

Highly-stable memristive devices based on poly(methylmethacrylate): CsPbCl₃ perovskite quantum dot hybrid nanocomposites



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ABSTRACT

Memristive devices based on cesium-lead-chlorine (CsPbCl₃) perovskite quantum dots (PQDs) embedded in a poly(methyl methacrylate) (PMMA) layer were fabricated to enhance their memory stability. Current-voltage (*I*-*V*) measurements on Al/PQDs embedded in PMMA layer/indium-tin-oxide devices at 300 K showed nonvolatile rewritable memristive behaviors. The maximum ON/OFF ratio of the current bistability for the devices was as large as 2×10^4 . The retention time for the devices was above 1×10^4 s, indicative of high stability of the device. The operating mechanisms of the devices related to the interaction between the PQDs and the PMMA matrix could be explained by adjusting the thickness of the active layer. Furthermore, the carrier transport mechanisms of the devices are described on the basis of the *I*-*V* results with the aid of the energy-band diagram. The carrier transport could be attributed to the space-charge-limited-current mechanism in the high resistance state and to Ohmic conduction in the low resistance state.

1. Introduction

Nonvolatile memristive devices based on hybrid nanocomposites are currently the subjects of considerable attention due to their superior advantages of low cost, simple fabrication process, and low power consumption, as well as their potential applications in memory devices, artificial learning, and logic computing [1–3]. Among the several types of nonvolatile memristive devices, hybrid inorganic/organic nanocomposites containing low-dimensional nanomaterials embedded in a polymer matrix have been particularly attractive because of interest in promising applications in ultra-high density and flexible nonvolatile memory devices [4–7]. Typically, the inorganic/organic nanocomposites for memory devices can be processed by direct mixing both low-dimensional nanomaterials and matrix materials. The low-dimensional nanomaterials, acting as charge-storage sites, play a key role in the performance of memristive devices. Metallic nanoparticles, carbon-containing nanomaterials, and semiconductor quantum dots (QDs) have been extensively used as charge-trapping materials [8–11].

Recently, perovskite materials have been successfully made into memristive devices, which are expected to show great potential in promising applications of next-generation memory [12–16]. Especially, perovskite QDs (PQDs) have a wide range of applications in the fields of solar cells, light-emitting devices, and optical biomarkers [17–22]. Some works concerning the electrical properties and the memory effects of memristive devices fabricated utilizing organic-inorganic hybrid

PQDs or perovskite nanoparticles have been performed [21–23]. However, the stabilities of organic-inorganic hybrid perovskite materials have always been questionable, which is the main obstacle significantly hindering reliable device operation and potential future applications [24–26]. Even though all-inorganic PQDs are thought to hold promise as a new class of materials for memristive devices, very few investigations on the operating mechanisms and the carrier transport mechanisms in memristive devices fabricated utilizing all-inorganic PQDs blended with an insulating polymer have been reported. Furthermore, studies on the carrier transport mechanisms for memristive devices based on all-inorganic PQDs are necessary since the enhanced efficiencies of such devices are expected to appear.

This paper reports data for the electrical bistabilities of and the carrier transport mechanisms in memristive devices fabricated utilizing CsPbCl₃ QDs blended with a poly(methyl methacrylate) (PMMA) polymer formed by using a simple spin-coating technique. PMMA, which has been widely used in memristive devices, was chosen as the matrix material due to its remarkable dielectric and insulator properties [27,28]. Transmission electron microscopy (TEM) measurements were performed to investigate the microstructural properties of the PQDs. Field-emission scanning electron microscopy (FE-SEM) measurements were performed to investigate the dispersion of the PQDs in the PMMA layer. Current-voltage (*I*-*V*) measurements and data fittings of the *I*-*V* curves were carried out to investigate the memristive properties and the carrier transport mechanisms in the devices with a structure of Al

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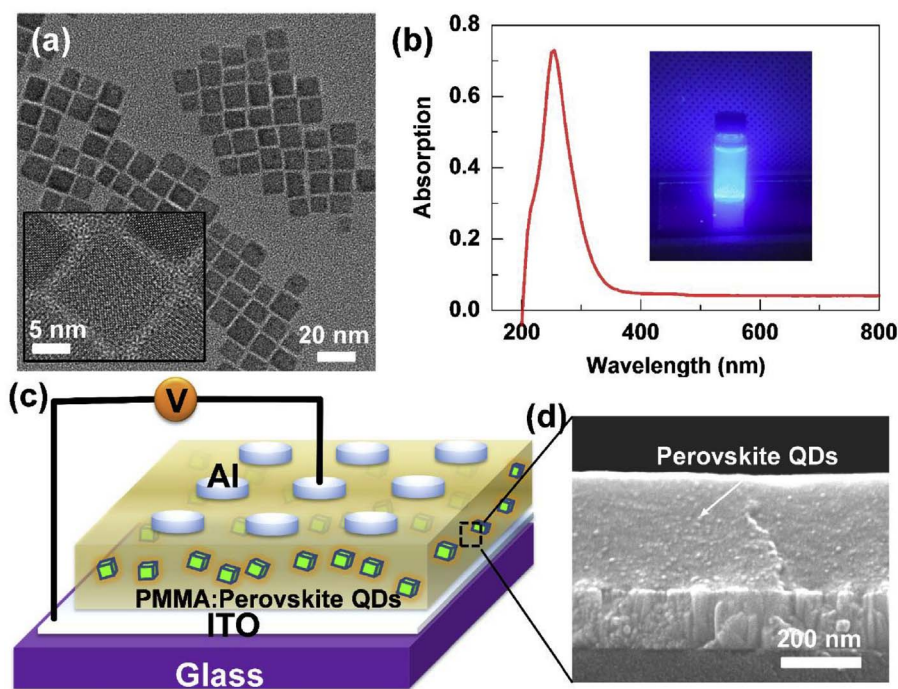


