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Memory behavior of multi-bit resistive switching based on multiwalled carbon nanotubes

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A B S T R A C T

Multiwalled carbon nanotube (MWCNT) displays peculiar electrical behavior, with its nano-configuration consisting of exceptional graphene flakes. As for MWCNTs blended into polymethyl methacrylate (PMMA), sandwiched Ni/MWCNTs(2 wt%):PMMA/ITO was manufactured to investigate into the multi-bit resistive switching regarding the turn-on compliance current as well as the thickness of the active layer. It bears ternary write-once read-many-times (WORM) memory, whose current proportionality between ON-state and OFF-state can exceed 10^7. Moreover, the memory performance, covering the long-term retention (>10^6 s), better endurance (>10^{12} cycles) and device-to-device profiles, confirms the excellent ternary memory of MWCNTs:PMMA. Concentration on multi-bit resistive switching in respect of MWCNTs underlies performance enhancement, higher integration and advanced architecture.

1. Introduction

Numerous eyes have been currently on organic-based resistive switching [1–4]. As an emerging kind of medium, it is favorable for structural simplicity, lower energy consumption, higher integration density, solution processibility, mechanical flexibility, capacity for three-dimensional stacking together with compatibility with complementary metal oxide semiconductor (CMOS) processing [5–7]. Nanodevices connected with metal oxides, organic small-molecule, polymer, nanocomposites and biomaterials are predominantly associated with organic light-emitting diodes, organic solar cell, organic field effect transistor, and biosensors. Organic materials, by means of design and synthesis, can be spin-coated or ink-jet printed on the plastic, glass, and CMOS hybrid integrated circuit.

Polymethyl methacrylate (PMMA) has remarkable dielectric and insulator properties. For the metal-insulator-semiconductor (MIS) memory structure, PMMA as dielectric layer is inserted by nanomaterials, such as carbon nanotubes (CNTs), graphene, Au–Pt–Ag, Au–Cu and CdSe, for which the memory performance is implemented by the charging and discharging effects [8,9]. Playing the role of matrix, PMMA blended with carbon nanomaterials appeals to great attention [10–13]. Carbon nanomaterials like graphene, CNTs, fullerene as well as its derivatives play a crucial role in organic-based memory [14–17]. Advance of CNTs for aspects of biosensor, supercapacitor, and lithium ion battery triggers the fulfillment of CNTs in the semiconductor industry [18–20].

Requirement of both storage ability and calculation speed increasingly unfolds the multi-bit memory in a logic memory cell based on organic materials [21–23]. In this work, the multi-bit resistive switching by blending PMMA with multiwalled carbon nanotubes (MWCNTs) has been successfully manufactured and characterized. Particularly, efforts are made to learn the electrical performance of the resistive switching, taking the compliance current and the thickness of the active layer into consideration.

2. Experimental

For carboxylic (–COOH) MWCNTs (purchased from Hengqiu Tech. Inc.), the –COOH content, internal diameter, external diameter, length, specific surface area, density and resistivity is 2 wt%, 5–10 nm, 10–20 nm, 10–30 μm, >200 m^2/g, ~2.1 g/cm^3, >100 s/cm, respectively. PMMA (molecular weight M_W = 120,000–150,000) was purchased from ARKEMA.

MWCNTs (2 wt%) and PMMA blend was dissolved into the chloroform. At ambient temperature, the solution was stirred by the magnetic stirrer for more than 24 h. The glass substrate, with the indium-tin-oxide (ITO, 6–9 Ω/□) deposited, was sequentially cleaned by the detergent, distilled water, acetone, and ethanol, and proceeded to be kept 40 °C in the vacuum furnace for 30 min. After

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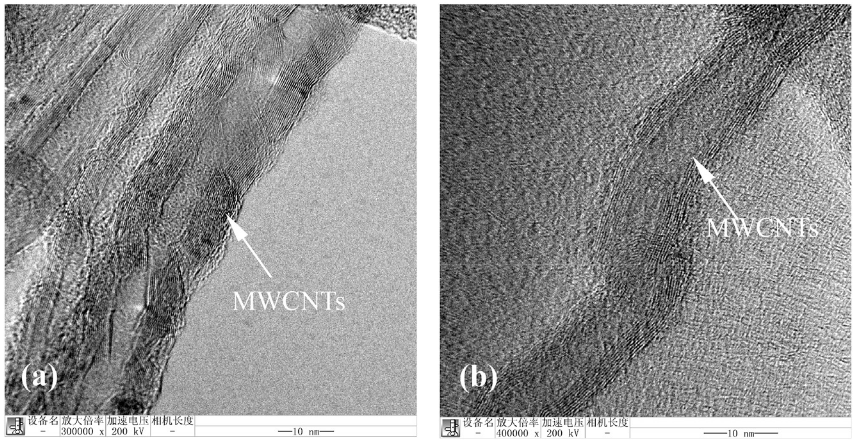

Fig. 1. TEM patterns of MWCNTs in (a) low and (b) high solution, respectively.

