

Self-Aligned Carbon Nanotube Thin-Film Transistors on Flexible Substrates With Novel Source–Drain Contact and Multilayer Metal Interconnection

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Abstract—This paper presents the development and characterization of self-aligned carbon nanotube thin-film transistors (CNT-TFT) on flexible substrates. The channel consisting of dense, aligned, 99% pure semiconducting single-walled CNT is deposited using the dip-coat technique on a sacrificial substrate and then transferred on the device substrate. The source, drain, and gate structures are formed by the ink-jet printing technique. A novel source–drain contact formation using wet droplet of silver ink prior to the CNT thin-film application has been developed to enhance source–drain contact with the CNT channel. Bending test data on CNT-TFT test structures show minimal change (less than 10%) in their performance. Moreover, a special multilayer metal interconnection technology is demonstrated for flexible electronics applications. Bending test data on via test structure show change in resistance by less than 5%.

Index Terms—Carbon nanotube (CNT), dip-coat technique, flexible electronics, single-walled carbon nanotube (SWCNT), thin-film transistor (TFT).

I. INTRODUCTION

INK-JET printing technique is a simple and cost-effective method to produce electronics. It is an attractive process for flexible electronics due to its noncontact and additive deposition technique. The technique does not require sacrificial resist or liftoff layers but rather deposits materials only where needed.

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Soluble organic materials have been used as the channel material for several state-of-the-art flexible electronics [1], [2]. However, the carrier mobility of organic semiconductor polymers is less than $1.5 \text{ cm}^2/(\text{V}\cdot\text{s})$ [3], [4]. This limits the organic polymer thin-film transistor's (TFT's) operating speed to only a few kilohertz. On the other hand, carbon nanotube (CNT)-based TFTs have seen tremendous improvements over the last five years due to their excellent mobility characteristics [5], [6]. Extremely high mobility of $100\,000 \text{ cm}^2/\text{V}\cdot\text{s}$ of individual CNTs has been reported [6]. CNT-TFT-based devices on flexible substrates have achieved high-field mobilities using ultrapure electronics-grade CNT solutions [7], [8] by ink-jet printing technique. CNT in solution can be printed; however, it often clogs up the nozzles. Other techniques such as dielectrophoresis [9], spin coat [10], and spray [11] to form CNT-TFT have been demonstrated. These techniques yield a random network of CNTs on the substrate. However, in order to retain the attractive properties of individual CNTs, it is desirable to have densely packed, perfectly aligned, horizontal arrays of CNT thin films as the channel of the TFT device. Most of the reported aligned CNT thin films are deposited on silicon or quartz substrates via the CVD technique [12]–[14]. This deposition technique is unsuitable for flexible substrates because of the high-deposition temperature.

In this study, we fabricate CNT-TFT using a combination of ink-jet printing and stamping (for CNT layer) techniques, with self-aligned-CNT channel, novel source–drain contact, and multilayer metal interconnect.

II. FABRICATION PROCESS

A. Ink-Jet Printing

Ink-jet printing is performed using the Dimatix DMP 2800 (Dimatix-Fujifilm, Inc., Santa Clara, CA) that uses piezoelectric printing cartridge (DMC-11610). The ink droplet dispensed from the ink cartridge has a nominal volume of 10 pL. One or more nozzles can be used for the printing. For small TFT structure used in this study, only one nozzle is used. For other large structures such as probing pads, via contact pads and other large elements, multiple nozzles up to 16 are used. The temperature of the platen and cartridge is set at room temperature. Silver ink from Cabot Corporation (CCI-300) is used for the source, drain, and gate electrode printing. Spin-on glass is used as dielectric ink. Customized waveform patterns in the printer are used to print silver and dielectric inks.

