Flexible, transparent single-walled carbon nanotube transistors with graphene electrodes



2010 Nanotechnology 21 425201

(http://iopscience.iop.org/0957-4484/21/42/425201)

Nanotechnology **21** (2010) 425201 (5pp) doi:10.1088/0957-4484/21/42/425201

# Flexible, transparent single-walled carbon nanotube transistors with graphene electrodes

**Sukjae Jang[1], Houk Jang[1], Youngbin Lee[1], Daewoo Suh[1], Seunghyun Baik[1,2], Byung Hee Hong[1,3] and Jong-Hyun Ahn[1,4]**

[1] SKKU Advanced Institute of Nanotechnology (SAINT) and Center for Human Interface Nano Technology (HINT), Sungkyunkwan University, Suwon 440-746, Korea
[2] School of Mechanical Engineering, Sungkyunkwan University, Suwon 440-746, Korea
[3] Department of Chemistry, Sungkyunkwan University, Suwon 440-746, Korea
[4] School of Advanced Materials Science and Engineering, Sungkyunkwan University, Suwon 440-746, Korea

E-mail: ahnj@skku.edu and byunghee@skku.edu

Received 30 July 2010, in final form 30 August 2010
Published 22 September 2010
Online at stacks.iop.org/Nano/21/425201

**Abstract**
This paper reports a mechanically flexible, transparent thin film transistor that uses graphene as a conducting electrode and single-walled carbon nanotubes (SWNTs) as a semiconducting channel. These SWNTs and graphene films were printed on flexible plastic substrates using a printing method. The resulting devices exhibited a mobility of $\sim$2 cm$^2$ V$^{-1}$ s$^{-1}$, On/Off ratio of $\sim 10^2$, transmittance of $\sim$81% and excellent mechanical bendability.

(Some figures in this article are in colour only in the electronic version)

## 1. Introduction

Flexible and transparent thin film transistors (TTFTs) have great potential in future display applications requiring both flexibility and transmittance, such as head-up and see-through displays embedded on the windshield of a car [1–3]. The traditional group of materials used to build TTFTs is the conducting (or semiconducting) oxides, including $SnO_2$, $In_2O_3$, ZnO, and CdO [4, 5]. These oxide materials provide good carrier mobility but generally lack mechanical flexibility. On the other hand, organic materials that can utilize low temperature processing are useful for flexible TTFTs, but their modest carrier mobility and environmental durability limit their range of application [6–8]. One of the promising alternatives is carbon based materials, such as single-walled carbon nanotubes (SWNTs) and graphene [1, 9–12]. These materials exhibit excellent mechanical and electrical properties owing to their unusual molecular structure [13–16]. As a representative example, transparent and flexible TFTs that use highly conductive carbon nanotube films as the electrodes of transistors were demonstrated with the transfer printing technique [9, 10]. This paper reports transparent, flexible TFTs with excellent performance, fabricated at room temperature using SWNT semiconducting thin films and graphene electrodes grown by chemical vapor deposition.

## 2. Experimental procedure

Figure 1 shows the device fabrication procedures, materials, and device structure as well as scanning electron microscopy (SEM) and optical microscope images. First, a purified SWNT solution (Nanointegris, IsoNanotubes-S™, semiconductor purity of 95%) was dropped onto a $SiO_2$ (thickness: 100 nm)/Si substrate and then placed in water for 30 min to remove the disperser and stabilize the adhesion between the SWNTs and the substrate. Next, a PET substrate coated with a graphene gate (G) electrode, and two intermediate films including graphene for the source (S)/drain (D) electrodes and SWNTs for the channels were prepared. For the S/D/G electrodes, two types of graphene film synthesized on a Ni(300 nm)/$SiO_2$(300 nm)/Si wafer and Cu foil (25 $\mu$m) catalyst layer by chemical vapor deposition at 1000 °C were used as the S/D electrodes and G electrode, respectively [17–19]. The average number of graphene layers grown on the Ni catalyst and Cu foil ranged from three to five and from mono- to bilayer, respectively. The graphene films grown on Cu foil for the G electrode were transferred to PET

