



Effect of device structure on the resistive switching characteristics of organic polymers fabricated through all printed technology



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ABSTRACT

Developments in organic bi-stable non-volatile memory devices have shown that organic materials are essential for the next generation of electrical memory unit owing to their low cost, high flexibility and large scalability. This study depicts an important aspect of organic memory devices by observing the effect of changing device structure on its switching characteristics. Memory devices with a bilayer and bulk-heterojunction structure were fabricated through an all printed technology by utilizing two organic polymers such as MEH: PPV and PMMA. Silver (Ag) was selected as the top and bottom electrode due to its high conductivity and easy processing. Though identical polymers were used in both device structures, but interestingly change in structure caused change in properties. It was observed that bilayer structure had much higher switching ratio and stability against various biasing cycles as compared to its bulk-heterojunction counterpart. Superior switching characteristics of bilayer structure were due to the presence of a well-defined interface between both polymers. Bulk-heterojunction device suffers the drawback of phase separation in a single organic layer between the two polymers.

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1. Introduction

After the physical realization of a memristor by HP laboratory in 2008 [1] whose theory was earlier proposed by Leon Chua back in 1971 [2], the memristor was given the status of fourth basic element. This resistive switching occurs due to the movement of charge carriers through the active layer sandwiched between two metal electrodes when external biased voltage is applied. The resistive switching based memory devices became the center of interest for researchers due to their various applications such as high scalability, digital logic design, non-volatile memory, low power dissipation, and neuromorphic research [3–5]. Organic resistive switches are considered to be the potential candidate to replace traditional transistors from memory and switching applications owing to their exceptionally small size, increasing off/on ratio, simple structure, low cost and easy fabrication process. Due to these significant potential applications of resistive switches in

electronic industry, various research efforts are currently dedicated for its realization as a commercial electronic component. These research efforts mainly focus on three categories such as varying active layer material, device fabrication technology and structure of device respectively. Materials ranging from insulators, polymers and inorganic semiconductors with memristive behavior have been explored and investigated extensively. Various sophisticated fabrication technologies including nanoimprint lithography (NIL), electrohydrodynamic atomization (EHDA), atomic layer deposition (ALD), sputtering, vacuum evaporation and inkjet printing have also been reported to manufacture resistive switching memory devices [6–8]. However, effect of changing device structure on the resistive switching properties of a memory devices has been neglected as compared to other two research categories of testing different materials and fabrication technologies respectively.

Therefore, in this study we have explored the effect of changing device structure on the resistive switching characteristics of a memory device by fabricating two devices including bulk-heterojunction and bilayer structure of active layer. Mostly, single active layer has been reported in memory devices [9–13]. Organic

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materials have shown excellent switching properties when used as a single active layer material [14–21]. Memory devices with hybrid active layer such as a bilayer of organic and inorganic semiconductors have also been reported in literature [22,23]. Effects of adding a metal and inorganic semiconductor nanoparticles as the trap states in the polymer active layer have also been explored [24,25]. However, very little work has been done on using two different organic polymers jointly to form the active layer of a memory device [26]. Therefore, we have opted for two organic polymers to explore the effect of changing structure on the resistive switching properties. Organic polymers of Poly [2-methoxy-5-(20-ethylhexyloxy)-(p-phenylenevinylene)] MEH:PPV and Poly [methyl methacrylate] (PMMA) were selected over other materials as the active layer for both devices owing to their low cost, low power consumption, light weight, low temperature processing, large area applications, high dissolvability in several solvents and ease of fabrication [27–29]. Furthermore, these polymers are green materials such that they are not harmful for the environment [30].

The configurations of bulk-heterojunction and bilayer structure device were Ag/MEH:PPV-PMMA/Ag and Ag/PMMA/MEH:PPV/Ag respectively. Bottom Ag electrodes were printed by a highly promising printing technique of reverse offset [31,32] while thin films of organic materials were deposited by using state of the art electrohydrodynamic atomization (EHDA) [33–35]. Top Ag electrodes were patterned by screen printing to complete the device fabrication [36,37]. PMMA was selected owing to its huge energy barrier that is vital for memory effect. It also offers a high density of trap states for the charge carriers. MEH:PPV acts as the charge trapping element while PMMA restricts the flow of those trapped charge carriers back to their original energy states due to its high value of dielectric coefficient. Energy barrier of PMMA and charge trapping property of MEH:PPV play a vital role in the nonvolatile memory behavior shown by our devices. This work draws an extremely important conclusion about the research of nonvolatile memory devices.

