



Effect of Ag doping and insulator buffer layer on the memory mechanism of polymer nanocomposites



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ABSTRACT

Resistive memory devices based on nanocomposites have attracted great potential for future applications in electronic and optoelectronic devices. The successful synthesis of aqueous CdSe nanoparticles has been provided with UV–Vis and Photoluminescence spectroscopy. The two terminal planar devices of CdSe nanocomposite have been fabricated. The effect of Ag doping and additional dielectric buffer layers on the memory devices have been studied by current–voltage (I – V) and capacitance–voltage (C – V) measurements. The devices show hysteresis loops in both positive and negative bias directions. The memory window has been found to be increased with both Ag doping and PVA layer addition. The charge carrier transport mechanism in the memory devices has been studied by fitting the I – V characteristics with the theoretical model, Space charge conduction model (SCLC). C – V hysteresis loop in both positive and negative bias directions indicate that both the electrons and holes are responsible for memory mechanism of the devices. The switching mechanism of the memory devices has been explained by charge trapping/detrapping model. The retention characteristics show good stability and reliability of the devices.

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1. Introduction

Nanocomposites (NCs) have attracted much attention due to their promising potential applications in electronic and optoelectronic devices [1–4]. The memory devices have become increasingly important in daily life due to the widespread use of portable electronic devices such as mobile phones, media players and personal data storage media such as compact discs and floppy discs [5,6]. Resistive Random Access Memory (RRAM) devices are one of the most important components in modern electronic devices to store information for logic operation [7–10]. RRAM exhibits resistive switching between a high resistance state (HRS) and a low resistance state (LRS). These two states represent the logic values 0 and 1, respectively. Depending on voltage polarity, the resistive-switching behaviour of an RRAM device can be classified as unipolar or bipolar. In unipolar RRAM, resistive switching is induced by a voltage of same polarity whereas for bipolar switching, one polarity is used to switch the device from HRS to LRS, and the opposite polarity is used to switch the device back to HRS. RRAM has advantages over other memory devices such as easy fabrication, simple structure, excellent scalability, fast switching,

high integration density, and good compatibility with the complementary metal oxide semiconductor (CMOS) technology [11–14]. Thus more attention is being focused on these devices.

There have been many studies on electronic memory devices based on metal nanoparticles, oxide and organic materials [15–17]. But the memory device using the organic/inorganic materials is still under the exploration stage. Basically there are two types of structures for memory devices: two terminal bistable memory devices [18,19] and three-terminal transistor like memory devices [20]. In two terminal memory devices, the organic/inorganic material is sandwiched between two metal electrodes. During its operation, the conductivity of the active layer is changed by applying a voltage across the device, allowing the bit of data to be stored [21]. The two terminal bistable memory devices have attracted attention due to their low operating voltage and simple fabrication process as compared to three terminal memory devices [22]. Recently, a new class of memory devices has been emerged on the basis of shape retaining property of polymers known as shape memory devices. Lu et al. [23,24] had studied the temperature dependent shape recovery effects in amorphous shape memory polymers. They studied the shape recovery behaviour using three thermodynamic constitutive frameworks: Arrhenius, Vogel–Fulcher–Tammann (VFT) and Bässler laws. Out of these, VFT law provide accurate results, which are in good agreement with the reported (literature) experimental results.

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Different materials and device structures are studied for the memory devices according to the requirements [25–27]. Polymer nanocomposites (PNCs) have currently emerged as excellent candidates for potential applications in next-generation memory devices [28]. These materials have advantages of low cost processing and are suitable for large area device fabrication [29]. In PNCs, the organic polymer not only acts as a stabilizer for the nanoparticles, but also the optical and mechanical properties of the polymer are combined with the specific features of the nanoparticles [30]. In PNCs based memory device, the nanoparticles act as charging and discharging islands in the polymer matrix. Both the insulating and conducting polymers can be used as a matrix in the PNCs for memory device applications [31,32]. The electrical hysteresis in the I - V characteristics is essential requirement for the memory phenomena in electronic devices. The memory devices are characterized by ON/OFF ratio. The value of ON/OFF ratio should be high enough to prevent the misreading of stored data. The mechanism of memory devices is uncertain and may be explained on the basis of many phenomena such as metal filament formation, charge storage in nanoparticles, and redistribution of oxide vacancies [33–36].

In this paper, we discuss the fabrication and characterization of two terminal memory devices with CdSe nanocomposite (NC) as an active layer sandwiched between two metal electrodes. The effect of PVA buffer layers on the memory performance of the devices have been investigated. The effect of Ag doping is also studied on all the devices, as the small amount of dopant in the semiconductor nanocrystals plays a significant role in changing the electronic properties of the semiconductor [37]. CdSe and Ag doped CdSe (Ag:CdSe) NC has been prepared by chemical synthesis technique. The prepared CdSe and Ag:CdSe NCs have been characterized by UV-Vis and photoluminescence (PL) spectroscopy. The memory performance of the devices has been investigated with current-voltage (I - V) and capacitance-voltage (C - V) measurements. To understand the charge carrier transport in memory devices, the I - V characteristics are fitted with the theoretical model. To check the reliability of the devices, the retention characteristics are performed.

