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Transparent and Flexible Supercapacitors with Single Walled Carbon Nanotube Thin Film Electrodes

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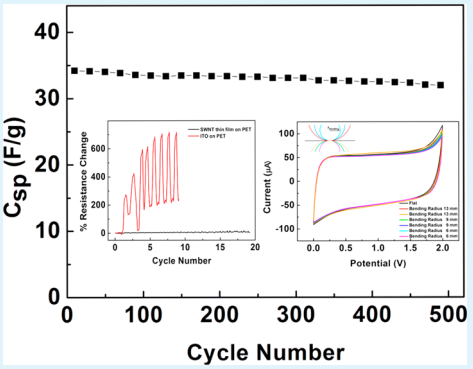
ABSTRACT: We describe a simple process for the fabrication of transparent and flexible, solid-state supercapacitors. Symmetric electrodes made up of binder-free single walled carbon nanotube (SWCNT) thin films were deposited onto polydimethylsiloxane substrates by vacuum filtration followed by a stamping method, and solid-state supercapacitor devices were assembled using a gel electrolyte. An optical transmittance of 82% was found for 0.02 mg of SWCNTs, and a specific capacitance of 22.2 F/g was obtained. The power density can reach to 41.5 kW·kg^{-1} and shows good capacity retention (94%) upon cycling over 500 times. Fabricated supercapacitors will be relevant for the realization of transparent and flexible devices with energy storage capabilities, displays and touch screens in particular.



KEYWORDS: *supercapacitors, single walled carbon nanotubes, transparent, flexible, polydimethylsiloxane, thin film*

1. INTRODUCTION

Supercapacitors have received a lot of attention due to their high specific power and moderate energy densities. They have a wide range of applications spanning from electric vehicles and pulse power systems to portable devices.[1−3] Significant efforts have been spent in the development of the basic components of many flexible and transparent electronic devices, opening possibilities for new device concepts and form factors; however, energy and power sources in those devices have retained their classic form factor. Therefore, it is of great interest to develop both transparent and flexible supercapacitors.[4−9]

Single walled carbon nanotube (SWCNT) thin film electrodes are a highly appealing candidate material for this purpose.[6,8] Owing to their high conductivity, permeability (resulting in high power density) and chemical inertness (long cycle lifetime), SWCNT thin films are promising candidates for active supercapacitor electrode materials.[7,11,12] For practical device applications, these characteristics together with the fact that carbon typically forms a purely double layer make SWCNT thin films unique. Other alternatives like pseudocapacitive materials, such as metal oxides (e.g., RuO_2, IrO_2 and MnO_2) and conducting polymers (polyaniline and polypyrrole), tend to suffer from reduced cyclability and power densities.[7,10,12] SWCNT thin film electrodes, in fact, have already been successfully demonstrated in prototype devices, such as solar cells,[13] photodetectors[14] and organic light emitting diodes.[15] An alternative transparent electrode candidate, conducting polymers, revealed high capacitance but showed rapid degradation, volumetric changes and limited cycle life.[16] In addition, conducting polymer electrodes are limited to work in a strict potential window. Possible overcharging and discharging during operation could easily damage the conducting polymers.[12] Chemical routes, on the other hand, have difficulties in the synthesis of conducting polymers.[12,17] Many conducting polymers have more than one oxidation step. Moreover, some polymers should be doped to increase their conductivities.[5,18] Charged state and the doping nature of the conducting polymers affect their electrochemical performance. Because of this, they are suited only for particular electrode and/or electrolyte systems. Therefore, it is difficult to use conducting polymers as electrodes in a symmetric supercapacitor assembly.[19]

Metal-oxide supercapacitors have a high specific capacity, but they are not inherently conducting and necessitate the use of metallic or conductive fillers in a composite structure.[20,21] In addition, the optoelectronic properties and especially flexibility of thin film crystalline metal oxide (e.g., MnO_2,[13] RuO_2,[22] ITO[23]) supercapacitors are far from that of the SWCNT thin film supercapacitors.[7,24] SWCNT thin film electrodes can be made highly transparent and can be simply deposited onto polydimethylsiloxane (PDMS) and polyethylene terephthalate (PET) substrates for the realization of flexible electrodes. The sheet resistance of SWCNT films can simply be tuned with the

