remarkably large cycle number of 130 000 cycles (Figure 3f). The initial increase of the specific capacitance upon cycling is likely due to the improvement in wetting of the SWNTs by the electrolyte. This cycling performance meets the requirements for commercial applications.[29] By using stretchable fabric sheets, instead of cotton sheets, as both the electrode substrates and the separator (Figure 3g), we were able to fabricate a fully stretchable SC. Figure 3h displays the specific capacitance of for a SC before and after being stretched up to 120% strain 100 times. The mass loading of SWNT is 0.24 mg/cm^2 and the specific capacitance for the stretchable SC is 62 F/g at 1 mA/cm^2. We observed no change in the specific capacitance after stretching. The stretchable SCs show high Coulomb efficiency (>99%) and good cycling stability, <6% decrease after 8000 cycles, which could be improved with better device sealing. Our stretchable SCs are attractive for powering the emerging stretchable or wearable electronics.[20]

One approach to further increase the specific energy of SWNT SCs is to incorporate pseudocapacitor materials such as MnO_2, RuO_2 or conducting polymers.[30,31] These pseudocapacitor materials have high reversible specific capacitance.

Previously, pseudocapacitor materials were either mixed with SWNTs, which significantly decreases the film conductivity, or coated on the surface of SWNTs but with only very small mass loading, ~0.03−0.2 mg/cm^2 to maintain good electrical contact with SWNTs.[32,33] In our case, as shown in the schematic in Figure 4a, MnO_2 was uniformly electrodeposited on the SWNTs. This was effective in significantly increasing the mass loading of the pseudocapacitor, while maintaining contact between SWNTs and providing a good electrical conduction path. MnO_2 was deposited along the conducting path which allowed intrinsically good contact with the SWNTs. Here the conductive cotton was utilized without mechanical pressing to take advantage of its open structure (Figure 4b). The open structure of cotton allows excellent deposition of the MnO_2 conformally along the fibers (Figure 4a, d, and e). The deposition is observed not only on the surfaces of the SWNT/cotton but also inside the layers of cotton fibers. The conductive cotton was peeled apart and SEM was taken of the interior cotton (Figure 4c). Conformal coating of MnO_2 on cotton fiber surface is observed, and the peeling leads to the partial delamination of MnO_2 from the cotton fibers (Figure 4c). Figure 4e reveals the flower structure of MnO_2 particles deposited on the SWNT surfaces. The MnO_2 nanoparticles are entangled with the SWNTs, which ensures good electrical contacts between them. To demonstrate the feasibility of this peudocapacitor approach for wearable power devices, SCs of SWNT/cotton with MnO_2 were tested with a 2 M aqueous Li_2SO_4 electrolyte. In this particular SC, the mass density is 0.24 mg/cm^2 for SWNTs and is 1.60 mg/cm^2 for the MnO2 which is 50 times higher than the reported value in literature.[32] The time required to charge the SCs for SWNT/cotton after MnO_2 deposition is significantly increased, suggesting a large charge capacity increase (Figure 4f). The areal capacitance with respect to the device increases by a factor of 24 after MnO_2 deposition. In literature, the electrodepostion of oxides typically increase the capacitance by a factor of less than 6.[34] The areal capacitance of the device reaches 0.41 F/cm^2, which is much higher than reported values with SWNT electrodes.[23−25] The specific capacitance data, considering the mass of both SWNTs and MnO_2 is plotted in Figure 4g. The specific capacitance when including the masses of both CNT and MnO_2 increases by a factor of 4 with MnO_2 deposition. Note that the specific capacitance of SWNTs here with Li_2SO_4 electrolyte is lower than the values in Figure 3b, which is likely due to better wetting between the organic electrolyte and SWNTs. Such wearable SCs with salt electrolyte also show excellent cycling stability (Figure 4h), with negligible change between the initial and the final specific capacitance over 35 000 cycles.