 

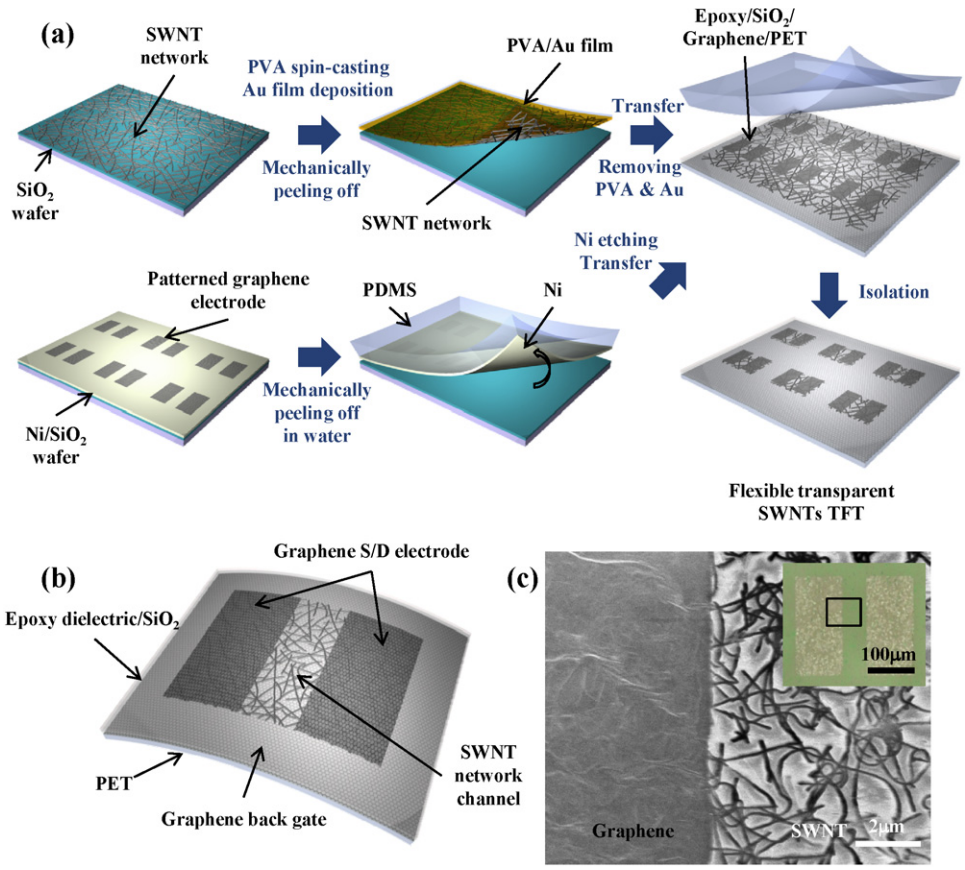


**Figure 1.** (a) Schematic diagram of the fabrication procedures for flexible and transparent thin film transistors that use SWNT networks for the semiconducting layers and graphene for the S/D/G electrodes. The process involves transfer printing of SWNTs and graphene to form the semiconducting and conducting layers of the devices. (b) Schematic structure of the flexible TTFTs on the plastic substrate. (c) SEM image of the interface between a source/drain graphene electrode (left) and SWNT network channel layer (right). Inset: optical microscope image of a typical device. Black box indicates the magnified region by SEM.

(thickness: 188 $\mu$m) substrates using a PMMA wet transfer method [20]. The graphene films on Ni for the S/D electrodes were printed on a poly(dimethylsiloxane) (PDMS) stamp after direct formation of the S/D electrode patterns on Ni films by conventional photolithography and reactive ion etching (RIE) with $O_2$ plasma [18].