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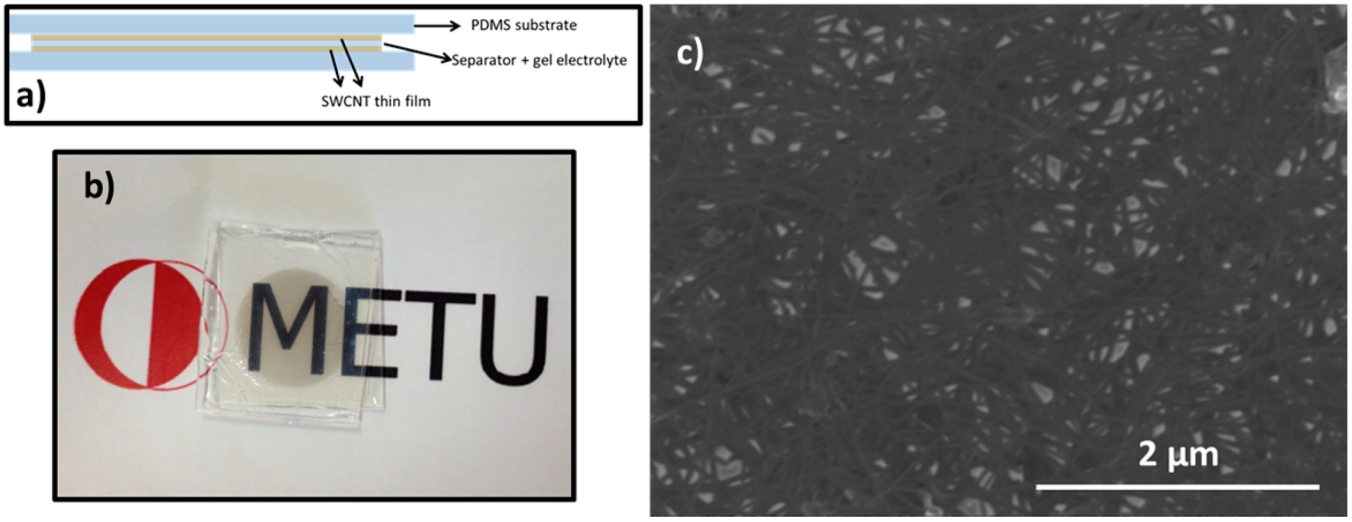

Figure 1. (a) Simple schematic and (b) photograph of the fabricated supercapacitors without carbon paste current collectors. (c) SEM image of a SWCNT thin film with a sheet resistance of 75 ohm/□.

nanotube density. Attained values are competitive to that of commercially used indium tin oxide (ITO) thin films.

Electrolytes are another fundamental component of supercapacitors. Aqueous electrolytes have high ionic conductivity, but they have distortion at near 1 V due to the electrolysis of water. Thus, they have a narrower voltage window than organic electrolytes.[25] Besides, they can damage electrode active materials and metallic contacts in the long-term. In addition, liquid electrolytes have the risk of leakage, which could deteriorate electrochemical performance and is harmful. This adds the requirement of stricter packaging configurations. Solid electrolytes are available, but their room temperature conductivity is lower than that of liquid electrolytes for high power applications. Organic gel electrolytes provide a good balance. They are physically more robust than liquid electrolytes while maintaining good ionic conductivity. In addition, organics also permit a large potential window up to 2.5 V.[26] The most commonly used gel electrolytes are composed of H_3PO_4 (or H_2SO_4) and PVA (poly(vinyl alcohol)).

In this work, we fabricate flexible and transparent SWCNT thin film based supercapacitors using a polymer based gel electrolyte on PDMS substrates. High electrical conductivity of SWCNT thin films allowed the possibility of fabricating supercapacitors without a separate charge collector.

2. EXPERIMENTAL DETAILS

PDMS preparation was done using Dow Corning's Sylgard 184 elastomer kit. PDMS base is a viscous liquid and becomes rigid and flexible upon the addition of the curing agent. Depending on the preparation parameters and thickness, one can tune its flexibility. PDMS base and the curing agent were mixed at a ratio of 10:1 in a Petri dish. Following mixing, PDMS was kept at room temperature for 1 h and then under vacuum for 12 h. Elastomeric PDMS substrate was then cut into small pieces. Size and thickness of the PDMS substrates were 2 × 2 and 0.3 cm, respectively.

