Push–Pull Archetype of Reduced Graphene Oxide Functionalized with Polyfluorene for Nonvolatile Rewritable Memory

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ABSTRACT: A solution-processable PFTPA-convalently grafted reduced graphene oxide (RGO-PFTPA) was synthesized by the 1,3-dipolar cycloaddition of azomethine ylide. Bistable electrical switching and nonvolatile rewritable memory effects were demonstrated in a sandwich structure of indium tin oxide/RGO-PFTPA/AI. The switch-on voltage of the as-fabricated device was around -1.4 V, and the ON/OFF-state current ratio was more than 10³. The ON-OFF transition process is reversible because the application of a high enough positive voltage can induce the reverse transfer of electrons, reducing the con-

INTRODUCTION Rather than encoding "0" and "1" as a charge stored in the cell of a silicon device, polymer memory stores data in an entirely different form, such as in a high- and low-conductivity response to an applied voltage.¹ As alternatives to the more elaborated processes of vacuum evaporation and deposition of inorganic and organic molecular materials, solution processes, including spin-coating, spraycoating, dip-coating, roller-coating, and ink-jet printing, can be used to deposit polymers on a variety of substrates such as plastics, inorganic wafers, glass, metal electrodes, and others.^{1(b)} The International Technology Roadmap for Semiconductors has identified polymer memory as an emerging memory technology since year 2005.²

When compared with silicon, graphene, which has a two-dimensional planar geometry that enables multilayer device architecture and simplifies future integration with complementary metal oxide semiconductor technology, exhibits superior electron mobility of up to 200,000 cm²/(V s), more than twice of that of the highest mobility conventional semiconductor.³ Additionally, the symmetry of the experimentally measured conductance indi-

ductivity back to its initial OFF state. Both the OFF and ON states are accessible and very stable under a constant voltage stress of -1 V for up to 3 h, or under a pulse voltage stress of -1 V for up to 10^8 continuous read cycles (pulse period = 2 μ s, pulse width = 1 μ s). © 2011 Wiley Periodicals, Inc. J Polym Sci Part A: Polym Chem 50: 378–387, 2012

KEYWORDS: conjugated polymers; functionalization of polymers; high performance polymers; polyfluorene; reduced graphene oxide; rewritable memory effect; synthesis

cates that the mobilities for holes and electrons should be nearly the same.⁴ Therefore, it would be very interesting to design and synthesize graphene-based solution-processed organic/polymeric materials for molecular optoelectronic and photonic devices. In our previous work, we designed a series of soluble donor-acceptor (D-A) type polymers for memory devices,⁵ among which graphene oxide (GO)-covalently functionalized polymer materials exhibit good bistable electrical switching and rewritable memory effects.^{5(g-i)} Some groups also reported the rewritable or, the write-once-read-many-times memory properties of polymer-grafted GO,⁶ and GO/reduced graphene oxide (RGO)doped polymer composites^{7–9} in recent years.

In contrast to graphene, its oxide form, GO, is an electrically insulating material owing of the disrupted sp² bonding networks, giving rise to a very low electron/hole transporting performance, which is unfavorable for construction of the optoelectronic devices such as organic light-emitting diodes (OLEDs), polymer light-emitting diodes (PLEDs), solar cells, and electroactive materials-based memory devices. On the other hand, electrical conductivity and electron/hole transporting

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properties of RGO can be recovered by restoring the π -network. Importantly, the preparation method for grafting organic or polymeric moieties onto the surface of RGO retained structural integrity of the RGO framework so that there is no loss of electronic structure. Up to the present moment, only fewer reports¹⁰ investigating covalently chemical modification of RGO.

It is well-known that the 1,3-dipolar cycloaddition reaction of azomethine ylides is one of the most versatile and widely applied methodologies for the functionalization of C₆₀.¹¹ Following this basic concept, the reaction between sarcosine, C_{60} , and the corresponding π -conjugated systems functionalized with one or two aldehyde groups, led to a variety of C_{60} -based full-fledged dyads and triads.^{5(a),11,12} The same approach can also be applied to the polymeric functionalization of RGO. In this contribution, we first synthesized a new polyfluorene (PF)-based conjugated polymer, namely poly{[4,4'-(4-(9-phenyl-9*H*-fluoren-9-yl)phenylazanediyl)diben zaldehyde]-[4,4'-(9H-fluorene-9,9-diyl)bis(N,N-diphenylbenze namine)]-(9,9-dihexyl-9H-fluorene)} (hereafter denoted as PFTPA-CHO), in which triphenylamine (TPA) acts as electron donor and hole transporting material,¹³ and then this polymer was directly used to react with RGO in the presence of an excess of N-methylglycine to produce a highly soluble RGO covalently functionalized with PFTPA (hereafter denoted as RGO-PFTPA, Scheme 1). The varieties, excellent optical and electronic properties, and high thermal and chemical stability of PFs make them an attractive class of materials for polymer-based optoelectronic devices. As a result, this polymer exhibited excellent nonvolatile rewritable memory effect, with a switch-on voltage of about -1.4V and an ON/OFF current ratio in excess of 10^3 .

