Demonstration of Addressable Organic Resistive Memory Utilizing a PC-Interface Memory Cell Tester

Byungjin Cho, Sunghoon Song, Yongsung Ji, Ho-Gil Choi, Heung Cho Ko, Jae-Suk Lee, Gun-Young Jung, and Takhee Lee

***Abstract*—We demonstrated nonvolatile 8 × 8 array organic memory devices utilizing a PC-interface memory cell tester. The organic memory devices composed of a Ag/poly(3-hexylthiophene-2,5-diyl) (P3HT)/p^+ poly-Si structure exhibited excellent memory performance properties, including stable switching behavior, proper statistical distribution, and long retention time. We succeeded in independently addressing and reading the data in the memory cell array using the PC-interface memory cell tester, opening an avenue toward more realistic organic memory device applications.**

***Index Terms*—Nonvolatile memory, organic memory, PC interface, resistive switching.**

I. Introduction

ORGANIC resistive memory devices have been considered as potential candidates for next-generation information storage because they are simple [1], flexible [2], and stackable [3]. In addition, they have outstanding electrical properties, such as a nondestructive reading process, a nonvolatile memory effect, a high ON/OFF ratio, and a fast switching speed, all of which meet the requirements for viable memory technologies [4], [5]. It is important to verify whether individual cells within a device have large ON/OFF differences and similar programming voltage values, and then, it is likely to correctly address and store data in all operating memory cells. Recently, Lin *et al.* have demonstrated PC-interfaced organic memory devices, which is considered to be a technologically advanced step toward the realization of practical organic memory devices [6]. The PC-interface demonstration requires a complex external system that includes addressing and sensing circuits in addition to reliable internal organic memory devices.

In this letter, we demonstrate nonvolatile 8 × 8 array organic memory devices utilizing a PC-interface memory cell tester. The tested organic memory devices comprised of a Ag/poly(3-hexylthiophene-2,5-diyl) (P3HT)/p^+ poly-Si structure exhibited reasonable memory requirement, including stable switching behavior, proper statistical distribution, and long retention time. With the assistance of a PC-based memory cell tester that interfaces crossbar-array organic memory devices and includes electronic circuitry for addressing and sensing, we were able to program and read the data in the memory array cells. Our demonstration may foster the potential availability of practical memory applications using organic memory devices. Our memory devices with a homogeneous single-layer polymer and a high ON/OFF ratio can be a promising candidate for the PC-interface cell demonstration. In addition, the cost is cheaper than other types of polymer memory devices. Potentially, junction size would be scalable to tens of nanometer scale, which is due to an operation mechanism based on nanosized Ag filament paths. Although p^+ poly-Si itself is too hard for bending applications, alternatively, if the device is placed on a neutral strain plane by sandwiching between a plastic substrate and an encapsulation layer, flexibility may be implemented in our organic memory devices.

II. Experimental Section

The 8 × 8 array organic memory devices were fabricated on a SiO_2/Si substrate. First, to make the bottom electrodes, a heavily doped p^+ poly-Si was patterned into eight lines with a 100-μm width using conventional lithography and a dry etching process. For the active layer, P3HT (purchased from Sigma-Aldrich Company) was spin coated on the patterned bottom electrodes. Finally, 100-μm-thick Ag top electrodes were deposited through electron beam evaporation. The final 8 × 8 crossbar array structure of the Ag/P3HT/p^+ poly-Si was completed. All the electrical measurements of the memory devices were conducted under an ambient atmosphere utilizing a semiconductor parameter analyzer (Agilent 4155C) and a PC-interface memory cell tester.

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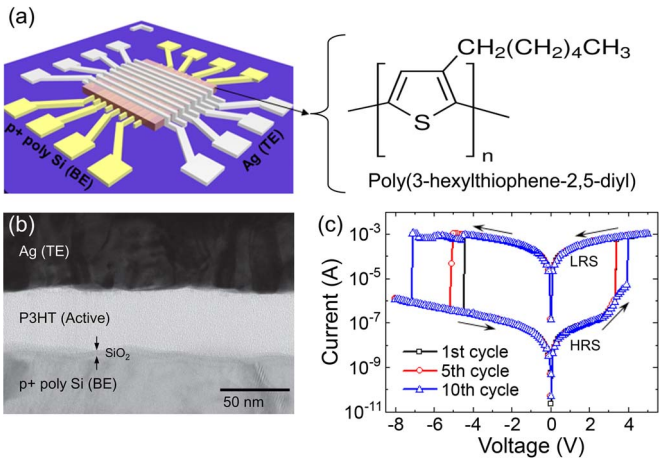

Fig. 1. (a) Schematic of P3HT-based organic memory devices with an 8 × 8 array cell structure. (b) TEM image showing each layer of Ag/P3HT/p^+ poly-Si. (c) $I-V$ characteristics of an organic memory device.