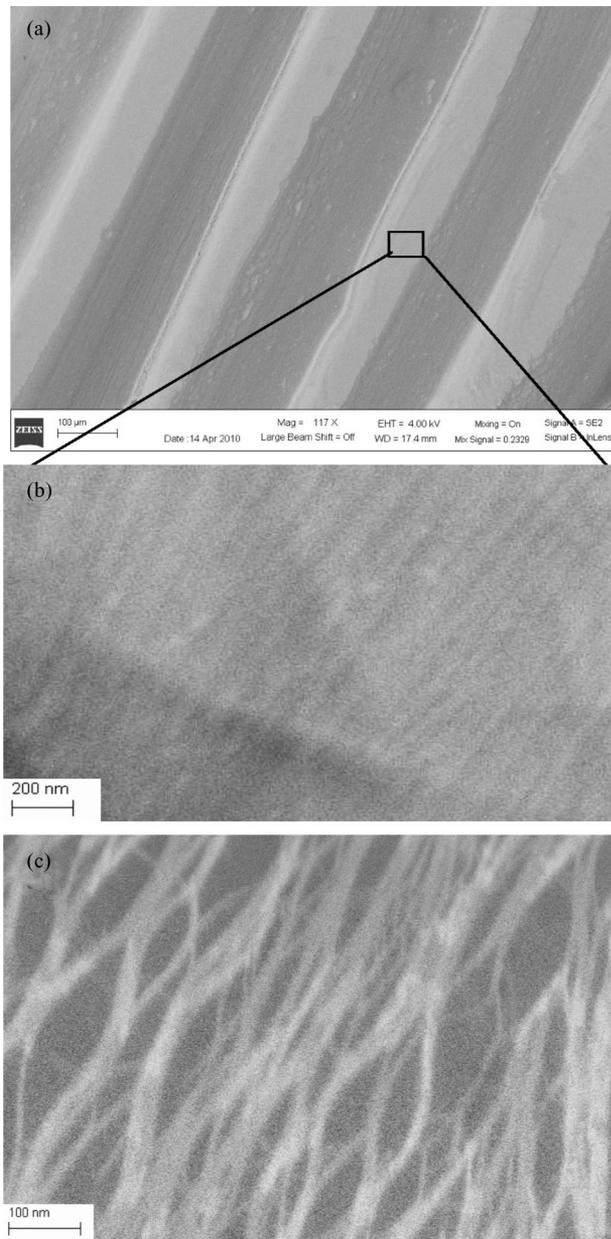
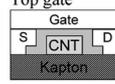
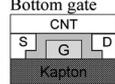


Fig. 1. (a) SEM image showing the stripe of CNT with surfactant. (b) Zoom-in SEM image of self-aligned CNT thin film with surfactant. (c) SEM image of a self-aligned CNT thin film after suspended in alcohol bath.

B. CNT Deposition

Using the dip-coat technique, CNTs can be self-aligned on the substrate [18]. The dip-coating technique has been widely used to deposit particles uniformly on different surfaces [15]–[17]. The main driving force for the convective transfer of CNTs is evaporation of the solvent. As the solvent starts to evaporate, the convection force transfers the CNTs to the contact line (solid–vapor–liquid interface), thus depositing CNTs on the substrate [15], [16]. Depending on the concentration, particle size, and drying speed, single or multiple layers can be obtained. We observe that stripes of the CNT thin film are obtained at high-drying speeds. This occurs when the capillary force, which pulls

TABLE I
COMPARISON BETWEEN TOP AND BOTTOM GATE INTEGRATIONS

Integration	Advantage	Disadvantage
 <p>Top gate</p>	<ul style="list-style-type: none"> ◆CNT channel deposited first ◆Dielectric & gate electrode cover the channel (passivating the channel) ◆CNT channel has good contact with the annealed source-drain metal printed on top of the channel 	<ul style="list-style-type: none"> ◆Dielectric material diffuses between CNTs and separate them ◆High thermal budget applied on the CNT channel.
 <p>Bottom gate</p>	<ul style="list-style-type: none"> ◆CNT channel deposited last – No interaction with dielectric and gate materials ◆Low thermal budget for CNT channel 	<ul style="list-style-type: none"> ◆CNT channel has bad contact with source-drain areas (solution identified in this paper) ◆No passivation – exposed to air (solution identified in the paper)

