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A Solution-Processable Donor–Acceptor Compound Containing Boron(III) Centers for Small-Molecule-Based High-Performance Ternary Electronic Memory Devices[**]

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Abstract: *A novel small-molecule boron(III)-containing donor–acceptor compound has been synthesized and employed in the fabrication of solution-processable electronic resistive memory devices. High ternary memory performances with low turn-on ($V_{Th1} = 2.0$ V) and distinct threshold voltages ($V_{Th2} = 3.3$ V), small reading bias (1.0 V), and long retention time ($> 10^4$ seconds) with a large ON/OFF ratio of each state (current ratio of "OFF", "ON1", and "ON2" $= 1:10^3:10^6$) have been demonstrated, suggestive of its potential application in high-density data storage. The present design strategy provides new insight in the future design of memory devices with multi-level transition states.*

In the past decades, resistive memory devices have drawn a lot of attention in the field of data storage[1] because of the limitation presented by existing materials and technologies in the semiconductor industry.[2] Unlike traditional silicon-based memory devices which are based on the amount of charges stored in the devices, the memory effect in resistive memory devices is based on the electrical bistability of conductance/resistance, where a low-conductance/high-resistance (OFF) state switches to a high-conductance/low-resistance (ON) state. Typical resistive memory devices can be fabricated by forming a composite active layer and switch with two electrodes. Metal oxide has been widely used as materials for the active layer,[1] while there has been a growing interest in the use of organic-based materials[3] as active layer because of the unique flexibility, light weight, good processability, large scalability, and data storage capacity, and low-cost potential of organic compounds.[4] To achieve electrical bistability in organic systems, donor–acceptor couples[3b,5] have to be employed and they have been demonstrated as one of the successful approaches involving electric-field-induced charge transfer in conjugated compounds. By varying the donor and acceptor strengths and the selection of different donors and acceptors, it is suggested that the memory effect can be modulated.[4a,5]

On the other hand, the memory states in most data-storage systems consist of "0" and "1" output signals, representative of the conventional binary logic states. In such systems, the overall data storage capacity scales as 2^n. To enhance the data storage capacity, there has been continuous effort on the increase in the number of n by reducing the bit cell size.[6] However, the bit cell size will eventually reach its limits due to difficulties in fabrication and data processing in nanoscale.[7] Besides that, the data storage capacity can be dramatically boosted by increasing the number of memory states from binary (2^n) to ternary (3^n). For semiconductor-based memory devices, this kind of multistate memory behavior has been rarely reported in the literature,[8] because of the difficulties in reliably controlling multiple states. On the contrary, multiple electroactive states can be facilely designed and engineered by controlling the donor–acceptor couple in organic materials. To achieve organic ternary memory devices, Lu and coworkers reported the use of "one donor–two acceptors" in thermally evaporable small molecules,[9] while Huang and coworkers achieved a solution-processable ternary device from the combination of two independent moieties with electrical bistabilty by using diblock copolymers.[10] However, there is still a lack of research and understanding for the development of new materials and concepts for solution-processable small-molecule based ternary memory devices.

There has been an increasing interest in boron(III)-based materials owing to their potential for optoelectronic and anion-sensing applications.[11] In particular, boron(III) diketonates are known to exhibit rich photophysical properties.[12] In spite of the multi-disciplinary functions in boron(III) diketonates,[12–14] the development of this class of compounds as active materials in organic memory devices has not yet been reported. In addition to our continuing interests in designing various functionalized boron(III)-based materials[15] and the recent investigation in organic memory devices,[16] we hypothesized that by utilizing the electron-accepting and electron-transporting ability of boron(III) diketonates,[17] organic memory devices with multi-level states could be readily achieved through rational design. Unlike the previous reported methods,[9,10] our design strategy is to append the boron(III) diketonates to the dithienyl substituted benzothiadiazole core, which is an electron acceptor and donor, respectively.[18] It is envisaged that

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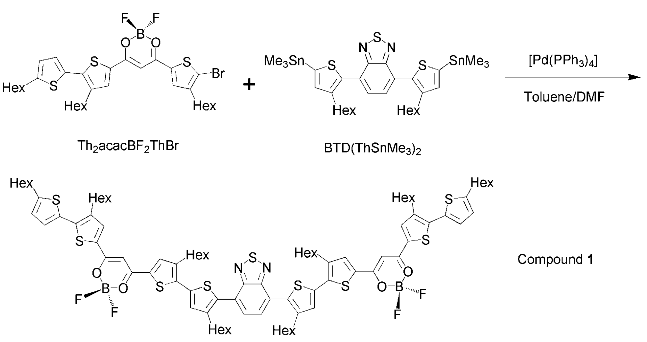
charge separation is induced upon increasing the applied bias. Subsequent electron transfer may occur from the benzothiadiazole core to the boron(III) diketonates through intramolecular and/or intermolecular electron transfer and may result in the formation of two independent electron trapping states, which may open up a new class of materials and concepts to achieve memory devices with ternary logic state. In order to test the hypothesis, herein we describe the design and synthesis of a new class of donor–acceptor compound containing boron(III) diketonates and the study of their ternary memory devices.

