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Uniform percolation of inkjet-printed polymer-semiconductor -wrapped carbon nanotube networks by blending with insulating polymer

Jiyoul Lee

Department of Graphic Arts Information Engineering, College of Engineering, Pukyong National University, Sinseonro 365, Nam-gu, Busan, 608-739, Republic of Korea

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ABSTRACT

The enhanced performance uniformity of thin-film transistors (TFTs) fabricated by inkjet printing of polymer-semiconductor-wrapped single-walled carbon nanotube (PSC/SWCNT) networks blended with an insulating polymer binder was investigated. The inkjet-printed PSC/SWCNT network TFTs exhibited varied device performance with a field-effect mobility of $6.15 \pm 6.75\ cm^2/V{\cdot}s$; the inkjet-printed TFTs fabricated using the poly(methyl methacrylate) (PMMA)-blended PSC/SWCNT layer as a channel exhibited relatively uniform device performance, with a field-effect mobility of $7.57 \pm 3.32\ cm^2/V{\cdot}s$. This notable difference in device performance uniformity is attributed to the blended PMMA, which prevented the PSC/SWCNTs from forming random aggregates and facilitated their uniform percolation when they were inkjet-printed.

1. Introduction

Single-walled carbon nanotubes (SWCNTs) are attractive nanomaterials in the development of flexible electronic devices such as optoelectronic devices, chemical and biological sensors, thin-film transistors (TFTs), logic circuits, and computers because of their extraordinary charge carrier mobility, outstanding optical properties in the infrared region, solution processability, and high mechanical flexibility [1–4]. In general, carbon nanotubes are known to form bundles, which alters their intrinsic one-dimensional properties and tunable optoelectronic characteristics. Thus, many researchers have focused on isolating individual carbon nanotubes using various techniques. Well-known methods for isolating semiconducting SWCNTs (sc-SWCNTs) involve the sorting of SWCNTs in solution, the removal of metallic SWCNTs (m-SWCNTs) after growth, interactions via surface functional groups, and chemical reactions with SWCNTs. In particular, the solution-based sorting of sc-SWCNTs via non-covalent functionalization has been demonstrated to be the most promising method for selecting pure sc-SWCNTs without degrading their electrical properties. Among the solution-based sorting methods, which include density gradient ultracentrifugation, column chromatography, partition separation, deoxyribonucleic acid wrapping, and conjugated polymer wrapping, the conjugated polymer wrapping method is regarded as a facile route toward the isolation of carbon nanotubes for the large-scale sorting of SWCNTs [5,6]. However, although the resulting conjugated-polymer-semiconductor-wrapped SWCNTs (PSC/SWCNTs) are well-sorted, when the inkjet-printing technique is used to maximize the merits of solution processability of the SWCNTs, the inherently poor jetability of SWCNTs results in randomly percolated PSC/SWCNT networks, which, in turn, leads to variable device performance in the PSC/SWCNT network-based TFTs.

Here, we applied blending systems involving insulating polymer binders to solve the problem of non-uniform performance of PSC/SWCNT TFTs. By introducing the insulating polymers into the PSC/SWCNT network, we improved the uniformity of the performance of inkjet-printed PSC/SWCNT network TFT devices. Our approach enables stable jetability of inks containing PSC/SWCNTs, thereby leading to uniformly percolated PSC/SWCNT network films. The PSC/SWCNT networks blended with the polymer binder resulted in high-performance PSC/SWCNT network-based TFTs with a relatively uniform field-effect mobility of $7.57 \pm 3.32\ cm^2/V{\cdot}s$.

E-mail address: jiyoul_lee@pknu.ac.kr (J. Lee).

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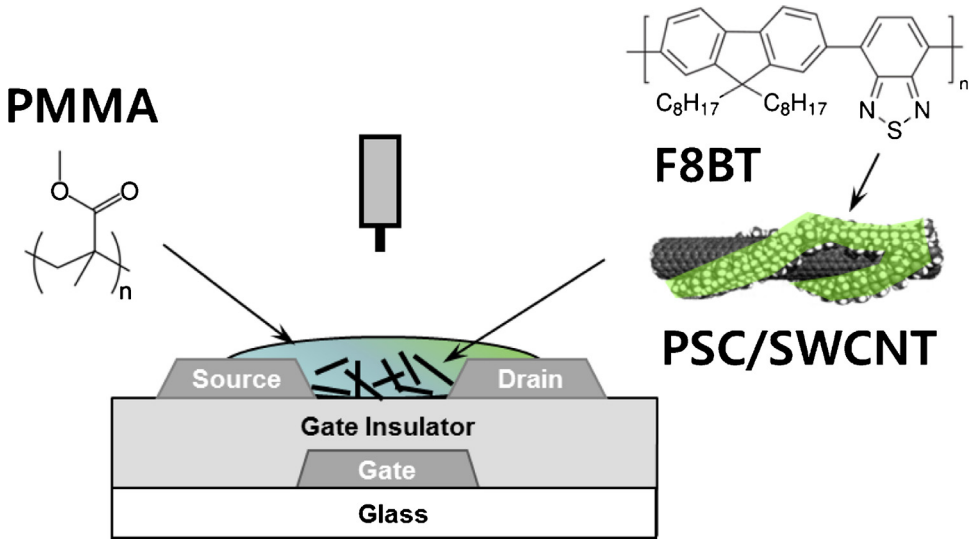

Fig. 1. Schematic of transistors and the PSC/SWCNT networks; the chemical structures of F8BT and PMMA used in this study are also shown.