SWCNT (P3 Carbon Solutions Inc.) thin films were deposited onto polydimethylsiloxane (PDMS), and polyethylene terephthalate (PET) substrates by vacuum filtration and consecutive stamping method.[27,28] As a substrate, PDMS is good in terms of its flexibility, chemical inertness and optically transparency. It also provides strong adhesion to various surfaces. It has already been used in microelectromechanical systems and microfluidic device fabrication, soft lithography, contact lens manufacturing and device encapsulation.[29−35]

Following deposition, SWCNT thin films were treated with nitric acid (HNO_3, 65%) for 3 h to improve their conductivity. The resulting circular thin films had an area of 2 cm^2. Sheet resistance and transmittance of the prepared SWCNT thin films was controlled with the filtration volume.

The composition of the gel electrolyte used in the SWCNT supercapacitors was $TBAPF_6$ (tetrabutylammoniumhexafluorophosphate):PMMA (poly(methyl methacrylate)):PC (propylene carbonate):ACN (acetonitrile) in a ratio of 3:7:20:70 by weight. The gel electrolyte was prepared by first dissolving $TBAPF_6$ and PMMA in ACN and slowly evaporating the ACN to reach a honey-viscous condition. A few drops of PC were added to decrease the vapor pressure of the gel electrolyte yielding a conducting transparent gel (~3 mS/cm).[26] It is worth noting that our electrolyte was stable with the PDMS substrates.

To fabricate supercapacitors, conductive carbon paste was used as an external current collector. A commercial Celgard separator (Celgard 3401) was used as the separator. It was soaked in the gel electrolyte and sandwiched between two SWCNT electrodes as shown in the schematic provided in Figure 1a. The supercapacitor was then sealed with epoxy.

The structure and morphology of the fabricated SWCNT thin films were characterized by scanning electron microscopy, (SEM, FEI Nova Nano SEM 430, operated at 10 kV). Electrochemical properties of the supercapacitors were investigated in a two-electrode configuration using potentiostat/galvanostat Gamry 3000 setup 48 h after fabrication. All measurements were conducted within 0 and 2 V potential window provided through the use of an organic gel electrolyte. Optical transmittance measurements were conducted using a Maya2000 Pro Spectrometer.

3. RESULTS AND DISCUSSION

A schematic and photograph of the fabricated supercapacitors are provided in Figure 1a,b, respectively. The full device without external conductive carbon paste current collectors is highly transparent, as can be seen in Figure 1b. The morphology of the SWCNT thin film uniformly covering the PDMS substrate surface is shown in the SEM image provided in Figure 1c. Four sets of transparent and flexible devices with different SWCNT densities were fabricated in this work. Sheet resistance of the thin films, their optical transmittance and corresponding SWCNT weights are tabulated and provided in Table 1.

Table 1. Sheet Resistance and % Transmittance (at 550 nm) Change with Respect to SWCNT Weight for the Fabricated Supercapacitors

SWCNT mass (total)	sheet resistance (ohm/□)	% transmittance
0.02	260	82
0.04	210	75
0.06	155	63
0.08	75	56

Gel electrolyte with a composition of salt:PMMA:ACN:PC has high transmittance and ionic conductivity. Controlled viscosity of the gel electrolyte allowed stable electrochemical performance during bending. Instead of commonly used H_3PO_4/PVA, the use of $TBAPF_6$/PMMA/PC/ACN gel electrolyte with a wettable separator is an important point of our study and a major reason in obtaining high transparency on full devices. To our knowledge, it is the first time that this gel electrolyte composition is used in a supercapacitor application. It provides higher transparency and a higher voltage window as compared to conventionally used H_3PO_4/PVA or H_2SO_4/PVA gel electrolytes.

Transmittance spectra for the fabricated supercapacitors with different SWCNT densities are provided in Figure 2. A

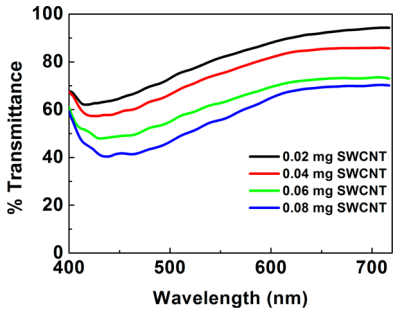

Figure 2. Optical transmittance characteristics of the full devices with respect to total SWCNT weight in both electrodes.