The target compound is prepared as shown in Scheme 1. Compound **1** can be facilely synthesized by bis-Stille cross-coupling between $Th_2acacBF_2ThBr$,[19a] which is prepared by modified literature methods, and $BTD(ThSnMe_3)_2$[19b] by using standard anaerobic reaction conditions. Air-stable compound **1** has been characterized by ^{1}H, ^{11}B{^{1}H} and ^{19}F{^{1}H} NMR spectroscopy, MALDI-TOF high-resolution mass spectrometry, and elemental analysis.



Scheme 1. Synthetic pathway for compound **1**.

In order to explore the potential of the compound in organic memories, a memory device has been fabricated, in which compound **1** has been sandwiched between indium–tin-oxide (ITO) bottom electrode and an aluminum top electrode, as shown in Figure 1 a. The film thickness of compound **1** and aluminum is found to be about 123 and 93 nm, respectively, from the SEM image of a cross section of the device, as depicted in Figure 1 b. The surface morphology of the thin film of compound **1** has been studied by atomic force microscopy (AFM). Figure 1 c shows the tapping-mode AFM topography and the corresponding cross-section profile of the AFM topographic image. The root-mean-square (RMS) roughness is found to be about ± 2 nm, suggestive of a smooth surface morphology. Such smooth surface is believed to benefit the charge injection from the electron to the film, leading to a lower threshold voltage.[4c] The X-ray diffraction patterns of compound **1** in powder and thin-film forms are shown in Figure S1 in the Supporting Information. The powder form of compound **1** exhibits diffraction peaks at $2\Theta = 5.8$, 6.4, and 24.5°, which correspond to d spacings of 15.24, 13.81, and 3.63 Å, respectively. On the other hand, a sharp peak at 5.2° and a broad signal from 16.5 to 29.1° with the maximum at 21.1°, which correspond to d spacings of 17.00 and 4.21 Å, respectively, are observed in neat film. The d spacing of the broad signal is from 5.37 to 3.07 Å, suggestive of a pronounced intermolecular π-π stacking in the thin-film sample, and such strong π-π interactions are believed to contribute towards the smooth surface of compound **1**, in which molecules with strong π-π interactions can facilitate better molecular packing and film-formation, resulting in smoother surface. Similar observation has been observed in the electronic absorption spectra of the solution and thin-film sample of compound **1**, as shown in Figure S2, in which the absorption band in thin film shows a bathochromic shift by 27 nm (866 cm^{-1}) when compared with that in solution with much broader absorption region, suggestive of the pronounced π-π stacking in thin films. It is also interesting to note that the lowest-energy absorption band of compound **1** is tentatively assigned as the mixing of π-π* transition of the benzothiadiazole ring system with some charge-transfer character from the benzothiadiazole moiety to the diketonate units. The optical bandgap is found to be 1.83 eV from the onset of the lowest-energy absorption band.

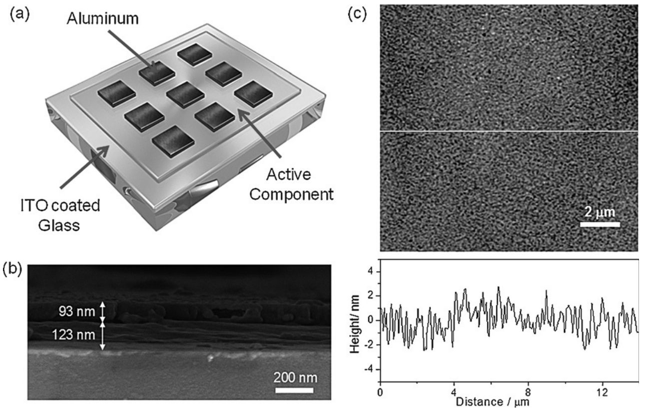


Figure 1. a) Schematic illustration of the ternary memory device. b) SEM image of a cross-section of the device. c) Tapping-mode (14×14 μm) AFM topography and the corresponding cross-section profile of AFM topographic image.