EXPERIMENTAL

General

Infrared (IR) spectra were recorded on a Nicolet Nagma-IR 550 spectrophotometer using KBr pellets. Molecular weights [number-average (M_n) and weight-average (M_w)] were determined with a Waters 2690 gel permeation chromatography (GPC) using a linear polystyrene standards eluting with tetrahydrofuran. Nuclear magnetic resonance (NMR) spectra were recorded on a Bruker 400 spectrometer at a resonance frequency of 400 MHz for ¹H NMR in deuterated solution with a tetramethylsilane as a reference for the chemical shifts. Raman spectra were taken at room temperature with a MicroRaman System RM3000 spectrometer and an argon ion laser operating at a wavelength of 785 nm as the excitation source. X-ray photoelectron spectroscopy measurements were carried on a Kratos AXIS HSi spectrometer with a monochromatized Al KR X-ray source (1486.6 eV photons) at a constant dwell time of 100 ms and pass energy of 40 eV.

All chemicals were purchased from Aldrich and used without further purification. Organic solvents used in this study were purified, dried, and distilled under dry nitrogen. Purified natural graphite was purchased from Shanghai Yifan's Graphite Co. GO was prepared according to a method reported in our previous work.⁵⁽ⁱ⁾ RGO was obtained by reduction of GO by thermal annealing to 900 °C at a heating rate of 2 °C min⁻¹

and with a flow of 3% H₂/Ar stream for 12 h. 2,7-Dibromo-9-phenyl-9-fluorenol (**1**),¹⁴ 4,4'-diformyltriphenylamine (**2**),¹⁵ and 9,9-bis(4-diphenylaminophenyl)-2,7-dibromofluorene (**4**)¹⁶ were synthesized according to the literatures.

Synthesis of 4,4'-[4-(2,7-Dibromo-9-phenyl-9H-fluoren-9-yl)phenylazanediyl]diben-zaldehyde (3)

To a stirred mixture of **1** (1.0 g, 2.4 mmol) and **2** (0.73 g, 2.4 mmol) in dichloromethane (100 mL), a solution of $BF_3 \cdot EtO_2$ (0.38 mL, 3.0 mmol) was added dropwise in dichloromethane (20 mL) under purified argon. Stirring was continued for 48 h at room temperature, and then 50 mL of ethanol and 150 mL of water were added to the reaction mixture. After phase-separation, the aqueous layer was repeatedly extracted with dichloromethane, and the combined organic layers were washed with water, dried over MgSO₄, filtered, and the solvent evaporated. The obtained crude product was further purified by column chromatography (SiO₂/petroleum ether-dichloromethane) to give compound **3** (0.94 g, 56%).

¹H NMR (CDCl₃, 400 MHz): δ /ppm = 9.91 (s, 2H), 7.79 (d, 4H), 7.62 (d, 2H), 7.53 (d, 4H), 7.30 (d, 2H), 7.27 (s, 1H), 7.14 (m,8H), 7.04 (d, 2H).

Synthesis of Polymer PFTPA-CHO

A mixture of **3** (44.6 mg, 0.06 mmol), **4** (206.8 mg, 0.26 mmol), **5** (200 mg, 0.34 mmol), Pd(PPh₃)₄ (2 mg, 0.002 mmol), K_2CO_3 (276 mg, 2 mmol), toluene (4 mL), and distilled water (1 mL) was stirred at 80 °C for 36 h under purified argon. Then, a solution of sodium diethyldithiocarbamate trihydrate (22.5 mg, 0.1mmol) in deionized water (1 mL) was added to the above system, and continued to stir at the same temperature for 12 h. The collected precipitate from methanol was further purified by Soxhlet extraction with acetone for 72 h to give a pale yellow solid (240 mg, 77%).