the liquid inward, builds up and exceeds the surface tension of the liquid. When this happens, a new contact line is formed as shown in Fig. 1. In this study, 99% pure semiconducting single-walled CNTs in surfactant solvent from Nanointegris, Inc., is used (S10-671, 0.1 mg in 10 mL aqueous solution) for the dip-coat technique to deposit CNTs on a silicon substrate (acting as a sacrificial substrate). The surfactants keep the CNTs from bundling with each other, which assists in the self-alignment process. Fig. 1(a) shows the SEM image of stripes of CNT, with the surfactant, formed using our process. Rinsing the substrate in 2-propanol causes the surfactants to dissolve away, thus leaving CNT on the substrate as shown in Fig. 1(b). Further suspending the substrate in 2-propanol for 15 min entirely removes the surfactants and exposes the CNTs as shown in Fig. 1(c). Note that some amount of CNT is also lost during this process. In this process, the stripe width can be controlled by temperature (evaporation speed), solvent properties, particle density, and substrate surface roughness.

C. Process Integration

Both top gate and bottom gate integration approaches are studied. Table I show the comparison of both integration schemes.

Due to its attractive advantages of low thermal budget and least interaction with dielectric and gate materials, the bottom gate integration approach is utilized in this study. In order to overcome the disadvantages, novel solutions have been addressed in this paper. The schematic of the bottom gate integration process flow is shown in Fig. 2. A Kapton polyimide substrate with a thickness of 127 μm is used. The gate electrode is first printed on the Kapton film using silver ink, followed by thermal annealing at 160 $^{\circ}\text{C}$ for 10 min. Spin-on glass is used as the dielectric material, which is also printed. The source and drain regions are printed using the same silver ink as used for gate electrode and under the same annealing conditions. The device channel length is 100 μm and width is 300 μm . For the small structure, only one nozzle is used during the printing in order to maintain resolution.

The 99% pure semiconducting CNT from Nanointegris, Inc., Skokie, IL, is deposited on the silicon substrate (as a sacrificial substrate) using the dip-coating technique, as discussed in

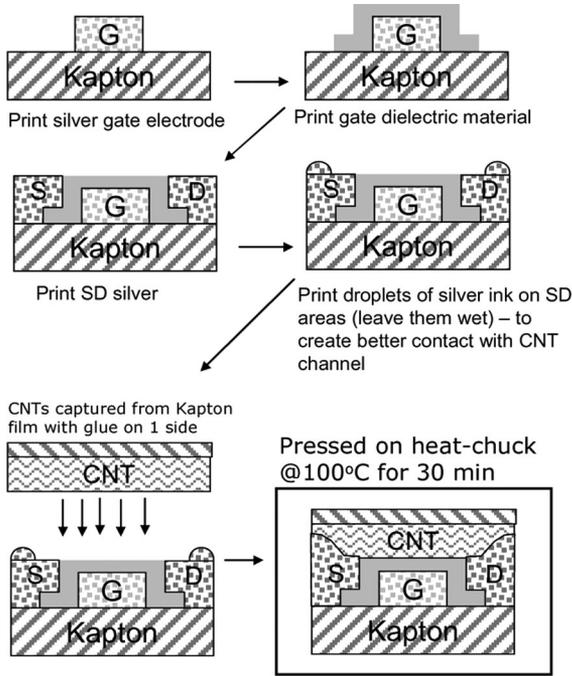


Fig. 2. Bottom gate integration with novel source-drain and CNT contact.

Section II-B. In order to transfer the CNT thin film onto the flexible Kapton polyimide substrate, another special Kapton substrate (25- μm thick), with adhesion coating on one side, is used to lift the self-aligned CNT thin film from the silicon substrate and lay it on top of the first substrate over the printed channel region. The Kapton with adhesive is left on the device in order to protect the CNT channel, thus, acting like a passivation layer.

Prior to bonding these two layers, a novel technique to enhance the contact between CNT and source-drain areas is developed. In this technique, droplets of silver ink are printed on the source and drain areas on the first substrate as shown in Fig. 3. These wet-silver ink droplets allow the silver liquid to “wet” the CNT thin-film area and enable good contact with the printed source and drain contact. In order to bond the two substrates, the device is left under pressure on a heat chuck surface at 100 °C for 30 min to enhance the bonding of the second Kapton substrate to the first substrate and eliminate any air pockets. Upon bonding and annealing, the source-drain contact junction is formed into the CNT thin film, thus, providing a good contact with the entire thin film. Without using the wet-silver droplets to enhance the contact, the ON current is very low ranging from a few nanoamperes to a few microamperes.