To transfer the SWNTs film from the wafer to the target plastic substrate, an Au thin film (thickness: 20 nm) and a 10 wt% poly (vinyl alcohol) (PVA) solution were formed on the SWNTs [21, 22]. The PVA/Au/SWNTs film attached to a PDMS stamp with patterned graphene S/D electrodes can be peeled off easily. After dissolving PVA in hot water (80 °C) and etching Au, a SWNTs film on a graphene/PDMS stamp was obtained. These SWNTs/graphene layers were transfer printed on $SiO_2$ (thickness: 100 nm)/graphene/PET substrates coated with a thin photocurable epoxy adhesive layer (SU8-2, Microchem Corp., thickness: 500 nm). Here, epoxy and $SiO_2$ served as a double layer gate dielectric. After completing all the printing processes, the devices were isolated by conventional photolithography and reactive ion etching with $O_2$ plasma.

Figure 1(b) shows a schematic diagram of the device structure of the SWNTs semiconducting channels and graphene electrodes. Figure 1(c) shows a SEM image of the boundary between the S/D graphene electrode (left) and SWNTs network channel layer (right) on the PET substrates. This image indicates that the SWNTs are dispersed completely with a density of $\sim$3 tubes $\mu m^{-2}$ and the S/D graphene electrodes cover the SWNTs perfectly. The inset shows an optical microscopy image of a typical device. The channel length of this device was 50 $\mu$m with a corresponding width of 200 $\mu$m. The black box indicates the region of the device magnified by SEM.

## 3. Results and discussion

Figure 2(a) shows the transmission spectra in the visible range of all the carbon based thin film transistors, in which SWNT networks and graphene films serve as the channels and electrodes, respectively. The average transmittance through the channel region (SWNT/epoxy/$SiO_2$/graphene) and source/drain region (graphene/SWNT/epoxy/$SiO_2$/graphene) at 550 nm, exclusive of the bare PET substrate effect, were 91.3% and 81.4%, respectively. The transmittance of the source/drain region decreased by 9.9% compared to the channel. This corresponds to the opacity of visible light through four layers of graphene because a monolayer film leads to an approximate 2.3% decrease in transmittance [23]. In addition, all the carbon based devices showed less variation

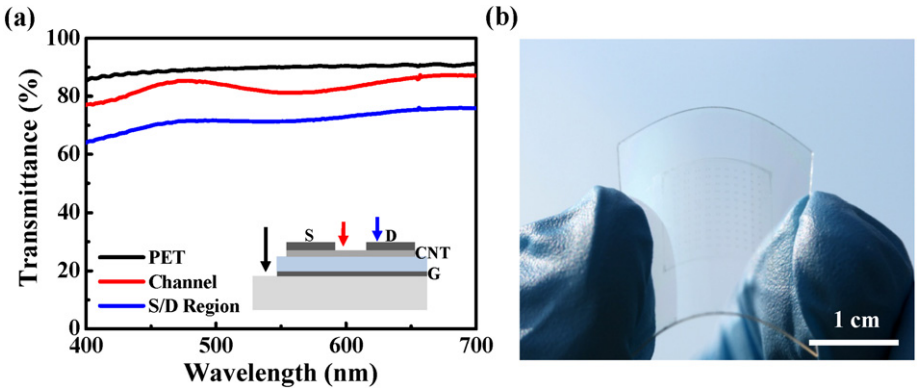


**Figure 2.** (a) Optical transmittance as a function of wavelength for the PET substrate (black line), through the channel region (red line) and through the source/drain electrode (blue line) of the device including PET substrates. (b) Optical images of a completed array of the TTFTs on the PET substrate.