¹H NMR (400 MHz, CDCl₃): δ /ppm = 9.87 (s, 1H), 7.80–6.96 (m, 95H), 2.10–1.99 (m, 10H), 1.20–0.94 (m, 30 H), 0.83–0.62 (m, 25H). GPC (tetrahydrofuran (THF), linear polystyrene as reference): $M_n = 1.72 \times 10^4$, $M_w/M_n = 2.2$.

Functionalization of RGO with PFTPA (RGO-PFTPA)

A mixture of PFTPA-CHO (100 mg), RGO (5 mg), and *N*-methylglycine (20 mg) in anhydrous *N*,*N*-dimethylformamide (50 mL) was refluxed for 4 days. After termination of reaction, 150 mL of methanol was added to the above mixture. The crude product was collected by filtration using a polycarbonate film (ϕ 0.45 μ m), washed with ethanol and dichloromethane, respectively, and then redissolved in DMF, and filtered through a double-layer filter paper to remove possible cross-linked byproducts. The filtration was poured into methanol to give a gray-black solid which was thoroughly vacuum-dried at 60 °C over night. Yield: 89 mg.

Theoretical Calculations

Molecular simulations of the electronic properties, including the optimized geometry, highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) of the basic unit (BU) of RGO-PFTPA, were carried out using the Gaussian 03 program package on a Hewlett-Packard Xeon Two Sockets Quad-Core workstation with 8 CPUs and 16 GB





SCHEME 1 Synthesis of PFTPA-CHO and RGO-PFTPA.

memory. Orbital energy levels were calculated with density function theory in B3LYP/6-31G(d) level. Vibrational frequencies were calculated analytically to ensure that the optimized geometries really correspond to the total energy minima.

Device Fabrication

The Indium tin oxide (ITO) glass substrates were precleaned in ultrasonic bath for 15 min each in detergent, deionized water, acetone, and isopropyl alcohol. A $50-\mu$ L RGO-PFTPA solution in toluene was spin-coated onto the precleaned ITO substrate. The resultant film thickness is around 100 nm as measured by a step-profiler. After drying, the film under reduced pressure (10^{-5} Torr) and at room temperature overnight, Al top electrodes were deposited onto the film surface via thermal evaporation at 10^{-7} Torr through a shadow mask. The *J*-V data reported were based on devices



FIGURE 1 IR spectra of (a) GO, (b) RGO, (c) PFTPA-CHO, and (d) RGO-PFTPA.

of 0.4 \times 0.4 $\rm mm^2$ in size and about 300 nm in thickness. The devices were characterized under ambient conditions, using a Hewlett-Packard 4155B semiconductor parameter analyzer with an Agilent 16440A SMU/pulse generator.

RESULTS AND DISCUSSION

PFTPA-CHO was synthesized through the Suzuki coupling reaction of the monomers 3, 4, and 5 in the presence of the tetrakis(triphenylphosphine)palladium(0) as the catalyst. This polymer is soluble in many common organic solvents, and by GPC analysis against a linear polystyrene standard was found to have $M_{\rm n}$ of 1.72 \times 10⁴, and a polydispersity of 2.2. RGO was functionalized by the 1,3-dipolar cycloaddition reaction between PFTPA-CHO, N-methylglycine, and RGO. It should be noted that in this reaction multifunctionalized PFTPA-CHO could have trouble forming network or crosslinked structures. As a matter of fact, a small amount of insoluble cross-linked byproducts have indeed been observed, but they can be easily removed from the crude product by filtering, as described in the "Experimental" section. In addition, this problem can also be effectively overcome by carrying out the addition reaction in the dilute solution. The resultant polymer-grafted RGO is soluble in some common organic solvents. The key to the development of RGO functionalized polymers is to form selectively soluble polymercovalently grafted RGO hybrid materials under very mild conditions.

As shown in Figure 1, the most characteristic features in the IR spectrum of GO are the absorption bands corresponding to the C=O carbonyl stretching at 1733 cm⁻¹, the O—H deformation vibration at 1412 cm⁻¹, the C—OH stretching at 1226 cm⁻¹, and the C—O stretching at 1053 cm^{-1.5(i),17} After reduction by thermal annealing, the characteristic absorption bands of GO nearly disappeared in the IR spectrum of RGO. As expected, the characteristic stretching vibration of aldehyde groups in the IR spectrum of PFTPA-CHO at 1697 cm⁻¹ is absent in the IR spectrum of RGO-PFTPA, suggesting that PFTPA was grafted onto the surface of RGO. Because of the overwhelming contributions of multiple polymer chains in

the RGO-PFTPA structure, the observed IR spectrum of RGO-PFTPA is in general similar to that of PFTPA and does not give additional structural information.