2. Materials and methods

2.1. Materials

All the solvents including acetone, ethanol (99.5%), and toluene were purchased from Sigma-Aldrich. Both organic materials MEH:PPV ($C_{18}H_{28}O_9$)_n and PMMA [$CH_2C(CH_3)(CO_2CH_3)$] were also purchased from Sigma-Aldrich. Highly conductive silver paste (60–80 wt% and 2.5 mΩ/sq/mil) to deposit electrodes was purchased from MicroPE® PARU.

2.2. Ink formation

0.5% w/v ink of MEH:PPV was prepared by dissolving 0.05 g of MEH:PPV powder in 10 ml of toluene. 0.5% w/v ink of PMMA was prepared by dissolving 0.05 g of PMMA powder in 10 ml of toluene. The composite ink of MEH:PPV and PMMA with 1% w/v was prepared by dissolving 0.05 g of each MEH:PPV and PMMA in 10 ml of toluene. The concentration of both polymers was kept the same in bilayer and bulk-heterojunction memory devices. The solutions were bath sonicated for 10 min followed by stirring on a magnetic stirrer for 2 h. The MEH:PPV and composite inks were kept in dark and cold environment because exposure to high temperature and white light changes the chemical properties of MEH:PPV. All the prepared inks with the chemical formula of MEH:PPV and PMMA polymers are shown in Fig. 1.

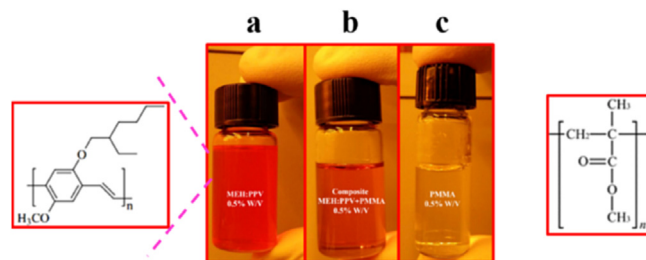


Fig. 1. Structural formulas of used organic semiconductors and optical images of synthesized solutions to deposit active layer thin films of both memory devices a) MEH:PPV b) Nanocomposite of MEH:PPV and PMMA c) PMMA.

2.3. Fabrication of bilayer and bulk-heterojunction memory devices

All the fabrication steps of bulk-heterojunction and bilayer memory devices are shown sequentially in the flow diagram of Fig. 2 from top to bottom labelled as A and B respectively.

The substrates were bath sonicated with acetone, ethanol, and deionized water for 15 min each prior to device fabrication. Cleaned substrates were passed through UV ozone treatment for 5 min to remove organic residues resulting in a better adhesion and proper wetting of thin films on the substrate surface. Bottom Ag electrodes for both devices i.e. bilayer and bulk-heterojunction structure were deposited on the cleaned glass substrates by a high resolution printing technique of reverse offset. Details of this relatively new and advanced printing technique can be found in our previous work [32]. These electrodes were cured at 120 °C for 15 min to establish its conductive properties. Thin film of MEH:PPV-PMMA composite was sprayed on top of bottom Ag electrodes through state of the art printing technique of EHDA for the bulk heterojunction structure of active layer. For the bilayer device, active layer of PMMA was sprayed on top of bottom Ag electrodes. MEH:PPV was sprayed over PMMA to complete the bilayer structure. Both layers were sprayed through EHDA printing system. Bulk-heterojunction and bilayer active layers were sintered in a furnace at 110 °C for 1 h each. Finally, top Ag electrodes were patterned through screen printing over bulk-heterojunction and bilayer thin films. The top electrodes were cured at 120 °C for 15 min.

Magnified optical images of as fabricated bilayer and bulk-heterojunction memory devices along with their schematic diagrams are illustrated in Fig. 3(a) and (b) while Fig. 3(c) and (d) illustrate the 3D schematic diagram of each memory device. The inset of Fig. 3(b) and (d) illustrates the cross section SEM images of bilayer and bulk-heterojunction active layer. Total thickness of active layers in both memory devices was nearly equal to each other to nullify the possible effect of thickness on the switching properties.

3. Results and discussions

3.1. Morphological characterization

Surface morphology of the prepared thin films was characterized by using a JEOL JSM –6310F FESEM. Fig. 4 shows a high magnification image of as deposited PMMA, MEH:PPV and their composite thin films. Fig. 4(a) shows the granular nature of PMMA thin film whereas Fig. 4(b) shows the micro-cracks on the surface of MEH:PPV thin film that is its intrinsic property. Fig. 4(c) shows the surface of MEH:PPV and PMMA composite such that the micro cracks of MEH:PPV were filled by the more compact thin film of PMMA. To find the purity of thin films, Fourier Transform Infrared

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