In this study, we report the use of porous conductive textiles as a platform for energy storage device applications. This porous conductor allows high mass loading of electrode materials and excellent access of electrolyte to those materials, which leads to outstanding device performance. Such textile-based power will boost the development and application of wearable electronics. With lightweight and cheap cotton or fabrics and the existing textile fabrication infrastructure, large-scale energy storage with low material and processing costs is also feasible. When combined with inks of other functional materials, other types of electronics could be created, such as wearable solar cells and batteries. These porous conductors with excellent properties will find a wide range of applications.

Acknowledgment. S.J. acknowledges support from The Korea Foundation for Advanced Studies. Y.C. acknowledges support from the King Abdullah University of Science and Technology (KAUST) Investigator Award (No. KUS-l1-001-12).

Supporting Information Available. This material is available free of charge via the Internet at http://pubs.acs.org.

REFERENCES AND NOTES

(1) Gniotek, K.; Krucinska, I. The basic problems of textronics. *Fibres Text. East. Eur.* **2004**, *12*, 13–16.

(2) Lukowicz, P.; Kirstein, T.; Troster, G. Wearable systems for health care applications. *Methods Inf. Med.* **2004**, *43*, 232–238.

(3) Park, S.; Jayaraman, S. Smart textiles: Wearable electronic systems. *MRS Bull.* **2003**, *28*, 585–591.

(4) Hyde, G. K.; Park, K. J.; Stewart, S. M.; Hinestroza, J. P.; Parsons, G. N. Atomic layer deposition of Conformal inorganic nanoscale coatings on three-dimensional natural fiber systems: Effect of surface topology on film growth characteristics. *Langmuir* **2007**, *23*, 9844–9849.

(5) Shim, B. S.; Chen, W.; Doty, C.; Xu, C. L.; Kotov, N. A. Smart Electronic Yarns and Wearable Fabrics for Human Biomonitoring made by Carbon Nanotube Coating with Polyelectrolytes. *Nano Lett.* **2008**, *8*, 4151–4157.

(6) Hecht, D. S.; Hu, L.; Gruner, G. Electronic properties of carbon nanotube/fabric composites. *Curr. Appl. Phys.* **2007**, *7*, 60–63.

(7) Sekitani, T.; et al. Stretchable active matrix organic light-emitting diode display using printable elastic conductors. *Nat. Mater.* **2009**, *8*, 494–499.

(8) *Handbook of Fiber Chemistry*; Lewin, M., Pearce, E. M., Eds.; Marcel Dekker Inc: New York, 1998.

(9) Pushparaj, V. L.; et al. Flexible energy storage devices based on nanocomposite paper. *Proc. Natl. Acad. Sci. U.S.A.* **2007**, *104*, 13574–13577.

(10) Yun, S.; Jang, S. D.; Yun, G. Y.; Kim, J. H.; Kim, J. Paper transistor made with covalently bonded multiwalled carbon nanotube and cellulose. *Appl. Phys. Lett.* **2009**, 95.

(11) Zhang, H.; et al. Regenerated-cellulose/multiwalled-carbon-nanotube composite fibers with enhanced mechanical properties prepared with the ionic liquid 1-allyl-3-methylimidazolium chloride. *Adv. Mater.* **2007**, *19*, 689–704.

(12) Islam, M. F.; Rojas, E.; Bergey, D. M.; Johnson, A. T.; Yodh, A. G. High weight fraction surfactant solubilization of single-wall carbon nanotubes in water. *Nano Lett.* **2003**, 269–273.

(13) Tracton, A. A. *Coating Handbook*; Marcel Dekker, Inc: New York, 2001.

(14) Hertel, T.; Walkup, R. E.; Avouris, P. Deformation of carbon nanotubes by surface van der Waals forces. *Phys. Rev. B: Condens. Matter* **1998**, *58*, 13870–13873.

(15) An, K. H.; et al. Supercapacitors using single-walled carbon nanotube electrodes. *Adv. Mater.* **2001**, *13*, 497.

(16) Parekh, B. B. F., G.; Eda, G.; Chowalla, M. Improved conductivity of transparent single-wall carbon nanotube thin films via stable postdeposition functionalization. *Appl. Phys. Lett.* **2007**, *90*, 121913.

