Sensors and Actuators A 234 (2015) 282–289

Contents lists available at ScienceDirect

Sensors and Actuators A: Physical

journal homepage: www.elsevier.com/locate/sna

Bistable electrical switching and nonvolatile memory effect based on the thin films of polyurethane-carbon nanotubes blends

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ARTICLE INFO

Article history:
Received 10 June 2015
Received in revised form 5 August 2015
Accepted 8 September 2015
Available online 12 September 2015

Keywords:
Carbon nanotube
Polyurethane
Flash
Memory
Conductance switching

ABSTRACT

Bistable nonvolatile memory devices with resistive switching characteristics were fabricated based on polyurethane (PU) doping with single-wall carbon nanotubes (SWCNTs). It has been demonstrated that the addition of SWCNTs in PU layer could enlarge the ON/OFF current ratio from 10^2–10^4 and keep a long retention time over 9 h. Besides, the increase of SWCNTs and charge traps induced by SWCNT reduced the current in OFF state and enhanced the memory window significantly. Furthermore, the resistive switching behavior of indium tin oxide/PU + SWCNTs/aluminum device was attributed to the formation and breakdown of SWCNTs percolated network structure in the PU composites.

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1. Introduction

Polymer-based resistive random access memory (RRAM) is rapidly emerging as the technology for nonvolatile memory applications. For the past few years, polymeric materials and their composites with carbon nanotube (CNT) have found a large number of applications in organic electronics [1,2]. On account of the high strength and high modulus of CNTs and their large aspect ratio, CNTs have been proved the most ideal reinforcement agent for polymers [3,4]. In particularly, the excellent electrical and thermal conductivity properties of CNTs are very favorable for preparing conductive polymer composites [5–7]. The interfacial interaction between CNTs and polymer matrix and the dispersion state of CNTs are the main factors determining the properties of CNT-based composites.

When a certain concentration is achieved, CNTs tend to form a percolated network structure in the matrix, which not only influences the behavior of polymer but also influences the electrical and thermal conductivity of composites [8]. Nevertheless, once the percolated network structure is formed, polymer composites usually exhibit largely enhanced electrical conductivity, and in this condition, the composites are changed from insulated material to semiconductor or even conductive material [9,10]. In recent years, conductance and resistive switching characteristics of polymer memory devices with CNTs have attracted much interest [11–13]. Different kinds of polymer-based RRAM materials have been developed through adding CNTs [14–17]. In the last years several attempts have been made to mix CNT with organic materials, Liu et al. found that with varying the CNTs content in PVK composite thin films, distinctly different electrical conductance behaviors, such as insulator behavior, WORM, flash, and conductor behavior were observed [11]. Later, by dopping various weight percentages of CNT in PVA, Pandurangan et al. suggested the characteristic behavior of an insulator, WORM, flash, and conductor effect is exhibited once again [14]. Kang et al. found that the device performance was substantially enhanced when the electroactive polymer (F12TPN) layer was doped with CNTs [16]. Hümmelgen et al. proposed that a very small quantity of functionalized multi-walled CNTs embedded in a PEDOT: PSS conducting polymer matrix show erasable and rewritable memory behavior [17]. Our previous work on electrical switching and memory effect in mixing SWCNTs with PEDOT: PSS systems showed the bi-directionally switchable WORM characteristics [18].

We have previously shown that the resistance switching memory characteristics based on the blend materials of oxadiazole acceptor and carbazole donor, due to the strong interaction between acceptor and donor, the devices performance has steadily improved [19]. In view of plenty of interesting works on electrical switching and memory effects in mixed polymer systems [20,21], the effect of mixing level on the electrical behavior of composite systems seemingly deserves further exploration.

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http://dx.doi.org/10.1016/j.sna.2015.09.016
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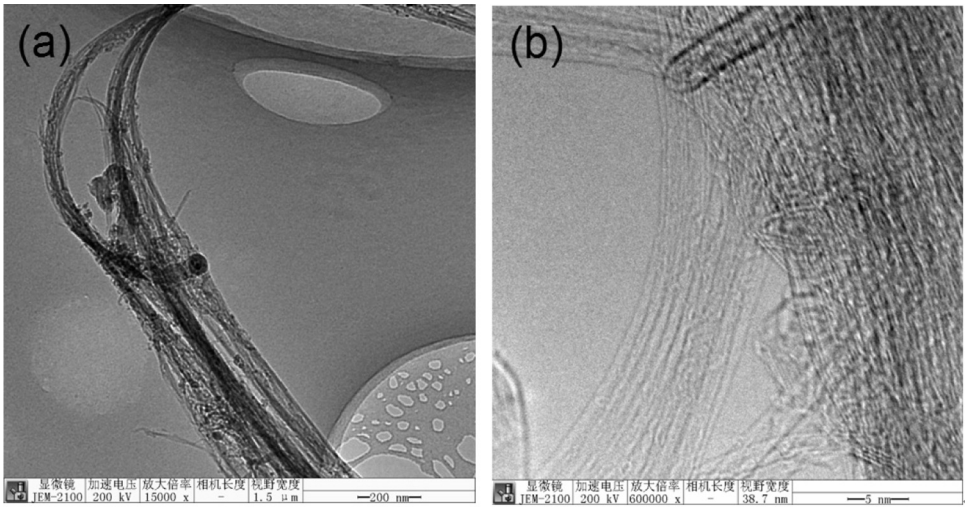