transmittance of 82% (at a wavelength of 550 nm) was obtained for the supercapacitors with 0.02 mg of SWCNTs. Transmittance was found to decrease with SWCNT density as shown in the figure, in agreement with the transparent supercapacitors in literature.[6,7,36]

Cyclic voltammograms of the resulting transparent and flexible supercapacitors are shown in Figure 3a. The cyclic voltammetry measurements revealed almost rectangular shapes at a scan rate of 20 mV·s^{-1}, as shown in Figure 3a. Denser films with higher conductivities showed higher capacitance values as compared to films with lower densities. The cyclic voltammetry measurements obtained at different scan rates are provided in Figure 3b. The deviation from the ideal capacitor shape was attributed to the internal resistance of the devices. The curves also show small and broad current peaks, which we ascribe to stray pseudocapacitance resulting from the functional groups on the SWCNTs or from the residual surfactant used for the thin film preparation process. Devices were found to be unstable above 2 V, which we attribute to the presence of functional groups on the SWCNTs reacting with the electrolyte beyond this voltage and instability of gel electrolyte components.

The functional and mechanical integrity of the fabricated supercapacitors was also investigated by characterizing electrochemical properties while bending the device down to different radii of curvature. This is made possible by the flexible nature of both electrodes and electrolyte. To investigate the flexibility of SWCNT thin films, resistance change in SWCNT thin film on PET substrates was monitored, provided in Figure 4a, during bending cycles.[37] A commercial ITO thin film on a PET substrate (Delta Technologies, 4−10 ohm) was used as a reference. SWCNT thin films provided higher flexibility than that of crystalline ITO films due to their web-like structure and mechanical stability of the individual SWCNTs. This is evidenced in the stable resistance of the SWCNT thin films upon bending, while the resistance of ITO thin film increases 7 times within 10 bending cycles. Cyclic voltammograms of the fabricated supercapacitors as a function of radius of curvature are shown in Figure 4b. Thin film supercapacitors on PDMS substrates were found to be flexible, and they could be repeatedly bent without significant variations in electrochemical properties including specific capacitance, power and energy density. The measurements indicated that mechanical bending does not have a significant influence on the ionic transport within the gel electrolytes or the quality of the interface between the gel electrolyte and SWCNT electrodes. Owing to easy ion transport and wide electrochemical window of gel electrolyte, flexed supercapacitors showed similar results to the flat counterparts.

The galvanostatic charge/discharge characteristics of the supercapacitors with respect to SWCNT loading measured at a current density of 1.25 A/g is shown in Figure 5a. A small internal resistance drop (IR_{drop}) was observed in measurements. The IR_{drop} was caused by equivalent series resistance, which includes electrode resistance, electrolyte resistance and contact resistance between the electrode and the electrolyte.[38] A significant increase in specific capacitance (F/g) with increasing SWCNT loading was expected due to the improved overall

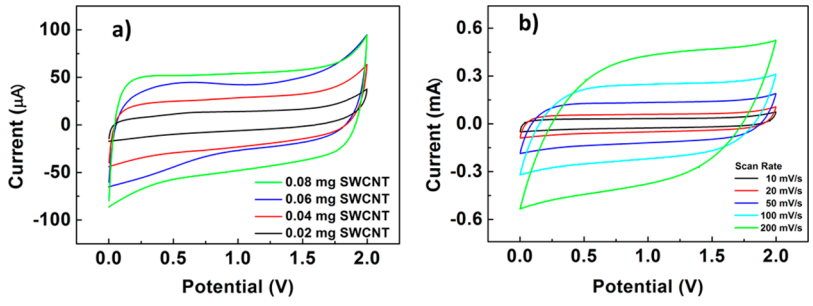

Figure 3. (a) Cyclic voltammograms of transparent and flexible supercapacitors with different SWCNT mass at a scan rate of 20 mV·s^{-1}. (b) Cyclic voltammograms at different scan rates for a SWCNT mass of 0.08 mg.