The memory behavior has been determined from the current-voltage performances of the as-fabricated device of Al/compound **1**/ITO, as shown in Figure 2 a. The first sweep of the as-fabricated device exhibits two abrupt increases in the current at the switching threshold voltages, namely 2.0 V (V_{Th1}) and 3.3 V (V_{Th2}), respectively. This process demonstrates the transition from a low-conductivity state, denoted as "OFF" state, to an intermediate-conductivity state, denoted as "ON1" state, and finally to a high-conductivity state, denoted as "ON2" state. The "ON1" state can be obtained in the second sweep of the other cells of the device over a voltage range of 0 to 2.8 V. The intermediate-conductivity state (ON1) remains during the third sweep and an abrupt increase in the current from "ON1" to "ON2" at about 3.2 V can also be observed. The high-conductivity state is maintained during the fourth sweep of the device over a voltage range of 0 to 5 V. The device shows good stability in the "ON2" state upon applying a reverse bias in the fifth sweep, suggestive of the "write-once-read-many" type of memory.[4] The three states exhibit a distinct current ratio of 1:10^{3}:10^{6} for the "OFF", "ON1" and "ON2" states, suggestive

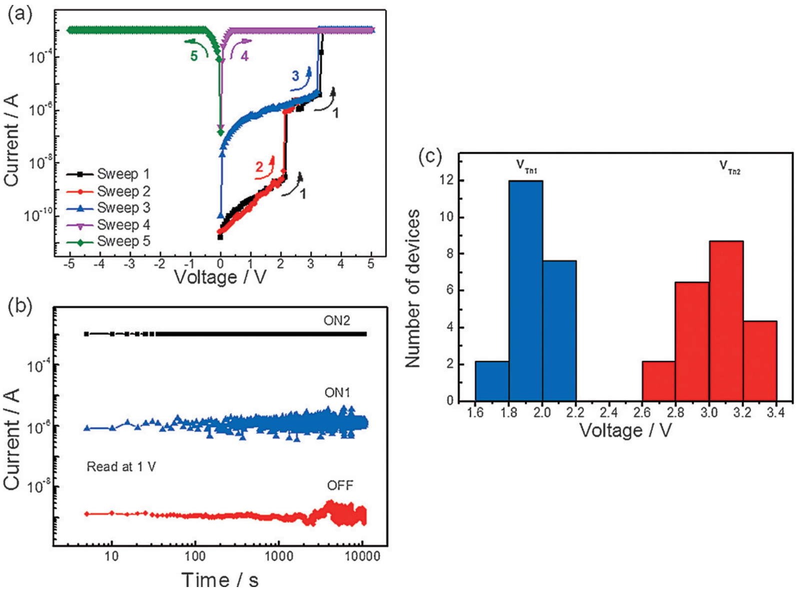


Figure 2. a) Current–voltage characteristics of an ITO/compound **1**/Al devices. b) Retention time of the ITO/compound **1**/Al devices in "OFF", "ON1" and "ON2" states under a constant stress (1.0 V). c) The bar chart of the number of devices against threshold voltage of "ON1" and "ON2" among 20 devices.

of a ternary memory device. The stability of the device under a constant stress is shown in Figure 2b. No significant degradation is observed in current for the "OFF", "ON1" and "ON2" states for at least 10^4 seconds during the measurement, suggesting that the device exhibits a good stability and a precise control of the three independent states with low misreading rate. Also, the patch-by-patch device performances have been investigated by the study of 20 individual devices, as shown in Figure 2c. The threshold voltage of "ON1" is found to range from 1.6 to 2.2 V, while that of "ON2" is from 2.6 to 3.4 V. About 1 V difference of writing voltage is observed between "ON1" and "ON2", suggestive of a good repeatability and a clear identification between the two states.

To understand the energy states of compound **1**, the electrochemical properties of compound **1** have been studied by cyclic voltammetry, as shown in Figure 3a. One quasi-reversible oxidation couple at +0.99 V ($\Delta E = 95$ mV) versus saturated calomel electrode (SCE), while two quasi-reversible reduction couples at −0.77 ($\Delta E = 66$ mV) and −1.26 V ($\Delta E = 70$ mV) versus SCE are observed. The first and second reduction couples are tentatively assigned as the reduction of the diketonates[15b,f] and the benzothiadiazole core,[18a] respectively. On the other hand, the first oxidation couple is assigned as dithienyl-substituted benzothiadiazole oxidation. The assignment has further been supported by the density functional theory calculation on the model compound **1′** (see the Supporting Information). As depicted in Figure 3b, the highest occupied molecular orbital (HOMO) is mainly the π orbital localized on the dithienyl benzothiadiazole. The lowest unoccupied molecular orbital (LUMO) and LUMO+1 are close in energy, which are the π* orbitals of the two aryl diketonates, while the LUMO+2 is the π* orbital localized on the benzothiadiazole core.

