All ink-jet-printed carbon nanotube thin-film transistor on a polyimide substrate with an ultrahigh operating frequency of over 5 GHz

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We report a flexible carbon nanotube (CNT) thin-film transistor (TFT) fabricated solely by ink-jet printing technology. The TFT is top gate configured, consisting of source and drain electrodes, a carrier transport layer based on an ultrapure, high-density (>1000 CNTs/μm^2) CNT thin film, an ion-gel gate dielectric layer, and a poly(3,4-ethylenedioxythiophene) top gate electrode. All the TFT elements are ink-jet printed at room temperature on a polyimide substrate without involving any photolithography patterning or surface pretreatment steps. This CNT-TFT exhibits a high operating frequency of over 5 GHz and an on-off ratio of over 100. Such an all-ink-jet-printed process eliminates the need for lithography, vacuum processing, and metallization procedures and thus provides a promising technology for low-cost, high-throughput fabrication of large-area high-speed flexible electronic circuits on virtually any desired flexible substrate. [DOI: 10.1063/1.3043682]

Printing thin-film transistors (TFTs) on flexible substrates at room temperature offers a cost-effective way to achieve mass production of large-area electronic circuits without using special lithography equipment. It is expected to provide an enabling technology for many emerging applications such as flexible displays, radio frequency identification (RFID) tags, electronic papers, and smart skins, just to name a few. Printed flexible electronics have been reported by using various organic semiconducting polymers.[1–3] However, the carrier mobility of organic semiconducting polymers is still less than 1.5 $cm^2/V\,s$,[1–3] which limits the device operation speed to only a few kilohertz. Carbon nanotube (CNT), a material with exceptional aspect ratio and great mechanical flexibility, has shown great promises as an active carrier transport material in making high-speed flexible field-effect transistors (FETs).[4–12] Extraordinary field-effect mobility as high as 79 000 $cm^2/V\,s$ was reported in the FETs based on individual CNTs.[5] Due to the ultrahigh field-effect mobility, CNT-based flexible FETs are capable of achieving high-speed (gigahertz) operation.[13–15] However, most of the reported FETs were based on CNTs grown using chemical vapor deposition (CVD),[16,17] which generally requires an extremely high temperature, typically >900 °C.[5,16,17] This represents a major obstacle to fabricating electronic devices on flexible substrates because most flexible substrates are unable to survive such a high CVD growth temperature. FETs based on solution-processable CNT thin films[6–12] can be fabricated at room temperature and are thus especially suitable for printed electronics on flexible substrates. However, the sidewalls of as-produced nanotubes are covered by amorphous carbon (α-C), which is a very common carbonaceous impurity.[18] Such impurities would tremendously restrict the transport of carriers in the formed CNT thin films and seriously limit the field-effect mobility of the CNT-TFTs.[18,19] High field-effect mobility CNT-TFTs can be achieved by using ultrapure electronics-grade CNT solutions.[18] High-speed (>300 MHz) CNT-FETs have been demonstrated by syringe dispensing a tiny droplet of an electronics-grade CNT solution on a flexible substrate.[20] In this paper, we report an all-printed CNT-TFT on a polyimide substrate. All the elements of the TFT are fabricated solely by using ink-jet printing technology without involving any photolithography fabrication steps. An ultrahigh operating frequency of over 5 GHz was demonstrated with an on-off ratio of over 100.

The schematic structure of the CNT-TFT is shown in Fig. 1(a). Figures 1(b) and 1(c) show the pictures of the CNT-TFT. The TFT is in a top gated configuration. It consists of source (S) and drain (D) electrodes, a carrier transport layer based on an ultrapure, high-density (>1000 CNTs/μm^2) CNT thin film, a gate dielectric layer, and top gate electrode (G). All of these TFT elements were printed on a DuPont™ Kapton® FPC polyimide film[21] by using an Optomec's M^{3}D Aerosol Jet® printing system.[22] The S and D electrodes were first printed on the Kapton® FPC polyimide film using UTDAg silver nanoink from UT-Dots,[23] followed by the thermal annealing at 130 °C for 30 min. The width of the S and D electrodes was 50 μm, and the separation between the S and D electrodes, i.e., channel length (l), is 100 μm. An active carrier transport layer was then printed using an ultrapure, electronic grade CNT solution (CJ-28) from Brewer Science, Inc. In a separate experiment, it was verified that a high-density CNT thin film (>1000 CNTs/μm^2) with a low content of amorphous car-

