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Enhancements of the memory margin and the stability of an organic bistable device due to a graphene oxide:mica nanocomposite sandwiched between two polymer (9-vinylcarbazole) buffer layers

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ABSTRACT

Current-voltage (I–V) curves for the Al/polymer (9-vinylcarbazole) (PVK)/graphene oxide (GO):mica/PVK/indium-tin oxide (ITO) devices at 300 K showed a current bistability with a maximum high conductivity (ON)/low conductivity (OFF) ratio of 2×10^4, which was approximately 10 times larger than that of the device without a PVK layer. The endurance number of ON/OFF switchings for the Al/PVK/GO:mica/PVK/ITO device was 1×10^2 cycles, which was 20 times larger than that for the Al/GO:mica/ITO device. The "erase" voltages were distributed between 2.3 and 3 V, and the "write" voltages were distributed between −1.2 and −0.5 V. The retention time for the Al/PVK/GO:mica/PVK/ITO device was above 1×10^4 s, indicative of the memory stability of the device. The carrier transport mechanisms occurring in the Al/PVK/GO:mica/PVK/ITO and the Al/GO:mica/ITO devices are described on the basis of the I–V results and the energy band diagrams.

1. Introduction

Nonvolatile resistive-switching devices have been attractive because of their potential applications in next-generation nonvolatile memristive devices due to their excellent advantages of low-power consumption, high-density storage, high-mechanical flexibility, low cost, and simple fabrication [1–5]. Among the various kinds of memristive devices, organic bistable devices (OBDs) based on hybrid inorganic/organic nanocomposites have emerged as excellent candidates for promising applications in flexible, mobile, portable memories due to their relatively simple fabrication process without the need for additional sources and drains [6–11]. Among the diverse nanomaterials, graphene-relevant materials with a monolayer thickness are predicted to have remarkable properties, which make them attractive materials for OBDs [12–16]. Especially, OBDs fabricated utilizing graphene-oxide (GO) thin films and conjugated-polymer functionalized GO films have currently emerged as excellent candidates for potential applications in next-generation electronic devices [17–19]. Graphene oxide sheets are oxygenated bearing hydroxyl and epoxide functional groups on the top and bottom surfaces of their sheet [20]. The presence of functional groups makes GO sheets electrically insulating. Also, graphene oxide is hydrophilic unlike graphene, Thus easily dispersible in hydrophilic solvents [21]. Due to its water solubility, GO can be transferred onto any substrates uniformly using simple methods such as spin-coating [22]. And water solubility makes it very easy to form GO thin films and the ultrathin thickness of a GO sheet is a beneficial aspect for high density fabrication which makes GO the excellent candidates for memristive devices [23]. Some investigations about the electrical characteristics of nonvolatile memory devices based on GO nanocomposites have been performed [24–26]. But The maximum high conductivity (ON)/low conductivity (OFF) current ratio of the GO-based OBDs is only approximately 10^3, so their stabilities should be improved to provide potential applications [27]. The degradation behaviors of the GO-based OBDs might originate from the lattice defects of the chemically functionalized GO [28,29]. Thus studies concerning the improvement of the characteristic of the graphene oxide-based material memories is important with both high memory margin and operating stability for the development of next generational memristive devices.

Because a muscovite-type mica bulk, which is similar to a GO layer, has a large band gap, the muscovite-type mica layer acts as an insulator layer. However, when multilayer mica nanosheets are prepared by using liquid-phase exfoliation, the number of required nanosheets is reduced. Because the bandgap

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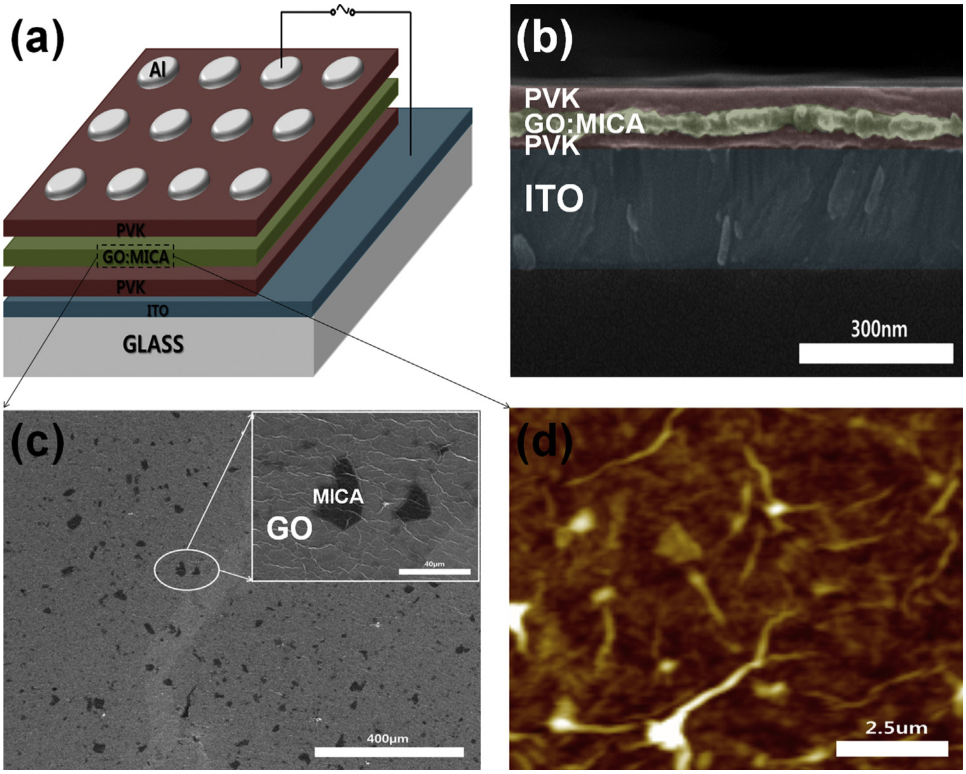