III. CHARACTERIZATION OF THE SELF-ALIGNED CNT-TFT

Fig. 4(a) and (b) shows the measured I - V characteristics (I_D versus V_{DS}) of the self-aligned CNT-TFT as a function of different gate voltages V_G . The transistor I - V characteristics are measured using precision semiconductor parameter analyzer (Agilent 4156C). At a gate voltage of 0 V gate, the device does not show pinchoff, most likely due to a small number of metallic nanotubes in the channel. At $V_G = -3$ V and

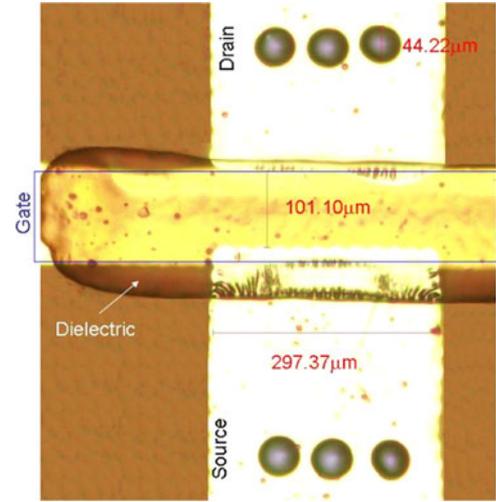


Fig. 3. Wet silver droplets on source-drain areas before bonding with a CNT thin film.

source-drain voltage V_{DS} of -1 V, high-drive current of 0.371 mA is obtained, which is in good agreement with the high density and self-aligned nature of CNTs on the device channel. As mentioned by Engel *et al.* [18], CNT-TFT can have the I_{on}/I_{off} ratio as high as 10^5 . At this channel length, percolation alone can give you much higher I_{on}/I_{off} ratios; let alone using material that is highly purified. However, any improvement for the on-state current will degrade the I_{on}/I_{off} ratio due to the increased number of metallic pathways between source and drain. In this experiment, we use a channel length of 100 μm , which is 20 times smaller than that reported in [18]. Due to the longer-channel length, a much smaller I_{on}/I_{off} ratio can be expected from the percolation model. We experimentally observe the I_{on}/I_{off} ratio of around 20. The restriction on the channel length comes inherently from the printing process. Further optimizations such as using the electrical burning-off technique to eliminate metallic pathways will be reported in a future publication.

Bending test is performed on the device. Fig. 5 shows the picture of bending test structures. Two different test structures are formed to evaluate the vertical and horizontal orientations of the transistor. Three different radii of curvature, 4.5, 3, and 1.5 mm, are used in this evaluation. Forward and backward bending tests are conducted as described in Fig. 5.

Fig. 6 shows the normalized drain current ($I_{D,test}/I_{D,original}$) plotted against the radius of curvature for the vertical test structure. In forward bending, lower drain current is observed, with up to 10% change, while no significant drain current difference is observed in the backward bending case. Larger current change for the vertical test structure can be attributed to the 300- μm channel width of the testing device subjected to the bending direction. Since the CNTs are captured by the thinner Kapton film that is bonded onto the thick Kapton substrate, forward bending may cause the thin Kapton film to move away from the substrate, while in backward bending, it causes the thin Kapton film to press against the substrate.

Fig. 7 shows drain current bending test data for the horizontal test structure. Smaller change in drain current is observed