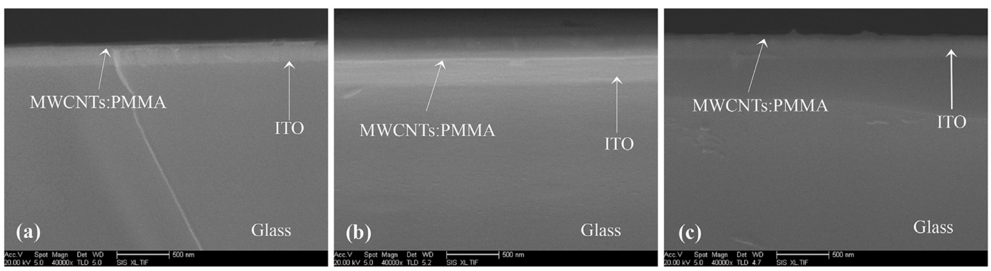

Fig. 2. SEM images of the cross-sections of the MWCNTs (2 wt%):PMMA nanocomposite film with the thickness (a) 30 nm, (b) 40 nm, and (c) 60 nm.

spin-coating the solution uniformly to fabricate an active layer at 3000 rpm, the solvent was eliminated from the coatings through the vacuum furnace in 10^{-5} torr and 60 °C for 6 h. Finally, a Ni electrode, 300 nm thick, was deposited on the top of the nanocomposite film, protected by the mask pattern with the diameter of 0.2 mm. It is noted that ITO acts as the anode while the Ni electrode can be regarded as the cathode.

MWCNTs and MWCNTs:PMMA nanocomposite films were characterized by JEM-2100 Transmission Electron Microscopy (TEM) and FEI Sirion Scanning Electron Microscope (SEM), respectively. Foss DS 2500 Infrared Spectrometer was employed to test Fourier Transform Infrared (FTIR) spectra of the MWCNTs:PMMA solution and nanocomposite films that were prepared by spin-coating on the ITO/Glass substrate. At ambient temperature, electrical characteristics of the resistive switching Ni/MWCNTs:PMMA/ITO were detected by KEITHLEY 4200-SCS Semiconductor Characterization System in air.

3. Results and discussion

Fig. 1 displays TEM patterns of MWCNTs in low and high solution. It is clear that MWCNT consists of single carbon nanotubes in the concentric shell, and particularly, the layer-to-layer distance approaches the interlayer spacing of graphene δ (~0.34 nm). As exhibited in Fig. 2, the cross-sections of the MWCNTs:PMMA

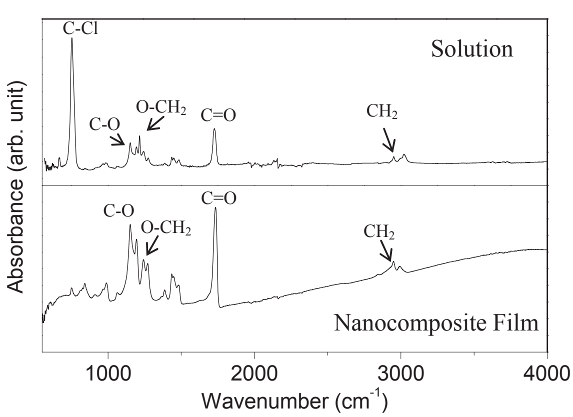

Fig. 3. FTIR spectra of the MWCNTs (2 wt%):PMMA solution and spin-coated nanocomposite film with the thickness of 30 nm.

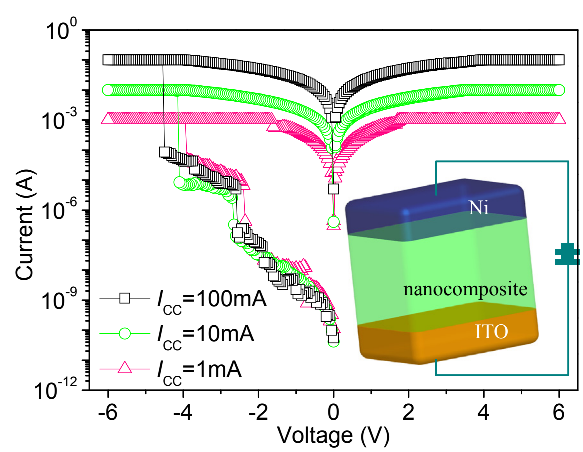

Fig. 4. *I-V* characteristics of Ni/MWCNTs:PMMA (30 nm)/ITO under the compliance current 100 mA, 10 mA and 1 mA, respectively, and its sandwiched configuration in inset.