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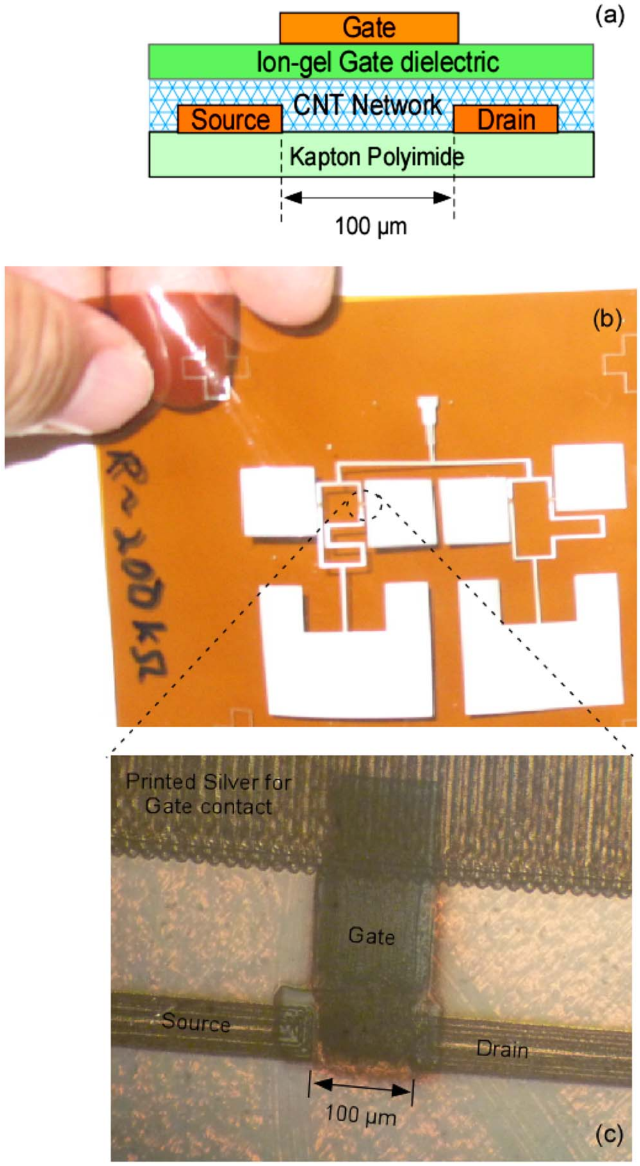

FIG. 1. (Color online) Printed CNT-TFT on a DuPont® Kapton® FPC polyimide film: (a) schematic structure (cross-section view), [(b) and (c)] picture of the CNT-TFT, (b) circuit, and (c) optical microphotography of the CNT-TFT (top view). The CNT-TFT is in a top-gated configuration. The channel width and length are 200 and 100 μm, respectively.

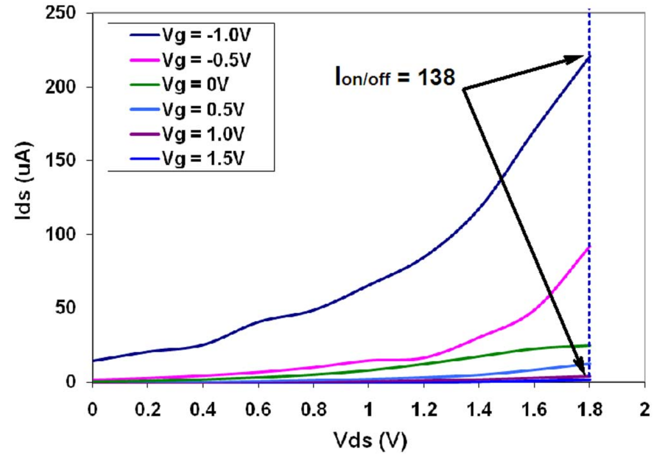

FIG. 2. (Color online) S-D *I-V* characteristics (I_{DS} vs V_{DS}) of the fabricated CNT-TFT at different gate voltages (V_G).

bon (α-C) can be obtained.[20] Multiple printing of the CNT solution between the S and D electrodes was performed to achieve a uniform CNT film with a S-D resistance of 200 kΩ. The CNT film was then air dried at room temperature. A thin layer of ion gel was then printed on top of the CNT film as the gate dielectric.[23] The TFT was finished by printing a conducting polymer poly(3,4-ethylenedioxythiophene) layer as the top gate electrode.[3] No postannealing or passivation step was performed after the printing of the TFT.

Figure 2 shows the S-D *I-V* characteristics (I_{DS} versus V_{DS}) of the CNT-TFT at different gate voltages (V_G). The gate voltages varied from −1.0 to +1.5 V. At the same S-D voltage (V_{DS}), lower S-D currents (I_{DS}) were observed as the gate voltage increased from negative to positive voltages. This indicates that the CNT network in this TFT is a *p*-type carrier (hole) transport layer.[19,20] At the S-D voltage (V_{DS}) of 1.8 V, a high S-D current (I_{DS}) of 221 μA and a low I_{DS} of 1.6 μA were obtained at the gate voltages of −1.0 and +1.5 V, respectively. The S-D current (I_{DS}) on-off ratio was 138. The large on-off ratio reveals a high content of the semiconducting type of CNTs in the CNT active layer. The gate-source leakage current (I_{GS}) was measured to be in the picoampere range. The low gate switching voltages of −1.0 and +1.5 V and the low leakage gate current indicate that the ion-gel layer is an effective gate dielectric.[23] Note that the *I-V* trace at zero gate bias is roughly linear with a S-D resistance of 72 kΩ. This is smaller than the initial 200 kΩ S-D resistance, indicating possible negative charges trapped in the ion-gel gate dielectric layer and functions effectively as a negative gate bias.