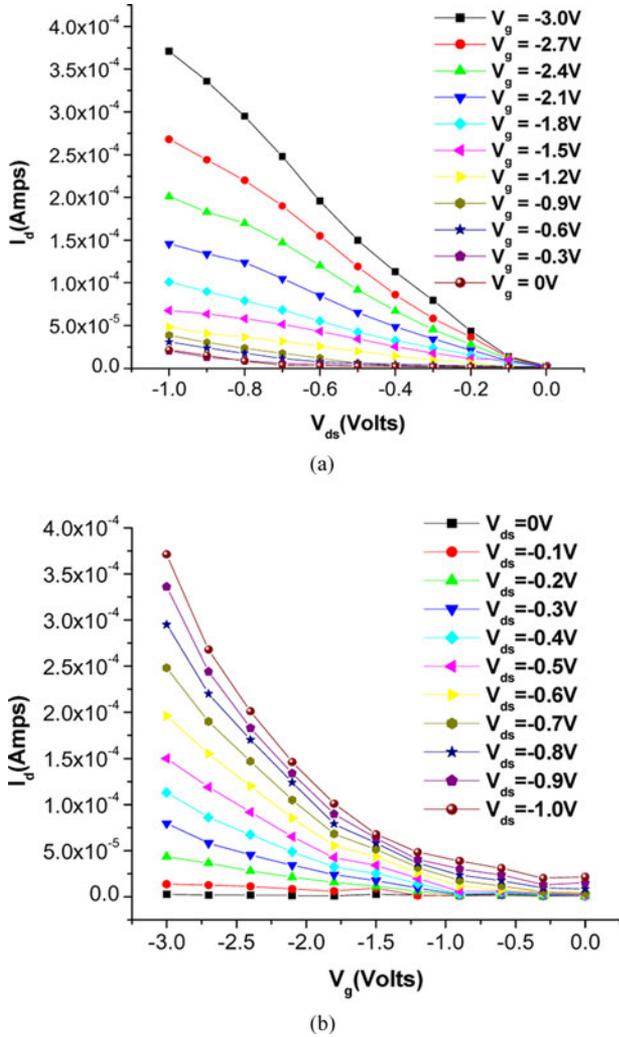


Fig. 4. (a) I - V characteristics (I_D versus V_{DS}) of the self-aligned CNT-TFT at different gate voltages. (b) I_D versus V_G of the self-aligned CNT-TFT.

(less than 6%). For the horizontal test structure, 100- μm channel length of the device is subjected to the bending direction. In the forward bending case, shorter channel length causes a small increase in current, and in the backward bending case, “stretched” channel length reduces the current (even though the thin top layer Kapton is pressed against the substrate).

IV. MULTILAYER METAL INTERCONNECT

In order to develop full systems such as large area printed phased array antennas utilizing CNT-TFTs as RF amplifiers, the signal layout scheme will be extremely complicated and prohibitive if performed in 1-D (i.e., all interconnection lines, other components, and CNT-TFTs printed in the same layer). Similar to silicon semiconductor devices, such complex-flexible electronics also require multiple layers and interconnections for its circuits. Especially, CNT-TFTs will all be formed in one layer and the signals will be routed to and from these FET to all other components through various stacked layers. Therefore, considering such a requirement for futuristic applications in mind, in this paper, we also report a technique to form multiple-layer