The significant structural changes from GO to RGO, and then to RGO-PFTPA are also reflected in their Raman spectra (Fig. 2). Upon excitation with 768 nm laser, the Raman spectrum of GO displays two prominent bands at about 1,315 (D-band) and 1,581 (G-band) cm⁻¹ with the I_D/I_G of 0.52. The D- and G-bands of RGO are located at 1,308 and 1,591 cm^{-1} , respectively, with a considerably increased D/G intensity ratio (2.07) compared with that in GO. This change implies a decrease in the average size of the sp² domains upon reduction of GO.¹⁸ Covalent graftings of the PFTPA chains onto the surface of RGO led to the blue-shift of the Dband and the red-shift of G-band, respectively, followed by the increase of the G-band intensity. This result is not in accordance with the previous reports in the literatures.^{5(a,c,d),10,19} Usually functionalization of GO and RGO would lead to the enhancement of the I_D/I_G ratio because the Dband has been used to monitor the process of covalent functionalization which transforms sp² to sp³ sites, whereas the G-band could be used to estimate the level and distribution of modification. In this study, the PFTPA moieties grafted onto the surface of RGO contain a large amount of sp^2 aromatic carbon atoms (see the structure of PFTPA). Therefore, although some sp² carbon atoms in RGO were transformed to sp³ carbon atoms, their amounts are still less than those of the sp^2 carbon atoms in the grafted polymer moieties. This is the reason why the I_D/I_G ratio is decreased from 2.07 for RGO to 0.85 for RGO-PFTPA.

The XPS spectra provide essential and useful information for the reduction of GO and for the covalent attachment of the PFTPA moieties onto the surface of RGO (Fig. 3). From Figure 3(a,c,e), it can be clearly seen that after reduction by thermal annealing, a very weak O1s signal is observed in both XPS spectra of RGO and RGO-PFPTA, this implying that a trace of residual oxygen functionalities still exist in the structure of RGO. The C1s XPS spectrum of GO [Fig. 3(b)]



FIGURE 2 Raman spectra of (a) GO, (b) RGO, and (c) RGO-PFTPA. $\lambda_{\text{ex}}=785$ nm.



FIGURE 3 XPS wide-scan spectra and C1s core-level spectra of the samples. The inset of (f) is the N1s core-level spectrum of RGO-PFTPA.

showed the peaks of carbon functionalities appeared at 283.9 (the C in C=C bond), 284.6 (the C in C-C bond), 286.4 (the C in C-O bond), and 288.4 (the C in C=O bond) eV.²⁰ The C1s XPS spectrum of RGO [Fig. 3(d)] showed three peaks at 283.8, 284.8, and 286.1 eV, followed by a notable enhancement of the sp^2 C signal intensity in C=C bond, and a considerable decrease of the sp^3 C intensity in C-O bond, this implying the most of oxygen functionalities in GO have been removed via thermal reduction. After covalent functionalization with PFTPA, the C1s XPS spectrum of RGO-PFTPA displayed the C signal of C-N bond at 285.5 eV. The N1s

XPS spectrum of RGO-PFTPA [see the inset of Fig. 3(f)] showed two peaks of nitrogen functionalities at 399.8 (the N in Ph_3N) and 400.6 eV (the N in pyrrole ring).

The intramolecular D-A structure usually allows charge transfer (CT) interaction or HOMO–LUMO electron transition to occur, as is evident from the photoluminescence spectrum of RGO-PFTPA (Fig. 4). By using 375-nm excitation light, the fluorescence emission exhibited by RGO-PFTPA was apparently quenched as compared with PFTPA-CHO. To confirm this, we measured the absolute fluorescence quantum yields



FIGURE 4 (a) UV/Vis absorption and (b) photoluminescence ($\lambda_{ex} = 375$ nm) spectra of the samples in THF.

of the samples in dilute THF solution by using a Horiba-Jobin-Yvon integrating sphere. The values are estimated as 51% for PFTPA-CHO and 40% for RGO-PFTPA. Such intramo-



FIGURE 5 TGA profiles of the samples measured in nitrogen (heating rate 10 $^{\circ}$ C/min, N₂ flow 100 mL/min).