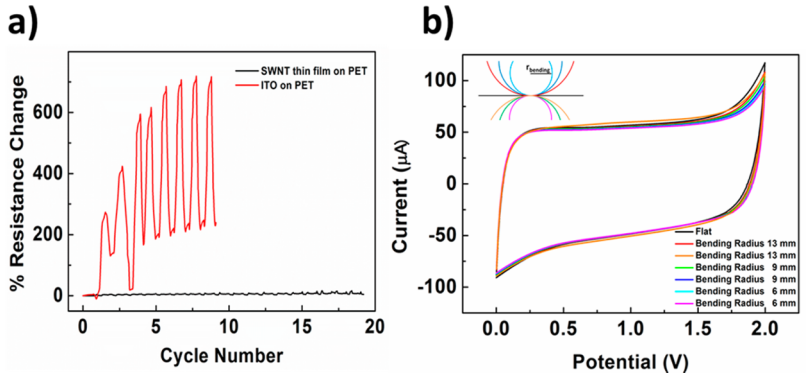

Figure 4. (a) Percent resistance change in ITO and SWCNT thin films on ITO during bending cycles down to a radius of 6 mm.[35] (b) Cyclic voltammograms for flexible supercapacitors while bent to different radius of curvatures for the 0.08 mg of SWCNT thin film at a scan rate of 20 mV·s^{-1}.

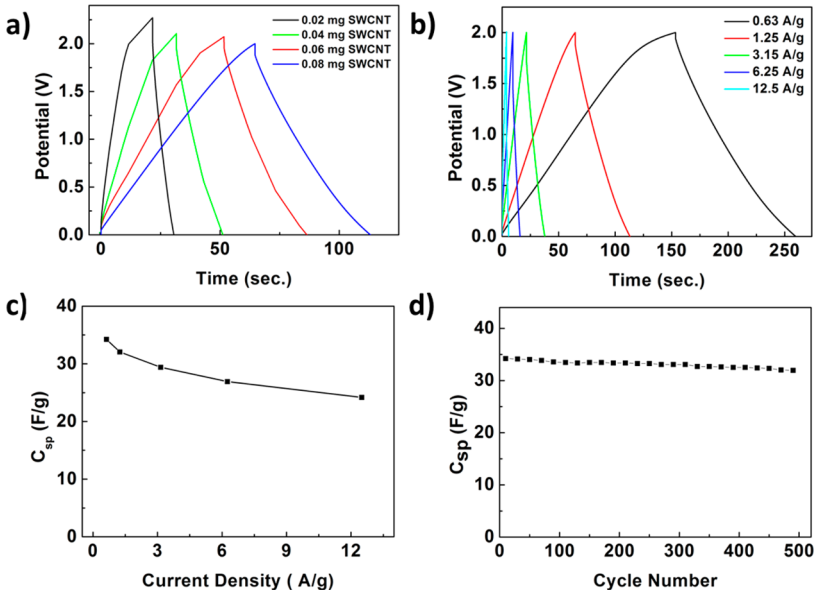

Figure 5. (a) Galvanostatic charge–discharge profile for supercapacitors at 1.25 A/g and (b) at different current densities. (c) Specific capacitance change with respect to potential scan rate. Lines are for visual aid. (d) Cycle performance of flexible supercapacitors at a current density of 1.25 A/g.

charge storage capacity. Porous nature of the SWCNT films facilitates the gel electrolyte access to the active area.[20] The galvanostatic charge/discharge characteristics measured at various current densities are provided in Figure 5b. Discharge profile of the fabricated supercapacitors was found to depend on the applied current and similar curve shapes have been obtained for different current densities.

The cell capacitances (C_{cell}) were calculated from cyclic voltammograms (Figure 3a) using the following equation:

$$C_{cell} = I/(dV/dt)$$

where I is the obtained current, m is the mass of SWCNTs on both electrodes and dV/dt is the scan rate. The capacitance of the supercapacitor with a SWCNT mass of 0.02 mg was calculated to be 25.2 F/g, whereas that for 0.08 mg SWCNT mass was 31.9 F/g. These measurements were taken at a scan rate 20 mV·s^{-1}. The specific capacitance (C_{sp}) was calculated using the galvanostatic charge–discharge curves of supercapacitors provided in Figure 5a and the following equation:

$$C_{sp} = I/m(dV/dt)$$

,where I is the applied discharge current, t is the discharge time, V is the discharge voltage and m is the mass of SWCNTs on both electrodes (10). C_{sp} estimated at a current density of 0.63 A/g was 34.2 F/g for the supercapacitor device with a SWCNT mass of 0.08 mg. Specific capacitance is plotted as a function of the discharge current density in Figure 5c. A serious specific capacitance loss was found to occur with increasing current density. It is commonly observed that the capacitive performance of electrode active materials decrease with an increase in current density. This is due to the difficulties in penetration and diffusion of electrolyte ions into a poorly conductive layer and/or thick electrode films.[39] The specific capacitance was 36.9 F/g at a potential scan rate of 10 mV·s^{-1}, and it decreased to 25.1 F/g at a potential scan rate of 200 mV·s^{-1} for the supercapacitors with 0.08 mg SWCNTs.