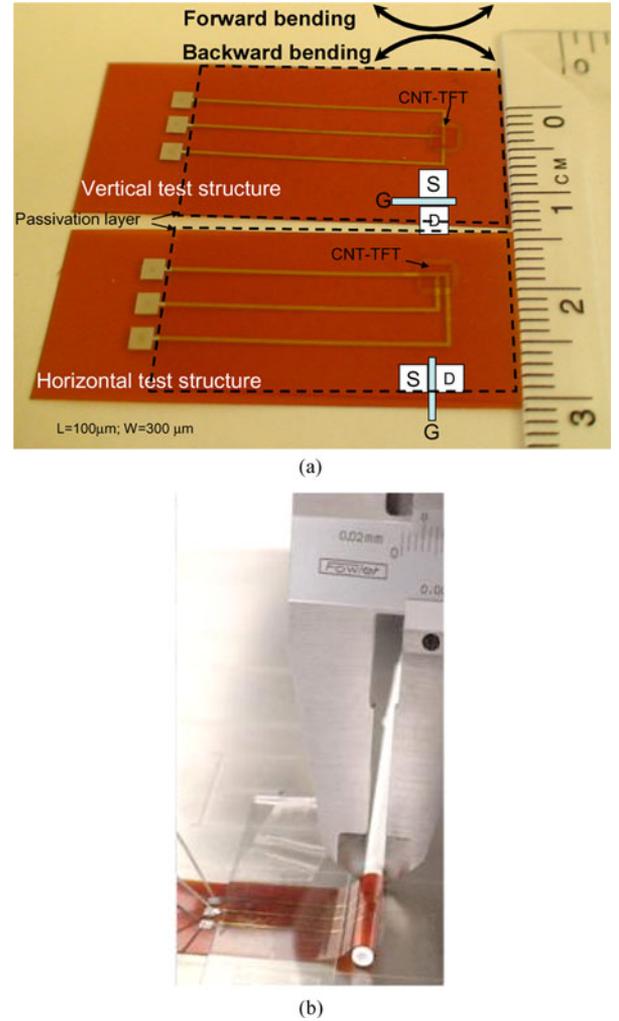


Fig. 5. (a) Bending test structure for vertical and horizontal devices. (b) Device under backward bending test at 1.5 mm radius of curvature.

metal interconnection on flexible substrates and show multi-layer interconnection to the fabricated CNT-TFT device. After forming the CNT-TFT device layer, a special Kapton substrate (25- μm thick), with adhesive coated on one side, is used to glue on top of the first substrate. Contact vias are formed prior to attaching, in order to obtain metal contacts with the gate contact pads on the bottom substrate. The via diameter used in this study is 1 mm (available cylinder blade to cut the via of that size). The adhesion of this layer is critical since any voids between layers will cause the liquid silver ink to spread between these layers due to the capillary effect. An annealing process is used after bonding these layers together by pressing against a heat chuck at 100 $^{\circ}\text{C}$ for 30 min. Then, the silver ink is printed on top of the top Kapton layer for the metal interconnection lines on the top substrate. The printed silver ink penetrates through the vias to contact the gates on the first layer. Following this, another annealing process is performed in order to evaporate the solvent in the silver ink and form a good Ohmic contact. Fig. 8 shows the schematic of multilayer metal interconnect integration for flexible electronics performed in this study.

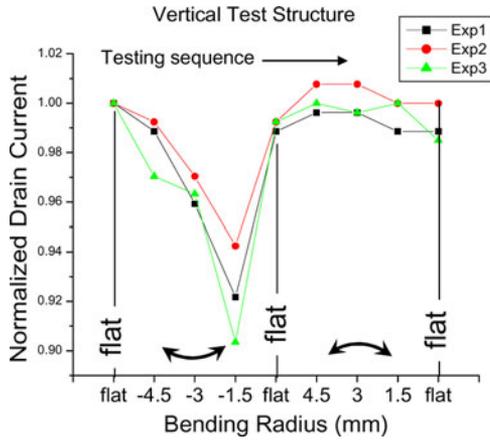


Fig. 6. Bending test data for the vertical test structure.

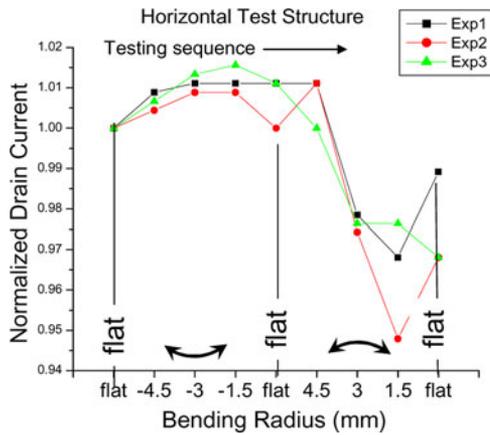


Fig. 7. Bending test data for the horizontal test structure.

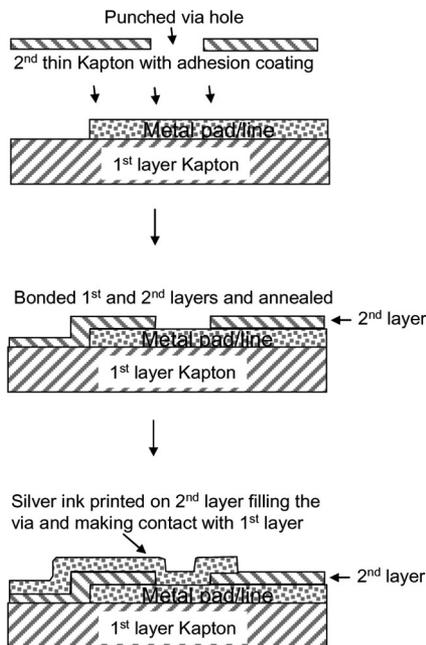


Fig. 8. Multilayer metal interconnect integration scheme.