2. Experimental details

2.1. Materials used

Cadmium acetate ($\text{Cd}(\text{CH}_3\text{COO})_2$) has been purchased from Qualigens fine chemicals, Sodium sulphite (Anhydrous purified) and Ammonia from Merck Specialities Pvt. Ltd, Selenium from Loba Chemie Pvt. Ltd., AgNO_3 from Universal Laboratories Pvt. Ltd., Poly(vinyl alcohol); PVA ($M_w = 140,000$) was obtained from HiMedia Laboratories Pvt. Ltd and used without further purification. Deionized water was used in all experiments and all other chemicals were of analytical grade.

2.2. Fabrication of nanocomposites

For CdSe NC preparation, sodium selenosulphate (Na_2SeSO_3) was chosen as the selenium source. Na_2SeSO_3 aqueous solution (0.50 M) was prepared by adding sodium sulphite and selenium powder into 100 ml of distilled water, stirred for 7 h at 60 °C. 3% PVA solution was prepared by adding 3.0 g PVA to 100 ml of deionized water and stirring at 60 °C until a viscous transparent solution was achieved. By adding ammonia, the pH of the solution was maintained at ~ 10 . Cadmium acetate (0.07 M) and Na_2SeSO_3 solutions were added with constant stirring into PVA solution with Cd:Se precursor ratio 1:1 at 60 °C. AgNO_3 (0.3 M) solution was prepared in de-ionized water. For Ag doping of CdSe, 10 μl of AgNO_3

solution was added to 10 ml of CdSe NC solution. The resulting solution was stirred for an hour. The colour of solution was observed to change from reddish to reddish brown with Ag doping. UV-Vis spectra of prepared nanocomposites was recorded in the wavelength range ~ 300 – 800 nm using single beam Spectroscan 30 spectrometer. The PL spectra was recorded using spectrophotometer LS-55 (Perkin Elmer Instruments) with accuracy = ± 1.0 nm.

2.3. Device fabrication and characterization

To study the memory effect of NCs, three sets of memory devices were prepared. Al substrate was used as a bottom electrode whereas Ag was used as a top electrode for all memory devices. Before film deposition, the Al substrates were cleaned with deionized water, and then washed with KOH pellets to remove native alumina layer and other soluble salts present on its surface. For the first set of devices (Set A), CdSe (or Ag:CdSe) NC solution was casted onto the Al substrate as an active layer. When the films were dried under ambient conditions, Ag top electrode was deposited on the NC thin film by thermal evaporation method. For the second set of devices (Set B), first a PVA buffer layer was deposited between the bottom electrode and the CdSe (or Ag:CdSe) NC layer.

Whereas, for the third set of devices (Set C), two PVA buffer layers were inserted in the device structure above and below the CdSe NC layer respectively. All the deposition processes were carried out at room temperature. The schematic architecture of the memory devices is shown in Fig. 1. The I - V measurements on the device were taken with Keithley 6517A electrometer. The C - V measurements were taken with Hioki 3522-50 LCR Hi-Tester. All the C - V measurements were done at 100 kHz. The applied voltage was swept between +4 V and -4 V for both I - V and C - V measurements.

3. Results and discussion

Fig. 2 shows the UV-Vis and PL spectra of aqueous CdSe and Ag:CdSe NC. CdSe NC shows an absorption edge around 575 nm which is blue shifted as compared to bulk CdSe (712 nm) due to quantum confinement effect. The intensity of absorbance for CdSe NC increases with Ag doping. For Ag:CdSe NC, a small broad hump around 406 nm appears. This peak at 406 nm is the characteristic of Surface Plasmon Resonance (SPR) [38].

The increase in absorbance intensity is due to the excitation of localized surface plasmon with the incident light. PL emission spectra was recorded at room temperature at an excitation wavelength of 380 nm. The photoemission peak for CdSe NC at 585 nm which is due to excitonic emission of CdSe nanoparticles [39] giving the evidence for successful synthesis of aqueous CdSe NPs. The photoemission peak has been found to be shifted to 589 nm for Ag:CdSe NC. The quenching of PL emission has been observed with Ag doping that may be due to the Ag ions which cover the surface of luminescent CdSe nanoparticles [40]. X-ray Diffraction (XRD), Transmission Electron Microscopy (TEM) and Fourier Transform Infrared Spectroscopy (FTIR) has also been conducted but these results are shown somewhere else [41].

The performance of the CdSe and Ag doped CdSe NCs has been studied for memory device application. To investigate the charge storage effect of CdSe nanoparticles (NPs), the I - V measurements of a device (Al/PVA/Ag) without the CdSe NPs were performed. Fig. 3 shows the I - V characteristics of Al/PVA/Ag device.

Fig. 4 shows the semi-logarithmic plot of I - V characteristics for set 'A', Al/CdSe NC/Ag and Al/Ag:CdSe NC/Ag devices respectively. Whereas, the I - V characteristics are shown in the insets of Fig. 4. The voltage scan of the devices is performed from 0 V to 4 V, 4 V

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