FIGURE 6 Current density-voltage (*J-V*) characteristics of a 0.16 mm² Al/RGO-PFTPA/ITO device (a); stability of the device in the ON and OFF states (b) under a constant stress of -1 V and (c) under a continuous read pulse with a peak voltage of -1 V, a pulse width of 1 μ s, and a pulse period of 2 μ s. The inset of (b) shows the device structure used in the measurement.

lecular quenching process may involve energy and/or CT between PFTPA as electron donor and RGO as electron acceptor. The UV/Vis absorption spectrum of PFTPA-CHO shows two peaks at 312 and 382 nm, with a relatively intensity ratio of 0.48. The main absorption peaks of RGO-PFTPA are located at 307 and 358 nm, respectively, with a relatively



FIGURE 7 (Left) Calculated molecular orbitals of the BU of RGO-PFTPA, and (right) electronic transition from the ground state to the CT state.

intensity ratio of 1.45. When compared with PFTPA-CHO, the maximal absorption peak of RGO-PFTPA is shifted to the blue by $\Delta \lambda = 24$ nm.

TGA curves of the samples are shown in Figure 5. GO is thermally unstable and suffers 15% weight loss upon heating to 100 °C. The main mass loss (\sim 30%) takes place around 200 °C and is ascribed to the decomposition of labile oxygen functional groups present in the material. A slower, steady mass loss (\sim 15%) of GO over the entire temperature range above 300 °C can be attributed to the removal of more stable oxygen functionalities. When compared with GO, RGO shows much increased thermal stability. When heated to 800 °C, about 8.12% weight loss was observed due to the removal of the residual oxygen functionalities. The thermal stability of RGO-PFTPA is apparently higher than that of PFTPA-CHO. At 800 °C, the amount of the PFTPA-CHO residue is 27.92%, whereas that of the RGO-PFTPA residue is about 56.06%.

The nonvolatile rewritable memory effect was observed from the characteristic J-V curves of the ITO/RGO-PFTPA/Al device [Fig. 6(a)] under ambient conditions. The J-V curves clearly displayed electrical bistable memory behaviors. The voltage applied to the device was varied in a cycle from 0 \rightarrow -3 \rightarrow $0 \rightarrow 3 \rightarrow 0 \rightarrow -3$ V. Starting with the OFF state (low-conductivity state) in the as-fabricated device, the current increases slowly with the applied negative voltage sweep (0 \rightarrow -3V). At the switch-on voltage of about -1.4 V, the current density increases abruptly from 10^{-2} to 10^2 A/cm² (Sweep 1), indicating device transition (writing process) from the OFF state to the ON state (high-conductivity state). After removing the power supply, the device is still in the ON state (Sweep 2). These results indicated that "OFF" and "ON" can be encoded from the corresponding low-conductivity state (before writing) and high-conductivity state (after writing), with an applied voltage (read voltage) in the range of 0 to -1.4 V. After reading the ON state in the positive



FIGURE 8 Experimental and fitted *J-V* characteristics of the Al/ RGO-PFTPA/ITO device in (a) the OFF state and (b) the ON state.

sweep (Sweep 3), a positively biased sweep can program the ON state back to the original OFF state at 1.65 V (erasing process). The OFF state of the device can be read (Sweep 4) and reprogrammed (Sweep 5) to the ON state again in the subsequent negative sweep. Such a cycle is a typical "writeread-erase-read-rewrite" process for a nonvolatile rewritable memory device. Although there are some minor shift of the write/erase voltages due to the possible influence of the electrical stress on the inherent electrical relaxation of the memory materials, as expected, our operation cycles can still be repeated with fairly good accuracy. From Figure 6(b,c), it can be clearly seen that both the OFF and ON states are accessible and very stable under a constant voltage stress of -1 V for up to 3 h, or under a pulse voltage stress of -1 V for up to one hundred million continuous read cycles (pulse period = 2 μ s, pulse width = 1 μ s), with a ON/OFF state current ratio of more than 10^3 .

