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Molecular Memory Nano-interfaced with Organic Molecules

Hyoyoung Lee[1], Mi-Hee Jung[2]

[1] Sungkyunkwan University
Center for Smart Molecular Memory, Department of Chemistry,
Cheoncheon-dong, Jangan-gu, Suwon 440-746, Republic of Korea
Phone: +82-31-299-4566 E-mail: hyoyoung@skku.edu
[2] Electronics and Telecommunications Research Institute
Thin Film Solar Cell Technology Research Team, Advanced Solar Technology Research Department,
161 Gajeong-dong, Yuseong-gu, Daejeon 305-350, Republic of Korea

1. Introduction

The organic non-volatile memory device has attracted a great deal of attention given its potential application in low cost, flexible, and large area electronics. Among organic memory devices, a memory based on the organic field effect transistor (OFET) is especially attractive because of its nondestructive readout and single transistor applications. However, the organic memory devices still do not sufficiently satisfy the criteria demanded in order to compete with other types of memory devices with the electrets being generally limited to polymer materials.

Until now, there has been no report on nonvolatile organic electrets using a nano-interfaced organic monomer layer as semiconductor materials, even though the use of organic monomer materials has become important for the development of molecularly interfaced memory and logic elements. Furthermore, to increase the retention time of nonvolatile organic memory devices, as well understand the intrinsic memory properties, molecular design of the organic materials is becoming an important issue.

Here, to demonstrate the concept of organic nonvolatile memory devices, we report an OFET memory device built on a silicon wafer and based on films of pentacene and a SiO_2 gate insulator that are separated by organic molecules acting as a gate dielectric.

We also propose novel push-pull organic molecules (PPOMs) containing triarylamine as an electron-donating group (EDG), thiophene as a spacer, and malononitrile as an electron-withdrawing group (EWG). The PPOMs were designed for charge transport control through differences of the dihedral angles induced by a steric hindrance effect of the side chains within the molecules, producing hysteresis and finally leading to the memory OFET device. It is expected that the higher dihedral angle within molecule gives the larger hysteresis.

2. Results and discussion

The new PPOMs that have benn submitted elsewhere were composed of triarylamine as an EDG, thiophene as a spacer, and malononitrile as an EWG in the molecular backbone. Herein, two types of molecules: R = H (2-((5-(4-(naphthalen-1-yl(phenyl)amino)phenyl)thiophen-2-yl)methylene)malononitrile), QH: R = CH_3 (2-((4-methyl-5-(4-(naphthalen-1-yl(phenyl)amino)phenyl)thiophen-2-yl)methylene)malononitrile), QMe.

As mentioned earlier, the thiophene spacer was designed for controlling the charge transport properties from triarylamine to malononitrile and vice versa. The charge transport would be affected by the dihedral angles derived from the steric hindrance of the side chains of the thiophene spacer as shown Fig. 1a.

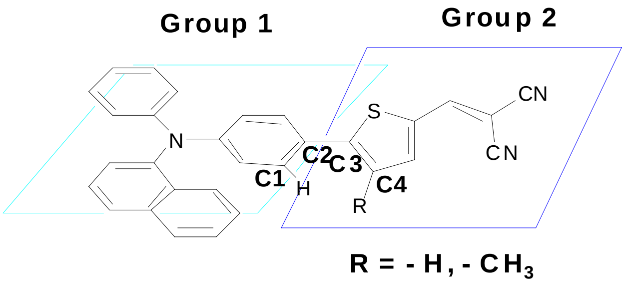

Fig. 1 Molecular structure of the PPOMs (QH and QMe)

For a realization of the OFET memory device as shown in Fig. 2, top contact OFET with the PPOMs was constructed on the heavily n-doped Si substrates with a 300 nm SiO_2 layer. Poly(methymethacrylate) (PMMA) was used as the dielectric layer. PMMA is a hydroxyl-free polymer that decreases the number of electron traps, providing the gate dielectric interface buffered to the organic molecules.[1] Gold electrodes, as source and drain contacts, were deposited onto the organic layer through the shadow mask. The channel length and width were 100 and 10,000 μm, respectively.