(17) Zhou, W. V., J.; Nemes, N. M.; Fischer, J. E.; Borondics, F.; Kamaras, K.; Tanner, D. B. Charge transfer and Fermi level shift

in p-doped single-walled carbon nanotubes. *Phys. Rev. B* **2005**, *71*, 205423.

(18) Geng, H. K., K.; So, K.; Lee, Y.; Chang, Y.; Lee, Y. Effect of Acid Treatment on Carbon Nanotube-Based Conducting Films. *J. Am. Chem. Soc.* **2007**, *129*, 7758.

(19) Iijima, S. Structural Flexibility of Carbon Nanotubes. *J. Chem. Phys.* **1996**, *104*, 2089.

(20) Kim, D. H.; Rogers, J. A. Stretchable Electronics: Materials Strategies and Devices. *Adv. Mater.* **2008**, *20*, 4887–4892.

(21) Yu, C. M., C.; Rong, J.; Wei, B.; Jiang, H. Stretchable Supercapacitors Based on Buckled Single-Walled Carbon-Nanotube Macrofilms. *Adv. Mater.* **2009**, *21*, 1.

(22) Wakelyn, P. J.; et al. *Cotton Fiber Chemistry and Technology*; CRC Press: Boca Raton, FL, 2007.

(23) Futaba, D. N.; et al. Shape-engineerable and highly densely packed single-walled carbon nanotubes and their application as super-capacitor electrodes. *Nat. Mater.* **2006**, *5*, 987–994.

(24) M. Kaempgen, C. K. C.; Ma, J.; Gruner, G. Printable CNT thin film supercap. *Nano. Lett.* **2009**, *5*, 1872.

(25) Bonnefoi, L. S., P.; Fauvarque, J. F.; Sarrazin, C.; Dugast, A. Electrode optimization for carbon power supercapacitors. *J. Power Source* **1999**, *79*, 37.

(26) Hu, L.; Choi, J. W.; Yang, Y.; Jeong, S.; La Mantia, F.; Cui, L.-F.; Cui, Y. Highly Conductive Paper for Energy Storage Devices. *Proc. Nat. Acad. Sci. U.S.A.* (2009, Early Edition) DOI: 10.1073/pnas.0908858106.

(27) Niu, C. M.; Sichel, E. K.; Hoch, R.; Moy, D.; Tennent, H. High power electrochemical capacitors based on carbon nanotube electrodes. *Appl. Phys. Lett.* **1997**, *70*, 1480–1482.

(28) Lee, J. Y.; Connor, S. T.; Cui, Y.; Peumans, P. Solution-processed metal nanowire mesh transparent electrodes. *Nano Lett.* **2008**, *8*, 689–692.

(29) Conway, B. E. *Electrochemical Supercapacitors — Scientific Fundamentals and Technological Applications*; Kluwer Academic/Plenum: New York, 1999.

(30) Subramanian, V.; Zhu, H. W.; Wei, B. Q. Synthesis and electrochemical characterizations of amorphous manganese oxide and single walled carbon nanotube composites as supercapacitor electrode materials. *Electrochem. Commun.* **2006**, *8*, 827–832.

(31) Arabale, G.; et al. Enhanced supercapacitance of multiwalled carbon nanotubes functionalized with ruthenium oxide. *Chem. Phys. Lett.* **2003**, *376*, 207–213.

(32) Ma, S. A., K.; Lee, E.; Oh, K.; Kim, K. Synthesis and characterization of manganese dioxide spontaneously coated on carbon nanotubes. *Carbon* **2007**, *45*, 375.

(33) Wang, Y.; Zhitomirsky, I. Electrophoretic Deposition of Manganese Dioxide-Multiwalled Carbon Nanotube Composites for Electrochemical Supercapacitors. *Langmuir* **2009**, *25*(*17*), 9684—9689.

(34) Deng, G. H.; Xiao, X.; Chen, J. H.; Zeng, X. B.; He, D. L.; Kuang, Y. F. A new method to prepare RuO2 Æ xH2O/carbon nanotube composite for electrochemical capacitors. *Carbon* **2005**, *43*, 1566–1569.