To gain insights into the geometry and electronic structures of RGO-PFTPA, computational studies were performed using density functional theory at B3LYP/6-31G level (Fig. 7). The accuracy of these methods was demonstrated by Schaefer and coworkers on electron affinities of aromatic compounds.²¹ To reduce the CPU time, a simplified model, in which the RGO entity contains 18 aromatic rings and the polymer segment only contains one repeat unit, was used to represent the BU of RGO-PFTPA. From the optimized structures, the electron distributions of both the HOMO and the LUMO were found to be located on the RGO entity. In contrast to GO, in which the ballistic charge carrier transport is significantly hindered due to scattering at the surface or edge defects of epoxide and carbonyl groups, the bandgap of graphene is theoretically zero, this leading to an excellent but nonswitchable electrical conductivity,⁹ and posing a major challenge for graphene electronics because the absence of a bandgap limits the ON/OFF ratio of digital devices. Upon covalent functionalization with the conjugated PFTPA polymer, the π -conjugation system of RGO was effectively disturbed, giving rise to an increase of energy bandgap. Both the HOMO-1 and HOMO-2 are located on the PFTPA moieties. Without inputting voltage, the electrons in the system are stable, this implying that the as-fabricated Al/RGO-PFTPA/ITO device is in the OFF state. The OFF state current can be fitted by a space-charge-limited current model [Fig. 8(a)]:

$$J = A\mu\varepsilon\varepsilon_0 \frac{V^2}{d^3} \exp\left\{\frac{0.891}{kT} \left(\frac{q^3V}{\pi\varepsilon\varepsilon_0 d}\right)^{1/2}\right\}$$
(1)

where J, A, μ , $\varepsilon\varepsilon_0$, d, V, k, T, and q are current density, positive constant, the mobility of charge carriers, the absolute permitivity of the complex, the thickness of the film, voltage, the Boltzmann constant, the ambient temperature, and the absolute value of the unit electronic charge (1.6 \times 10⁻¹⁹ C), respectively. At the switching threshold voltage, electrons transit readily from HOMO to LUMO within the RGO moiety, forming the locally excited state. However, the direct electron transition from HOMO-1 and HOMO-2 to HOMO is restrained due to the absence of overlapping between HOMO-1 or HOMO-2 and HOMO (Fig. 7). HOMO-2 is distributed in the molecular fragment directly bonded to the RGO nanosheets, whereas the HOMO-1 is separated from the HOMO on the RGO nanosheets by the fluorene moieties. Therefore, the spatial hindrance will deter electron transition from HOMO-1 to HOMO. Alternatively, electron transition from HOMO-2 to HOMO will be easier to occur, filling the holes generated in HOMO, followed by the spontaneous electron transition from HOMO-1 to HOMO-2. In this case, the CT state is formed. Along with the increase of CT interaction, dual-channel charge transport pathways will form via inter-plane hopping in RGO, and switch the Al/RGO-PFTPA/ITO device from the OFF state to the ON state.^{1(c)} The ON state current can also be fitted by an Ohmic current model [Fig. 8(b)]

$$J = B \frac{qnV}{d} \tag{2}$$

where B is a positive constant and n is the density of charge carriers. The intensive electron delocalization in RGO can effectively stabilize the conductive CT state, resulting in the nonvolatile nature of the ON state. However, application of a



reverse positive bias to the device can extract electrons from the graphene, and program the device back to the OFF state.

CONCLUSIONS

The reaction between sarcosine, RGO, and PFTPA-CHO, led to a π -conjugated polymer-covalently grafted RGO memory material (RGO-PFTPA). This material exhibited good solubilities in several common organic solvents and good thermal stability. When compared with PFTPA-CHO, the maximal absorption peak of RGO-PFTPA is shifted to the blue by $\Delta \lambda = 24$ nm, and the fluorescence emission exhibited by RGO-PFTPA was apparently quenched due to the CT between PFTPA as electron donor and RGO as electron acceptor. The J-V curves of the ITO/ RGO-PFTPA/Al device clearly displayed nonvolatile rewritable memory performance. Both the OFF and ON states are accessible and very stable under a constant voltage stress of -1 V for up to 3 h, or under a pulse voltage stress of -1 V for up to 10^8 continuous read cycles (pulse period = 2 μ s, pulse width = 1 μ s), with a ON/OFF state current ratio of more than 10³. As this material can be processed in solution, RGO-PFTPA provides great opportunities for device fabrication at low cost and with ease. With the excellent electrical, mechanical, and thermal properties of graphene, this bistable switching behavior makes RGO-PFTPA a promising alternative or supplement to present silicon-based semiconductor technology.

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