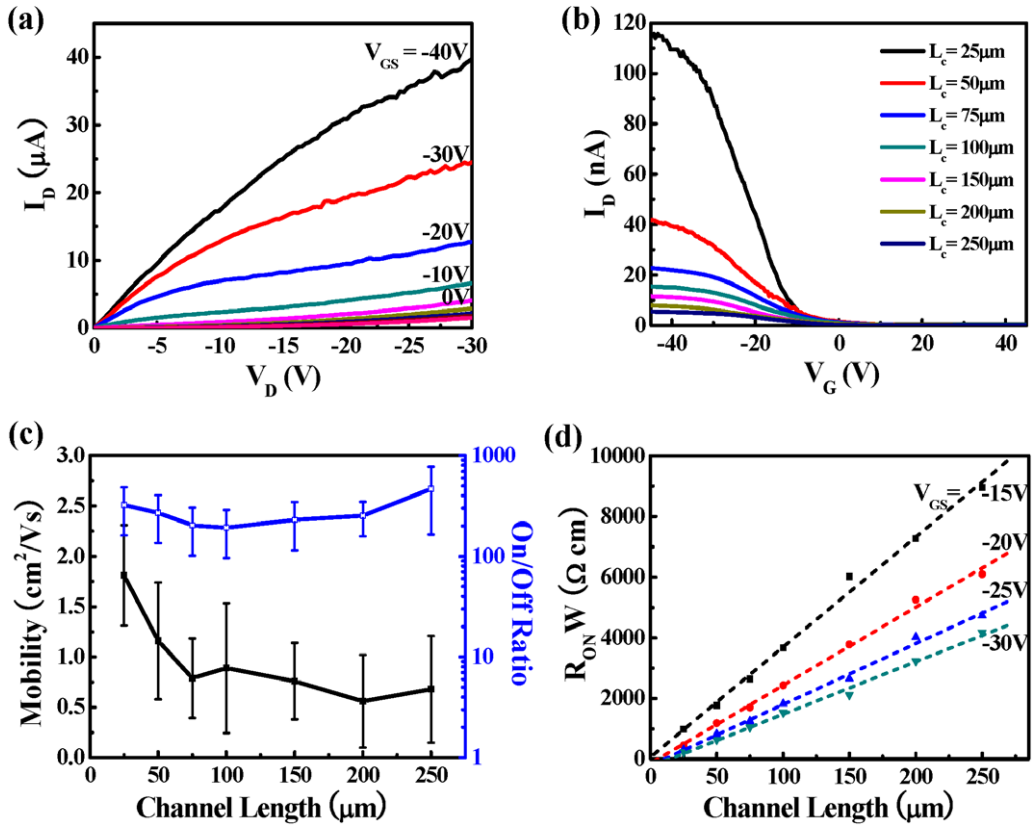


**Figure 3.** (a) Current–voltage characteristics of a TTFT with a 200 $\mu$m channel width and a 25 $\mu$m channel length (gate voltage, $V_{GS}$: $-40$ to 40 V from top to bottom; 10 V steps). (b) Transfer characteristics of TTFTs with different channel lengths on the PET substrate. The channel widths were 200 $\mu$m and the drain/source voltage ($V_{DS}$) was $-0.1$ V in all cases. (c) Effective device mobility and on/off ratios as a function of the channel length. (d) ON channel resistance as a function of the channel length at different gate voltages (gate voltage, $V_{GS}$: $-15$ to $-30$ V from top to bottom; 5 V steps).

in transmittance than ITO and a comparable degree of transparency to oxide material based devices [24]. Figure 2(b) shows an image of the device substrate to demonstrate the optical transmittance and mechanical bendability of these devices.

Figure 3(a) shows the current–voltage characteristics of a representative device with a 200 $\mu$m channel width and 25 $\mu$m channel length (gate voltage, $V_{GS}$: $-40$ to 40 V from top to bottom; 10 V steps). The unipolar p-type devices showed an effective mobility of $\sim$2 cm$^2$ V$^{-1}$ s$^{-1}$, on/off ratio of $\sim 10^2$ and threshold voltage of $\sim -7$ V. Figure 3(b) presents the transfer characteristics of the devices with different channel lengths ($L_C$) between 25 and 250 $\mu$m on a PET substrate. The channel widths were 200 $\mu$m and the drain/source voltage ($V_{DS}$) was $-0.1$ V in all cases. The $\sim$90% devices exhibited stable operation without failure. Device failure was caused mainly by gate leakage. Figure 3(c) summarizes the effective device mobility and on/off ratios of the devices as a function