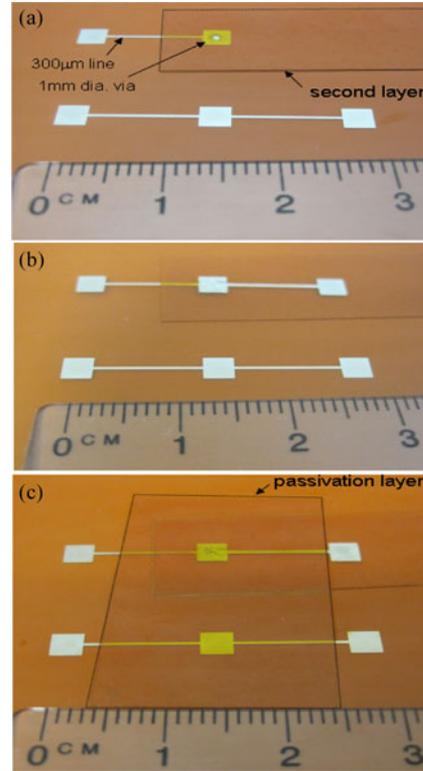


Fig. 9. (a) Via development test structure before forming via and second layer. (b) Via formed and contact line was printed on the second layer. (c) Protected or passivated layer to protect the circuit.

In order to evaluate the type of contact formed, we form a test structure as shown in Fig. 9. From the resistance measurements, we observe no resistance difference between the two contact pads with and without the interconnect via. Fig. 9(a) and (b) shows via development test structure, pre- and post silver ink printing on the second Kapton layer. To protect the metal contact line and via, the Kapton substrate with adhesive coating is bonded on top [see Fig. 9(c)]. This layer acts as a passivation or protection layer for the circuit and protects it from environment as well as mechanical strength.

Fig. 10 shows the bending test for the via test structure. Lower resistance value is observed when bending to small radius of curvature. It indicates that the via is not completely filled with silver ink. Silver thickness on flat surface is measured to be around $0.4 \mu\text{m}$. The via diameter is 1 mm , and the metal interconnect line is $300 \mu\text{m}$ wide. In forward bending, the via might squeeze the silver particles together causing lower resistance data, while in backward bending, the top passivation Kapton layer applies pressure to the silver particles in the via.

V. SUMMARY

In this paper, we demonstrate a self-aligned CNT-TFT using 99% pure semiconducting nanotube on flexible Kapton substrate. A novel source-drain contact is developed to enhance the contact with the CNT channel. Bending test data show minimal changes (less than 10%) in their performance.

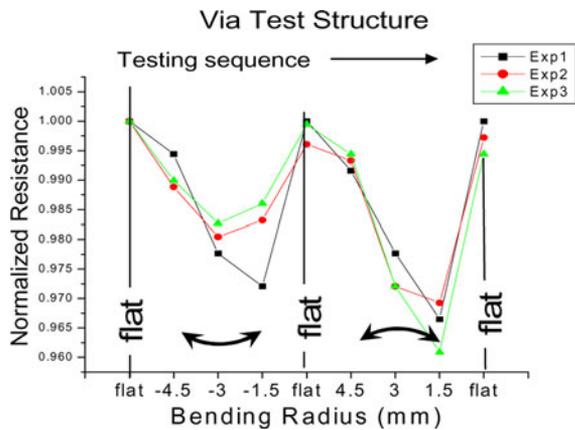


Fig. 10. Bending test data for via test structure.

Multiple-layer interconnect integration is also demonstrated on a flexible-substrate circuit. These developments lift strong constraints for flexible electronics, providing an open route for realistic applications using self-aligned CNT-TFT and multiple-layer interconnection for flexible-electronics technology.

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