2. Experimental procedure

Fig. 1 shows a schematic of transistors and PSC/SWCNT networks, where poly(9,9-dioctylfluorene-*alt*-benzothiadiazole) (F8BT) was used as a wrapping conjugated polymer [7] and poly (methyl methacrylate) (PMMA) was used as an insulating polymer binder material. The PSC/SWCNT-based TFTs were fabricated in a coplanar (bottom-gate bottom-contact, BGBC) structure, as shown in Fig. 1. For a gate electrode, a 50 nm-thick aluminum layer was thermally evaporated onto the glass substrate and photolithographically patterned. An organic–inorganic hybrid dielectric material composed of siloxane- and zirconium-based compounds ($\varepsilon_r = 3.2$) was spun at 3000 rpm for 1 min to form a 1.2 μm-thick layer, which was subsequently cured at 200 °C for 2 h. A 50 nm-thick silver layer was then thermally evaporated and photolithographically patterned to form the source/drain contacts with a width and length of 200 μm and 20 μm, respectively.

Commercially available SWCNTs prepared via a high-pressure carbon monoxide (HiPCO) process (Meijo Nanocarbon) were used for separation and functionalization experiments. The SWCNTs were first dispersed in an H_2O solution with a 2% (w/v) sodium cholate surfactant and sonicated; they were subsequently subjected to ultracentrifugation to remove SWCNT bundles and other impurities. The resulting solution contained individually suspended SWCNTs at a final concentration of 0.005% (w/v). In order to obtain an electronically homogeneous sc-SWCNT species, m-SWCNTs were removed from the pristine SWCNTs using a single cascade of the density-induced separation method, as described in the literature [8,9]. Fig. 2(a) shows the radial breathing mode (RBM) results for the Raman spectra of the separated sc-SWCNTs at an excitation energy of 2.41 eV; these results indicate that the m-SWCNTs were removed after separation. Next, a typical dispersion experiment was conducted to prepare the PSC/SWCNTs. As-separated sc-SWNTs (0.1 mg) and F8BT (10 mg) were suspended in 10 mL of *ortho*-dichlorobenzene (DCB, purchased from Sigma-Aldrich). The mixture was sonicated in a bath-type sonicator for 10 h to yield a DCB solution of the F8BT-wrapped SWCNTs. A 3 mg/mL solution prepared by blending the PMMA with 5 wt% of PSC/SWCNTs was also formulated and mixed for 1 h with continuous stirring to obtain a homogeneous solution. Finally, the prepared solutions were inkjet-printed using a Dimatix printer onto a channel area and annealed at 100 °C for 30 min to evaporate the

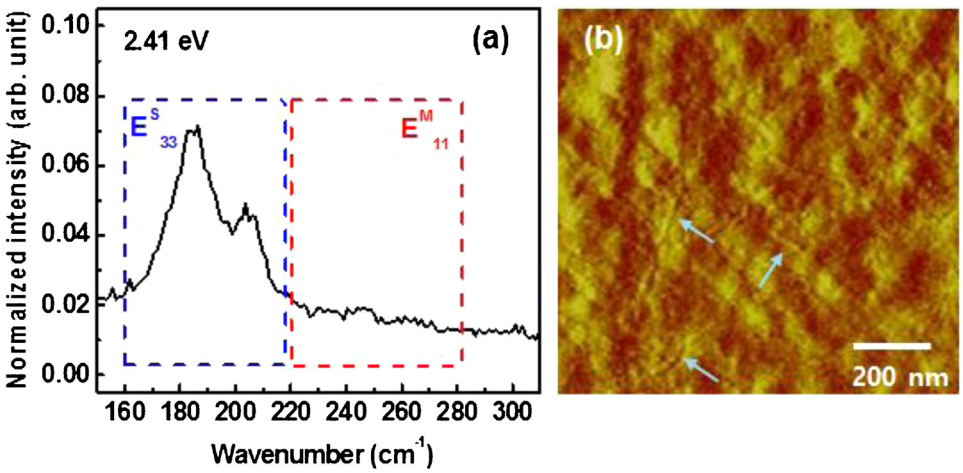

Fig. 2. (a) RBM regions in the Raman scattering spectra of the separated sc-SWCNTs; the excitation energy was 2.41 eV. (b) Tapping-mode AFM image of the PSC/SWCNT network blended with the polymer binder. The arrows show the sc-SWCNTs dispersed in the PMMA binder.