The energy level of the HOMO and the LUMO is found to be −5.33 and −3.59 eV, respectively, by using ferrocenium/ferrocene couple as internal reference. The energy bandgap was determined to be 1.74 eV, which is similar to that determined from the UV/Vis study. Also, the energy level estimated from the second reduction potential is found to be −3.12 eV. Based on the results obtained from cyclic voltammetry, the energy barriers for hole injection and electron injection are found to be 0.53 and 0.71 eV, which are determined by the energy difference between the work function of the ITO electrode (−4.8 eV) and the HOMO level of compound **1** and between the work function of Al electrode (−4.3 eV) and the LUMO level of compound **1**, respectively, as illustrated in Figure 3c. These results suggest that the hole injection from the electrode to the film of compound **1** is an energetically favorable process.

On the basis of our findings, the mechanism of field-induced conductivity is proposed to involve a stepwise charge transfer process. At a low voltage bias, low conductivity is observed because the energy is insufficient to overcome the injection barrier. When the applied bias reaches the first threshold voltage, the charge carrier can be injected from the dithienyl-substituted benzothiadiazole to the diketonates,

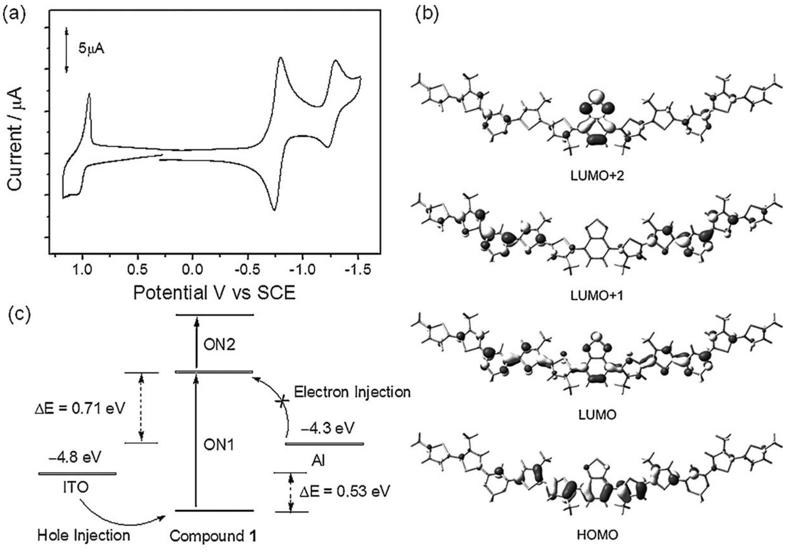


Figure 3. a) Cyclic voltammograms of compound **1** in dichloromethane (0.1 mol dm^{-3} $n\mathrm{Bu_4NPF_6}$). Ferrocenium/ferrocene couple ($\mathrm{FeCp_2}^{+/0}$) was used as internal reference for HOMO energy level (HOMO = −4.8 eV at half-wave potential). Scan rate = 100 mV s^{-1}. b) Spatial plots (isovalue = 0.03) of the selected molecular orbitals for **1′**. c) Schematic diagram of the charge injection process in the compound **1** based memory device.

which is supported by the fact that the threshold voltage of "ON1" is closely matched to the energy bandgap obtained from the electrochemical measurement. By further increasing the applied bias, the second charge transfer process from the diketonates to the benzothiadiazole is probably responsible for the occurrence of the "ON2" state, which results in the memory device with ternary logic (Figure 3c). In comparison to previous works,[9] the high performance ternary memory device properties are believed to be due to the high reversibility of the reduction couples in compound **1**, as revealed by cyclic voltammetry.

In conclusion, a novel approach and a new class of materials for small-molecule based solution-processable organic memory devices with ternary logic state have been demonstrated. The ternary memory effect is due to two independent electron trapping processes for the boron(III) diketonates and benzothiadiazole, in which the superior performance relative to those reported previously may be attributed to the high reversibility of the reduction couples in the boron(III) diketonate and benzothiadiazole moieties. The present design strategy provides a new insight in the future design of memory devices with multi-level transition states. In addition, a large variety of organic-based materials with different types of memory and tunable memory performances can be achieved by utilizing the present facile molecular design. Further investigations on the modulation of the memory response from the introduction of different aromatic systems into the boron(III) diketonates and the mechanistic studies on the memory effect are now in progress.

Keywords: boron · donor–acceptor systems · organic electronics · ternary memory devices

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