Fig. 1. TEM of SWCNTs (a) Low resolution (b) High resolution.

Good scalability, a simple structure, low cost and easy processing of polymer memories, make them suitable for the current memory technology. Recently, incorporating the CNTs into polymer to improve the solubility and film-forming properties from solution has been widely studied to simplify the device fabrication process. In the present study we have utilized an insulating polymer, polyurethane (PU) to fabricate a PU + SWCNT composite layer for the memory device. We have chosen PU as the base matrix due to the following advantages; it has a good film forming, low cost, is water soluble, easily processed, and good adhesive nature for applications in electronic devices. Also, it is considered a good host matrix for CNTs and small molecule. Here, we introduced different contents of SWCNTs into PU and investigated the resistive switching behavior of indium tin oxide/PU + SWCNTs/aluminum (ITO/PU + SWCNTs/Al) device. It is revealed that the addition of SWCNTs in PU could enlarge the ON/OFF current ratio from 10^2–10^4 and keep a long retention time over 9 h. Furthermore, different from our previous study about polymeric materials and their composites with CNT, the ITO/PU+SWCNTs/Al device exhibited another nonvolatile switching behavior (flash memory effects).

2. Experiment

2.1. Materials

The PU aqueous solution containing 6 wt.% PU was provided by Sigma–Aldrich. SWCNTs were purchased from NanoLab Inc. (Beijing, China). The outer diameter, length, specific surface area, and purity of the SWCNTs were 1–2 nm, 5–30 μm, 450 m^2/g, and 95%, respectively. Transmission electron microscopy (TEM) measurements were carried out to investigate the micro-structural properties of those SWCNTs, as shown in Fig. 1.

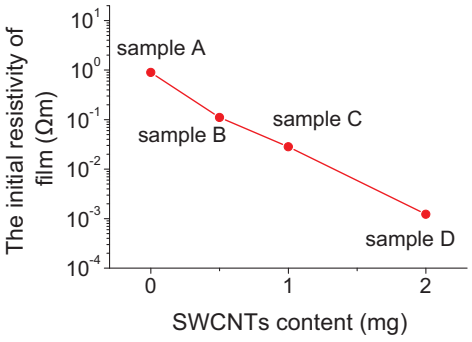

Fig. 3. The initial resistivity of PU + SWCNTs film.

The carboxyl-functionalized SWCNTs were first prepared *via* ultrasonication in 1:3 (volume ratio) mixture of concentrated nitric acid/sulfuric acid at 70 °C for 8 h. Then the acidic form of SWCNTs was converted into sodium salt by ultrasonication in 5 mmol/l aqueous NaOH solution. The sodium salt form of SWCNTs was refluxed with tetrabutylammonium hydrogensulfate and 1-bromododecane until the suspension became clear and the esterified SWCNTs precipitated out of the solution. The black solids were collected and dried in a vacuum oven at 70 °C overnight. The functionalized SWCNTs of different mass (see Table 1) were added into PU aqueous solution (1 ml, 6 wt.%), denoted as PU + SWCNTs. The mixed solution was ultrasonicated for 50 min.

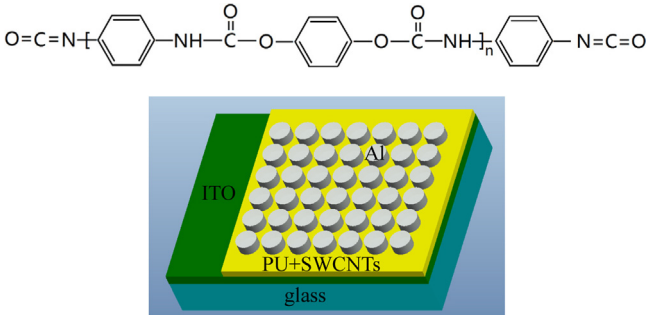

Fig. 2. Schematic of the memory device with the ITO/PU + SWCNTs/Al structure. Inset: chemical structure of PU.

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