Fig. 1. (a) Transmission electron microscopy image of the PQDs. Inset represents the high-resolution TEM image of one typical PQD. (b) Ultraviolet–visible–infrared absorption spectrum of the PQDs solution. Inset represents a photograph of the PQDs solution taken under a 365 nm UV light. (c) Schematic diagram of the Al/PQDs:PMMA/ITO memristive device. (d) Cross-sectional scanning electron microscopy image of the PQDs:PMMA nanocomposite film deposited on the indium-tin-oxide electrode.

electrode/CsPbCl₃ QDs embedded in an insulating PMMA polymer layer/indium-tin-oxide (ITO) electrode. The thickness of the active layer and the mass fraction of the PQDs in the active layer were adjusted to investigate the operating mechanisms. The possible carrier transport modes of the memristive devices were described by using several models to fit the experimental I–V data [29–33]. These observations provide important information for further exploration of PQD-based, flexible memories.

2. Experiment details

TEM measurements were carried out with the diluted colloidal solution of PQDs in order to obtain nanostructural formation on the PQDs. Fig. 1(a) shows a typical distribution of the PQDs. The QDs are seen to have a cubic shape with a typical size of about 12 nm. A HRTEM image showed the presence of clear lattice fringes, as shown in the inset of Fig. 1(a). Fig. 1(b) presents an ultraviolet (UV)-visible-infrared absorption spectrum of the PQD solution. The PQDs are of high purity because only a single absorption peak at a wavelength of 300 nm is observed in the absorption spectrum. The inset of Fig. 1(b) presents a photograph of the PQD solution taken under 365-nm UV light.

Fig. 1(c) shows a schematic diagram of the fabricated memory devices with an Al/PQDs:PMMA/ITO structure, i.e., a PMMA active layer containing PQDs and separating the Al and the ITO electrodes. The colloidal PQDs (purchased from PlasmOChem GmbH, AA151221-1) were dissolved in chloroform at a concentration of 10 mg/ml, followed by magnetic stirring for 4 h at room temperature. The PMMA (purchased from Aldrich Co., 1000969280) was dissolved in chloroform at a concentration of 10 mg/ml, followed by magnetic stirring for 4 h at room temperature. PQDs:PMMA solutions with PQD concentrations of 1.25, 2.44, and 4.76 wt% were prepared by mixing the as-prepared PQD solution and the PMMA solution. Then, the mixed solutions were magnetically stirred for 4 h at room temperature. After the PQDs:PMMA solution had been deposited on the ITO film by using a spin-coating technique, an Al electrode with a thickness of about 150 nm was thermally evaporated on the PQDs:PMMA composite through a metal mask at a system pressure of 1×10^{-6} Torr. The thickness of the active layer was adjusted by changing the rotating speed during the spin-coating process and the duration of that process. The electrical properties of the

devices were measured by using a Keithley 2400 Digital Source Meter. A bias voltage was applied to the top Al electrodes during all measurements, with the bottom ITO electrodes being grounded. Fig. 1(d) shows a cross-sectional FE-SEM image of a PQDs:PMMA nanocomposite formed on an ITO-coated substrate. The PQDs can be clearly identified, as shown by an arrow, and are seen to have been uniformly dispersed in the PMMA matrix during the spin-coating process.

3. Results and discussion

The device with a PQD mass fraction of 2.44 wt% showed the best electrical properties with the largest ON/OFF ratio because the PQDs, acting as charge-storage sites, play a key role in the performance of memristive devices. The nanocomposites with very much higher or lower PQD concentrations do not show satisfactory memristive behaviors. Thus, only the electrical behaviors for the memristive devices based on PQDs:PMMA active layers with an optimized PQD mass fraction of 2.44 wt% are discussed; the I–V data are shown in Fig. 2.

The voltages across the devices were varied a cycle from -3.5 to 0 to 3.5 to 0 to -3.5 V. The I–V curves for the device with a 150 nm-thick active layer clearly show counterclockwise electrical hysteresis behaviors, which are essential features of bistable devices. State “1” and state “0” correspond to the relatively low resistance state (LRS) and the relatively high resistance state (HRS), respectively. The device transits from the LRS to the HRS at an applied voltage of 2.6 V. The bistable transition from the LRS to the HRS is equivalent to the “Reset” process in a digital memory cell. Then, the device maintains the HRS after the transition is completed. The LRS can be achieved by applying a small reverse bias voltage of -300 mV. This is equivalent to the “Set” process of the digital memory cell. The maximum current ratio between the LRS and the HRS for the Al/PQDs:PMMA/ITO device at 1 V is as large as about 2×10^4 , which is equivalent to the “Reading” process in a digital memory cell.

Fig. 3(a) shows the endurance characteristics of the device. The cycling frequency in the measurement was about 0.3 Hz. No obvious deterioration was observed during the measurements. The retention characteristic at LRS and HRS obtained for the device is presented in Fig. 3(b). The different conduction states remain until the retention time of 10×10^4 s at ambient conditions and the clearly distinguished

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