III. Results and Discussion

Fig. 1(a) shows a schematic of the organic memory device with an 8 × 8 array of cells and a chemical structure of the P3HT polymer (the active layer). The device is composed of a Ag/P3HT/p^+ poly-Si structure, as shown in the cross-sectional transmission electron microscopy (TEM) image [see Fig. 1(b)]. The thickness of the active polymer layer was measured to be ∼50 nm. The bottom electrodes (p^+ poly-Si) were treated with O_2 plasma to enhance the ON/OFF ratio. This was due to the increase in thickness in an interfacial silicon oxide layer that determined the initial resistance value of the memory device. The representative current–voltage (I–V) behavior of the organic memory devices exhibited typical bipolar switching, as shown in Fig. 1(c). A variable voltage source was applied to the top electrodes (Ag) while the bottom electrodes (p^+ poly-Si) were grounded. In the first voltage sweep from 0 to 4.5 V, the current gradually increased in the low-voltage region and then abruptly increased at a higher voltage of ∼4 V, indicating a set process from a high-resistance state (HRS) to a low-resistance state (LRS). When the voltage was swept back to 0 V, the LRS was well maintained, and this behavior is indicative of nonvolatile memory. In the second sweep toward the negative polarity, the device recovered to the original HRS, thereby showing a reset process from the LRS to the HRS. A high ON/OFF ratio greater than 10^3 and stable I–V curves were obtained in our organic memory devices. The origin of the resistance change is associated with high conductive filament paths, resulting from the diffusion of the top metal into the polymer layer [7]–[9]. Particularly, our memory system is based on a creation-and-rupture process of redox-controlled Ag bridges [7]. Specifically, a positive voltage applied to the top electrode (Ag) leads to the oxidation of Ag atoms, as described by $Ag \rightarrow Ag^+ + e^-$. The resulting Ag^+ cations diffuse through the organic layer, and then, Ag is precipitated near the bottom electrodes (p^+ poly-Si) through a reduction mechanism ($Ag^+ + e^- \rightarrow Ag$). With the successive precipitation of Ag, metallic bridges connecting the two electrodes are formed. Conversely, under a negative voltage, electrochemical dissolution occurs along the metallic bridge, resulting in the rupture of the Ag metallic bridges. We

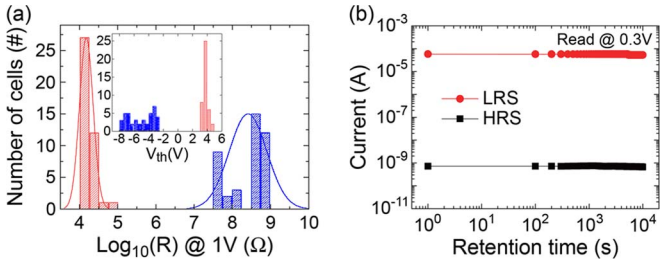

Fig. 2. (a) Histogram of ON and OFF resistance values for P3HT-based organic memory devices. (Inset) Histogram of set and reset threshold voltages. (b) Retention time of the organic memory device.

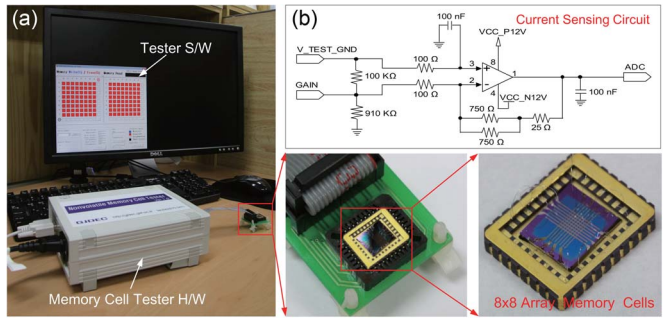

Fig. 3. (a) Optical image of the PC-interface memory cell tester used to independently address the data in the memory cells. At right, two enlarged images show that the organic devices are connected to a package embedded in a socket on a plastic IC chip. (b) Current-sensing circuit to read the resistance state of each memory cell.

also found a strong proportional relationship between the set and rest currents to support the filamentary conduction mechanism [10]. Therefore, it was reported that the P3HT active polymer with a strongly coordinating heteroatom (S or N) can support the reproducible filament formation behavior [8].