Capacity retention is another key factor in determining the supercapacitor performance. To explore this, fabricated transparent and flexible supercapacitors were charged and discharged over 500 cycles. No significant degradation in specific capacitance was obtained, as shown in Figure 5d. Quantitatively, 94% of C_{sp} was retained over 500 cycles, resembling the decent capacity retention in the fabricated devices.

Our results showed that SWCNT thin film electrodes work well in supercapacitor devices with nonaqueous gel electrolytes. No damage was visually observed on the SWCNTs and carbon paste contact points due to contact with the electrolyte at the end of the measurements. This is an important feature of our supercapacitors because the polymer-based gel electrolyte enabled the fabrication of transparent and flexible supercapacitors without any deterioration in performance.

Energy density and power densities of the fabricated supercapacitors were calculated using the following equations:

$$E_{cell} = (C_{cell}V^2)/2m$$

$$P_{cell} = V^2/(4\cdot ESR\cdot m)$$

,where V is the applied potential, m is the total mass of SWCNTs in both electrodes and the ESR is effective serial resistance calculated from Figure 5a using the following equation:

$$ESR = IR_{drop}/2I$$

High ESR of the supercapacitors was attributed to the absence of a separate charge collector, serial resistance of the SWCNT thin films, resistance of the gel electrolyte and the contact resistance between the carbon paste contact with SWCNTs. High flexibility and strong physical adhesion between SWCNTs and PDMS substrate preserved the specific capacitance and power density without any deterioration upon bending. Owing to the unique properties of SWCNTs, good electrochemical performance was obtained from the fabricated supercapacitors. Minimizing the ESR would increase the specific capacitance and power density. Specific capacitance, power density and energy densities were found to be 34.2 F/g, 21.1 $kW\cdot kg^{-1}$ and 18.0 $kWh\cdot kg^{-1}$, respectively. According to the Ragone plot, power density of the fabricated supercapacitors with gel electrolytes was found to lie in a low power range. Truly flexible supercapacitor devices necessitate use of solid-state electrolytes to prevent leakage. When compared with other solid-state supercapacitors in literature,[8,40] our results might seem moderate in terms of specific capacitance. However, when transparency is concerned, our results are found to be quite competitive to those in literature.[4,6] In this work, SWCNT thin films were used both as the current collector and host for the double-layer. Therefore, an increase in specific capacitance would require higher SWCNT mass, which necessitates sacrifice in transparency. Flexibility and transparency, simultaneously, would not be possible if a separate current collector was used. It is possible to further improve the transparency of the fabricated devices through the reduction of SWCNT bundles within the thin films. These absorb a significant proportion of the light and are responsible for a decrease in optical transmittance of the thin films. Power densities can be improved through the deposition of nanosized oxide particles and/or polymers onto SWCNTs.

4. CONCLUSIONS

In this study, we have demonstrated solid-state, highly flexible and transparent supercapacitors with binder-free SWCNT thin film electrodes. We have examined the effect of SWCNT density, thus the SWCNT mass, on the electrochemical properties of the supercapacitors. SWCNT density controlled the sheet resistance of the electrodes and transmittance of the overall devices. A typical EDLC behavior in a potential window of 0 to 2 V was obtained. High conductance of the SWCNT thin films eliminated the use of extra charge collectors. High bendability without significant deterioration in device properties was obtained. The measured specific capacitance, maximum power and energy density were 34.2 F/g, 21.1 $kW\cdot kg^{-1}$ and 18.0 $kWh\cdot kg^{-1}$, respectively for 0.08 mg SWCNT devices. The supercapacitors showed excellent stability and the variation in C_{sp} was found to be less than 6% over 500 charge/discharge cycles. In addition, the specific capacitance was found to change by less than 2% through bending the devices down to a radius of 6 mm. Finally, a low cost and rapid route for the fabrication of supercapacitors is presented. These supercapacitors will be critical components of transparent and flexible devices.

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Notes

The authors declare no competing financial interest.

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