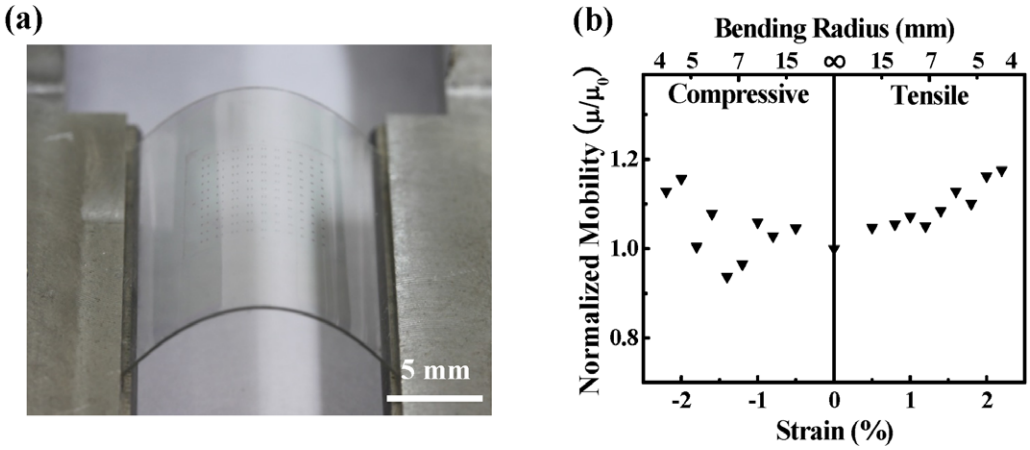


**Figure 4.** (a) Optical image during the bending test. (b) The change in the normalized effective device mobility $\mu/\mu_0$ for TTFTs as a function of the bending induced strain and bending radius.

of the channel length. The on/off ratios ranged from $10^2$ to $10^3$ and the effective mobility ranged from 0.5 to 2 $cm^2\ V^{-1}\ s^{-1}$, indicating that these values are not dependent on the channel lengths [25–27]. This channel length invariance of the mobility indicates that the contact resistance has a negligible effect on the devices. More detailed analysis using standard transmission line models was performed to evaluate the contact resistance between graphene and SWNT [28]. Figure 3(d) shows the linear scaling of the on-state resistance ($R_{ON}$) as a function of the channel length at different gate voltages (gate voltage, $V_{GS}$: −15 to −30 V from top to bottom; 5 V steps). The contact resistance, as determined from the intercept of the linear fit of the $R_{ON}$ versus $L_C$ plot in the linear region, was negligible compared to the channel resistance within the range of channel lengths investigated. This small resistance suggests good electrical contacts between the SWNTs and graphene electrodes. Graphene possesses a molecular structure basically identical to that of SWNTs, and the strong interaction between graphene and SWNTs could result in excellent interface contact. The relatively low on/off ratio of these devices results from the metallic pathway included in a random network of CNTs, which can be improved by higher purity refining. In addition, the use of aligned nanotubes instead of random networks as a channel can improve the mobility of devices simultaneously [1, 25, 29, 30].

To confirm the performance of all carbon based transistors for flexible device applications, electrical measurements were performed under bending conditions. Figures 4(a) and (b) show optical images of the bent device under tensile strain and the device mobility in the linear regime, which was normalized to the value in the unbent state $\mu_0$, as a function of the strain and bending radius, respectively. Only small changes in $\mu/\mu_0$ were observed in this range of strains up to ∼2.2%, corresponding to a bending radius of 4.25 mm. A strain >2.2% caused failure of the gate dielectric layer made with epoxy and $SiO_2$, which can be detected by large gate leakage currents. These results suggest that the SWNTs and graphene TTFTs may have robust bending properties, compared to the oxide semiconductors.

## 4. Conclusion

Transparent, all carbon based thin film transistors consisting of SWNTs semiconductors and graphene electrodes were fabricated. These devices exhibited good electrical, optical and mechanical properties. In particular, the graphene electrodes showed excellent contact properties with the SWNT channel materials. Overall, the characteristics of all carbon based devices observed in this study are very promising for future flexible electronic applications.