Fig. 3 shows transfer characteristics of the OFET devices. The dielectric pristine SiO_2 and PMMA polymer layers[2] used as control experiments herein have been shown to retain injected charge. In our results, the pristine OFET with a SiO_2 layer, as depicted in Fig. 3a, or PMMA, as shown in Figure 3b, shows only weak hysteresis with a memory window (ΔV_T) less than 5 V. Amazingly, the transfer characteristics of the PPOMs-FET devices show that there is a clearly visible hysteresis between the two source-drain currents for gate voltage sweep in the negative and positive directions shown in Fig. 3c and 3d. In the transfer characteristics of the OFET with QH (OFET-QH device) and QMe (OFET-QMe device), the memory window of the OFET-QMe was wider than that of the OFET-QH and as expected, the drain current of the OFET-QMe decreased more than that of the OFET-QH. For the OFET-QMe, the ON/OFF ratio was about 10^2 (two orders of magnitude difference in current between the high conductance ON state and low conductance OFF state). The "ON" current was 30 nA at a gate voltage of -50 V. The "OFF" current appeared to be 0.6 nA at the gate voltage of 50 V.

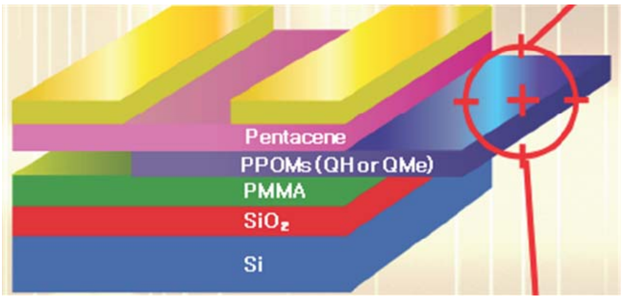

Fig 2 Schematic of the OFET memory device

For the possible mechanism, the "ON"-"OFF" switching phenomenon was achieved by the conversion of the "anion" and "neutral" states. When a gate voltage is applied to the neutral QH and QMe, they undergo a gradual conformational change from the neutral ("OFF" state) to the anion ("ON" state) species. Then, when the gate voltage is turned-off, the anion species ("ON" state) of the QH and QMe gradually changes to the neutral ("OFF" state) species. The difference in the QH and QMe is the response time as QH changes more quickly and the operation voltage range of QMe is wider than QH.

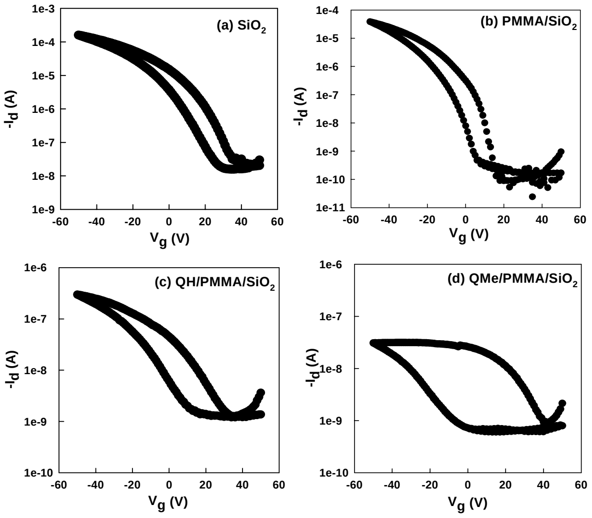

Fig. 3 Transfer characteristics of the OFET demonstrating the hysteresis with PPOMs (QH and QMe).

In partial summary, the larger tilted structure caused by the steric hindrance gives higher charge trapping and reduces the charge mobility more and finally produces higher hysteresis. Thus, the memory window of QMe is larger than that of QH.

4. Conclusion

The observed hysteresis effect occurred due to a charge trapping on the conduction channel comprising PPOMs and charge de-trapping with high potential energy barrier caused by the steric hindrance effect of the nano-interfaced organic semiconductor, PPOMs. The hysteresis window of the OFET-QMe was wider than that of the OFET-QH.

Acknowledgement

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