Fig. 1. (a) Schematic diagram of the Al/PVK/GO:mica/PVK/ITO glass device. (b) Cross-sectional scanning electron microscopy (SEM) image of the PVK/GO:mica/PVK/ITO glass structure. (c) SEM image of the GO:mica layer. (d) AFM image of the GO:mica layer.

energy of the mica layer decreases with decreasing number of mica layers, these bandgap engineered 2D mica nanosheets present an exciting opportunity for achieving unique physical properties in 2D materials and may lead to diverse applications in 2D electronic devices [30]. Even though some studies concerning the electrical properties of OBDs fabricated utilizing hybrid inorganic/organic nanocomposites containing GO have been performed, investigations on the electrical bistabilities, the memory stabilities, and the carrier transport mechanisms of OBDs fabricated utilizing exfoliative mica nanocomposites blended with GO and sandwiched between two PVK layers by using a simple spin-coating method have not been reported yet. Furthermore, studies on the memory stabilities and the carrier transport mechanisms of OBDs based on GO:mica nanosheet nanocomposites are very important for improving device efficiencies.

In this work, we describe the enhanced memory margins and the improved operating stabilities of OBDs based on a GO:mica nanocomposite, where the nanocomposite is sandwiched between polymer (9-vinylcarbazole) (PVK) layers to form a PVK/GO:mica/PVK structure. Scanning electron microscopy (SEM) and atomic force microscopy (AFM) measurements were conducted to investigate the structural properties of the PVK/GO/mica/PVK nanocomposite layer. Current-voltage (I–V) measurements were performed to investigate the memory performance of the fabricated devices. Retention and endurance measurements were carried out to investigate the stability and the durability of the devices. A dramatic increase in the difference between the high and the low conduction states in the fabricated OBDs containing a PVK buffer layer is observed. This behavior has been described in this paper on the basis of the experimental results, as are, based on the fitting results for the I–V curves and the energy band diagrams, the mechanisms underlying the enhanced properties of the devices.

2. Experimental details

Nanocomposites consisting of GO and a mica layer sandwiched between two PVK layers were prepared on indium-tin-oxide (ITO)-coated glass substrates by using a spin-coating technique. The ITO-coated glass substrates, which had been chemically-cleaned using acetone, methanol, and deionized water, were dried by using N_2 gas with a purity of 99.9999%. A solution made of 1-wt% PVK in chlorobenzene was used for the preparation of the buffer layers. The GO solution was prepared from natural graphite by using a modified Hummer's method [31] and was exfoliated into GO (2-wt%) by sonication for 2 h in ethanol. The exfoliated muscovite-type mica particles were fabricated by using ultrasonication, centrifugation, and ultrasonication [25]. Both solutions were ultrasonically treated to obtain a uniform suspension. The solution was mixed with the exfoliated muscovite-type mica and was dispersed uniformly in ethanol, and GO was formed with a mass ratio of 1:1, based on the concentrations of GO and the mica solution, by using an ultrasonic treatment at 27 °C for 3 h in order to achieve a solution of exfoliated mica and GO.

The solution of GO:exfoliated muscovite-type mica can be dissolved uniformly in ethanol. The 1-wt% PVK solution was spin-coated on a cleaned ITO glass substrate at 1000 rpm for 5 s, 3000 rpm for 10 s, 5000 rpm for 30 s, 3000 rpm for 10 s, and 1000 rpm for 5 s. After the sample had been spin-coated, it was thermally annealed at 135 °C for 20 min to remove the solvent. The GO:mica trapping layer was spin-coated onto the PVK tunneling layer at 500 rpm for 5 s, 1000 rpm for 5 s, 2000 rpm for 30 s, 1000 rpm for 5 s, and 500 rpm for 5 s. After the GO:mica trapping layer had been deposited on the PVK layer, the device was thermally annealed at 100 °C for 20 min. Subsequently, another top PVK buffer layer was formed by using the same solution as was used for the bottom PVK layer, and the layer was spin-coated onto the GO:mica trapping layer at 1000 rpm for 5 s, 2500 rpm for 10 s, 4000 rpm for

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