High-speed device operation was characterized by using a transimpedance amplifier,[14,20] as illustrated in Fig. 3(a), where R (120 Ω) and Z_L (50 Ω) are, respectively, the printed resistor and effective load of a microstrip transmission line with a characteristic impedance Z_0 designed at 50 Ω. Due to the CNT-TFT's high input impedance and low output impedance, this circuit configuration can effectively function as a transimpedance amplifier.[24] The cutoff frequency is predominantly determined by the transit time across the CNT channel length.[24] The small signal response of this voltage buffer stage is shown in Fig. 3(b). The pink trace is the 5 GHz input signal (V_{in}) biased at −1.0 V using a standard bias tee. The green trace is the corresponding output signal (V_{out}). The peak-to-peak values of the input and output signals were, respectively, 210 and 10 mV, indicating a transimpedance gain of <0.05. The low gain is due to the low resistance of the printed resistor. By optimizing the resistance of the printed resistor, a high transimpedance gain can be expected. Note that the output signal could follow the input signal without any waveform distortion, indicating excellent linearity. In the previous work,[20] we demonstrated a syringe-dispensed printable CNT-TFT with an operating speed of over 312 MHz by using external resistor and load. The operation speed of the circuit was not limited by the CNT-TFT but rather by the long wires from the probes to the external circuit board. By printing the integrated CNT-TFT with input and load transmission lines, a considerable operation speed improvement was achieved, indicating the high-speed property of CNT-based flexible transistors.

In this paper, we demonstrated an all-ink-jet-printed CNT-TFT on a DuPont® Kapton® FPC polyimide film by

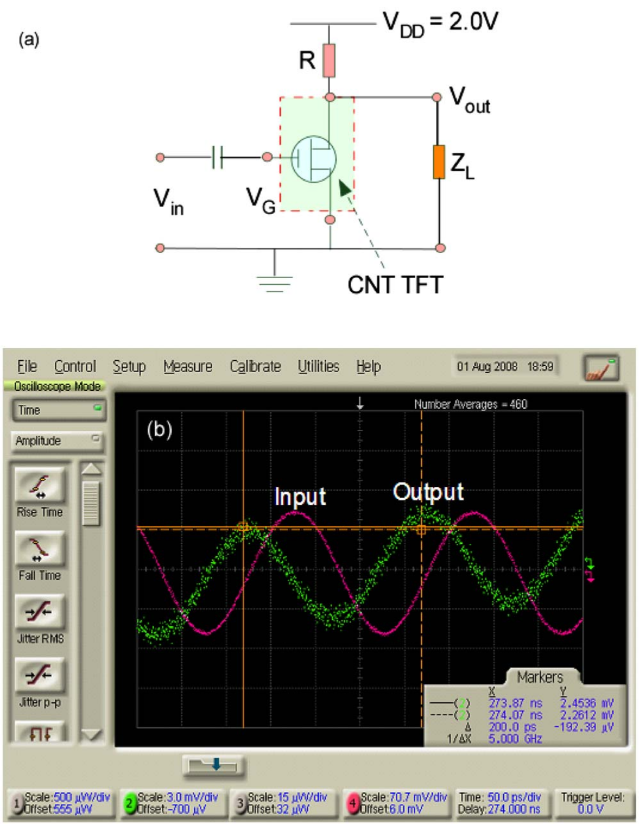

FIG. 3. (Color online) Demonstration of high-speed modulation of the fabricated CNT-TFT: (a) schematic circuit diagram and (b) small signal response of the voltage follower at 5 GHz.

using an Optomec's M^3D ® Aerosol Jet printing system. A high-speed (5 GHz) TFT with a large high on-off ratio of over 100 was obtained. The use of this ultrapure CNT solution enabled the ink-jet printing compatibility of CNTs with room-temperature fabrication of electronic devices on flexible substrates, while additional lithography patterning, etching, and vacuum metallization upon the formed CNT thin film were no longer needed. The preliminary printed CNT-TFT and the initial high-speed performance demonstration indicate the feasibility of the CNT-based all-printed flexible high-speed TFT technology. Such ink-jet printing of flexible TFTs at room temperature would enable mass fabrication of large-area electronic circuits on virtually any desired flexible substrate at low cost and high throughput for many emerging applications such as flexible displays, RFID tags, electronic papers, and smart skins.

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