It is important to statistically evaluate the memory performance parameters of the operating cells. We examined statistical distribution of ON and OFF resistance values and threshold voltages extracted from I–V curves of operating cells. The ON and OFF resistance values were well separated each other with an interval over 10^2, as shown in Fig. 2(a). The inset in Fig. 2(a) shows the threshold voltage distribution. The set threshold voltages were distributed between 3 and 5 V, whereas the reset threshold voltages exhibited relatively wider distribution from −2.5 to −8 V. The devices maintained a high ON/OFF ratio without serious current fluctuation over a tested retention time of 10^4 s [see Fig. 2(b)].

By utilizing the PC-interface memory cell tester hardware and software, we could easily address and read the memory cells of the packaged devices embedded in a socket on an integrated circuit chip [see Fig. 3(a)]. The resistance of the selected memory cell was measured from the current-sensing circuit, as illustrated in Fig. 3(b). More specifically, the logic states of "0" and "1" were determined by the threshold resistance value of 1 MΩ, i.e., if the resistance value of the cell ranged from 700 Ω to 1 MΩ, it was defined as "1"; if the resistance value ranged from 1 to 90 MΩ, it was defined as "0". In cases when the measured resistance was less than 700 Ω or larger than 90 MΩ, it was considered to be a short or a disconnection, respectively.

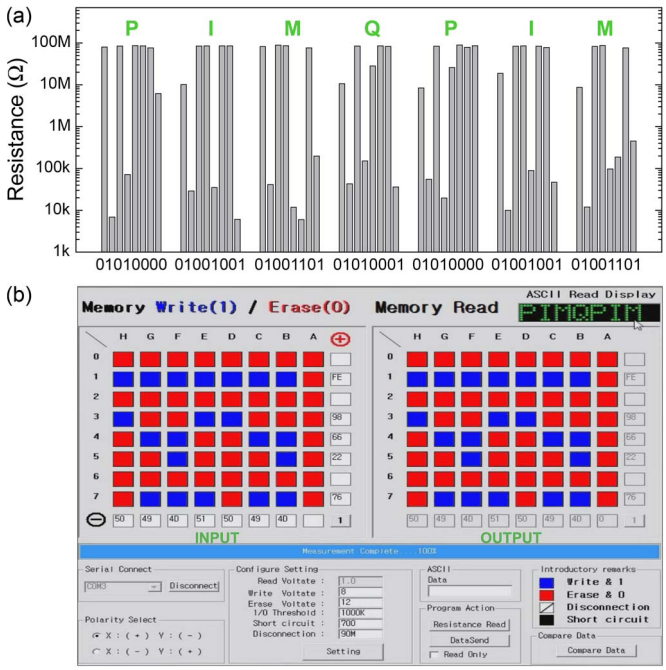

Fig. 4. (a) Logic states of "PIMQPIM" data programmed by the PC-interface memory cell tester. (b) Final image of the demonstration process to address the data into each memory cell with the PC-interface memory cell tester software, showing that the output data are well matched with the input data.

A byte is composed of a column line (8 bits). For example, the letter corresponding to "01010000" is "P." In fact, we intended to store 8-B data of double "PIMS" (abbreviation of "Program for Integrated Molecular Systems"), but unfortunately, all of the memory cells in the seventh row and the eighth column did not correctly operate due to device failure. Instead, 7-B data representing "PIMQPIM" were programmed in our demonstration, as shown in Fig. 4(a). Note that "Q" was displayed instead of "S" due to the failure of the seventh row, and the last letter was not written due to the failure of the eighth column. Such a programming failure seems to come from the fabrication problem of some electrode lines if considering that all the cells in the eighth column and the seventh row line did not work. The yield of memory cells can be improved by etching p^+ poly-Si uniformly and controlling Ag deposition rate carefully. Nevertheless, our PC demonstration of organic memory devices can be considered as a meaningful step toward the application of a practical memory device.

Each cell was sequentially programmed, and the final image of the demonstration (to show the logic states programmed by the tester software) is displayed in Fig. 4(b). In this figure, the red and blue cells represent "0" and "1," respectively. The input data (writing, shown at left) were perfectly consistent with the output data (reading, shown at right). The system visualized letters corresponding to the ASCII codes, as shown in the image in the upper-right corner of Fig. 4(b). The erasing capability of the memory cells could be also demonstrated by utilizing the PC-interface memory cell tester (data not shown here).

IV. Conclusion

In conclusion, we have fabricated 8 × 8 crossbar-array organic memory devices with a Ag/P3HT/p^+ poly-Si structure and demonstrated addressable nonvolatile memory operations utilizing a PC-interface memory cell tester. This operation was possible due to excellent memory performance, including stable switching behavior, good statistical distribution, and long retention time. We succeeded in independently addressing and reading the data in the memory cell array using the PC interface and displaying alphabetical letters on a computer screen. Our study demonstrates a promise toward more realistic organic memory device applications.

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