## Acknowledgments

This work was supported by the National Research Foundation of Korea funded by the Ministry of Education, Science and Technology (2010-0015035, 2009K000179 and 2009-0081966) and a Grant-in-Aid for Industrial Source Technology Development Programs from the Korea Ministry of Knowledge Economy (No. 10033309).

## References

[1] Cao Q and Rogers J A 2009 *Adv. Mater.* **21** 29
[2] Forrest S R 2004 *Nature* **428** 911
[3] Wager J F 2003 *Science* **300** 1245
[4] Nomura K, Ohta H, Ueda K, Kamiya T, Hirano M and Hosono H 2003 *Science* **300** 1269
[5] Nomura K, Ohta H, Takagi A, Kamiya T, Hirano M and Hosono H 2004 *Nature* **432** 488
[6] Horowitz G 1998 *Adv. Mater.* **10** 365
[7] Dimitrakopoulos C D and Malenfant P R L 2002 *Adv. Mater.* **14** 99
[8] Gelinck G H *et al* 2004 *Nat. Mater.* **3** 106
[9] Cao Q, Hur S-H, Zhu Z-T, Sun Y, Wang C, Meitl M A, Shim M and Rogers J A 2006 *Adv. Mater.* **18** 304
[10] Cao Q, Zhu Z, Lemaitre M, Xia M, Shim M and Rogers J A 2006 *Appl. Phys. Lett.* **88** 113511
[11] Cao Q, Kim H, Pimparkar N, Kulkarni J P, Wang C, Shim M, Roy K, Alam M A and Rogers J A 2008 *Nature* **454** 495
[12] Snow E S, Perkins F K, Houser E J, Badescu S C and Reinecke T L 2005 *Science* **307** 1942
[13] Yakobson B and Avouris P 2001 *Top. Appl. Phys.* **80** 287
[14] Lee C, Wei X, Kysar J W and Hone J 2008 *Science* **321** 385
[15] Zhang Y, Tan Y W, Stormer H L and Kim P 2005 *Nature* **438** 201
[16] Odom T W, Huang J L, Kim P and Lieber C M 1998 *Nature* **391** 62

[17] Kim K S, Zhao Y, Jang H, Lee S Y, Kim J M, Ahn J H, Kim P, Choi J Y and Hong B H 2009 *Nature* **457** 706
[18] Lee Y, Bae S, Jang H, Jang S, Zhu S E, Sim S H, Song Y I, Hong B H and Ahn J H 2010 *Nano Lett.* **10** 490
[19] Bae S *et al* 2010 *Nat. Nanotechnol.* **5** 574
[20] Reina A, Jia X, Ho J, Nezich D, Son H, Bulovic V, Dresselhaus M S and Kong J 2009 *Nano Lett.* **9** 30
[21] Hur S H, Khang D Y, Kocabas C and Rogers J A 2004 *Appl. Phys. Lett.* **85** 5730
[22] Hur S H, Park O O and Rogers J A 2005 *Appl. Phys. Lett.* **86** 243502
[23] Nair R R, Blake P, Grigorenko A N, Novoselov K S, Booth T J, Stauber T, Peres N M R and Geim A K 2008 *Science* **320** 1308
[24] Dehuff N L, Kettenring E S, Hong D, Chiang H Q, Wager J F, Hoffman R L, Park C H and Keszler D A 2005 *J. Appl. Phys.* **97** 064505
[25] Artukovic E, Kaempgen M, Hecht D S, Roth S and Gruner G 2005 *Nano Lett.* **5** 757
[26] Snow E S, Novak J P, Campbell P M and Park D 2003 *Appl. Phys. Lett.* **82** 2145
[27] Kumar S, Pimparkar N, Murthy J Y and Alam M A 2006 *Appl. Phys. Lett.* **88** 123505
[28] Luan S and Neudeck G W 1992 *J. Appl. Phys.* **72** 766
[29] Hu L, Hecht D S and Gruner G 2004 *Nano Lett.* **4** 2513
[30] Kumar S, Murthy J Y and Alam M A 2005 *Phys. Rev. Lett.* **95** 066802