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# Ultraviolet/ozone and oxygen plasma treatments for improving the contact of carbon nanotube thin film transistors

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#### ABSTRACT

Carbon nanotube thin film transistor (CNT-TFT) is an emerging technology for future macroelectronics, such as chemical and biological sensors, optical detectors, and the backplane driving circuits for flat panel displays. The mostly reported fabrication method of CNT-TFT is a lift-off based photolithography process. In such fabrication process, photoresist (PR) residue contaminates the interface of tube-metal contact and deteriorates the device performance. In this paper, ultraviolet ozone (UVO) and oxygen plasma treatments were employed to remove the PR contamination. Through our well-designed experiments, the UVO treatment is confirmed an effective way of cleaning contamination at the tube-metal interface, while oxygen plasma treatment is too reactive and hard to control, which is not appropriate for CNT-TFTs. It is determined that 2–6 min UVO treatment is the preferred window, and the best optimized treatment time is 4 min, which leads to 15% enhancement of device performance.

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#### 1. Introduction

As an emerging thin film transistor (TFT) technology, carbon nanotube thin film transistors (CNT-TFTs) have shown high mobility [1–3], high on-current ( $I_{on}$ ) with high on/off current ratio [3–5]. Therefore, CNT-TFTs are regarded an promising TFT technology for future macroelectronics, such as chemical and biological sensors, optical detectors, and the backplane driving circuits for flat panel displays (FPDs) [1,6]. Though the performance of CNT-TFTs is superior in some aspects as compared with those currently commercialized TFT technologies, its overall performance and fabrication processes are still far from the best optimization.

The metal-tube contact has been confirmed critical for the performance of CNT-TFTs [2,7,8], which should be well controlled during the device fabrication. However, the reported contact resistances of CNT-TFTs are usually very high [2]. Usually metal was deposited on top of the CNT to form side-bond contacts. The unique perfect one dimension structure without dangling bonds of CNT leads to weak metal-tube coupling. Thus the electron injection at the metal-tube interface was limited which results in relatively high contact resistance ( $R_c$ ) [9,10]. However, the device fabrication process also affects the  $R_c$  of the final devices seriously. CNT-TFTs are usually fabricated by a lift-off based photolithogra-

phy process [11-13]. In such process, photoresist (PR) is coated onto the CNT film with following exposure and developing steps. Thus the source/drain electrodes are defined by the opening windows in the PR layer. Then metal were deposited and followed by lift-off using solvents, e.g., acetone or N-methyl pyrrolidone (NMP). PR residue usually exists in the window area which is sandwiched between the metal electrodes and the CNT film (Fig. 1e), and leads to high  $R_c$  in nano devices. Actually the PR residue problem exists in fabrication of many kinds of nano devices through a lift-off process, such as graphene field effect transistors (FETs) [14,15], MoS<sub>2</sub> FETs and so on [16]. This problem is not a concern in the fabrication of conventional TFTs based on amorphous silicon (a-Si), low temperature poly silicon (LTPS) and amorphous oxide semiconductors (e.g., InGaZnO, IGZO) [17]. In commercial TFTs fabrication, an etching based photolithography process is usually used, so that the affection of PR residue on the contact is avoided [18]. Unfortunately, such etching based photolighography process for fabrication of CNT-TFTs has not been well developed [13]. PR residue can also be cleaned by oxygen plasma in conventional device fabrication, where the device channels are bulk semiconducting materials. However, in contrast to conventional bulk semiconducting materials, oxygen plasma cause serious damage to nano materials and hence deteriorates performance of the fabricated devices. Therefore, a generous method for cleaning the PR residue in nano device fabrication should be developed.

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Fig. 1. (Color online) (a)-(h) Process flow of the fabrication of CNT-TFTs with integrated UVO treatment step for improving the contact.

We have developed an ultraviolet ozone (UVO) treatment method for cleaning the PR residue at the metal/graphene interface, which can improve the metal-graphene contact effectively [14,15]. UVO is a common cleaning process used in semiconductor device research and manufacturing, and in applications requiring critically clean interfaces such as those involving the assembly of molecules on metal or oxide surfaces for which aggressive plasma and ion bombardment processes cannot be tolerated [19]. During the UVO treatment process, the atomic oxygen is produced and the organic contaminant molecules were excited by the UV illumination. Then the atomic oxygen reacts with the contaminant molecules to form simpler volatile molecules, such as CO<sub>2</sub>, H<sub>2</sub>O, N<sub>2</sub> etc. This surface cleaning process is very gentle, which introduces much less structural damages to the surface than oxygen plasma. It is straightforward to think of integrating this method to the fabrication process of CNT-TFTs. Though CNT is often regarded as seamless roll up of graphene, it is known that CNTs are more reactive than graphene due to the curvature effect [20]. Whether the CNTs can survive in the UVO treatment is not known. Therefore, it is necessary to investigate whether the UVO treatment is applicable for CNT-TFTs.

In this paper, UVO was employed to clean the PR contamination at the contact interface of CNT-TFT. The cleaning process was monitored by atomic force microscope (AFM), which revealed that PR residue can be effectively cleaned. Electrical measurements confirmed that the contact was improved. However, due to its relative higher reactivity, the time window of UVO treatment for CNT-TFTs is narrower than that of graphene devices. As a comparison, oxygen plasma treatment was also studied. The results indicate oxygen plasma is too aggressive, which is hardly applicable for CNT-TFTs.

#### 2. Experimental

High purity semiconducting (>99.9%) single-walled carbon nanotube (SWCNT) solution was purchased from Beijing Huatan Co. Ltd. The diameters of the CNTs are about 1.4–1.6 nm and the

tube lengths are about  $1.5-2 \mu m$ . Conventional solution deposition method, just soak the SiO<sub>2</sub>/Si substrate in the solution, was used to fabricate CNT network films [12]. By controlling the soaking time and the solution concentration, uniform CNT film with desired tube density was obtained. The fabrication process flow of CNT-TFTs is shown in Fig. 1. CNT film was firstly patterned to the channel film of TFTs by photolithography and oxygen plasma etching, as shown in Fig. 1d. Then the contact area was defined in a second photolithography step. After opening the windows for metal contacts in the PR layer, the substrate was placed into a commercial UVO cleaner or inductively coupled plasma (ICP) etcher (Section 1 in Supporting Information) for removing the PR residue prior to metallization. Then Ti/Pd (0.3/40 nm) was evaporated and followed by lift-off. In the device fabrication process, S1813 (Microposit S1813 G2) was used for PR in the first photolithography step. While in the second photolithography step, double layer PR, LOR 3A (from MicroChem) below S1813, was employed for easy lift-off. Electrical properties of CNT-TFTs were characterized by Keithley 4200 SCS. The morphology of CNT film and PR residue were monitored by AFM.

#### 3. Results and discussion

We first evaluated the PR residue on the CNT surface and the effectiveness of UVO cleaning of the residue by AFM. Usually high density CNT films are preferred for TFT fabrication because higher tube density leads to higher mobility and current. However, too high tube density results in high surface roughness, which hinders observing CNTs and substrate below simultaneously. Since the PR residue layer is very thin, a smooth substrate surface can be used as a reference which is great helpful for evaluating the effectiveness of UVO cleaning. Therefore, relative low density CNT film was used for this purpose and the same area was monitored by AFM for comparison during each step of PR cleaning by UVO. Fig. 2a shows the typical AFM image of the as-deposited CNT

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