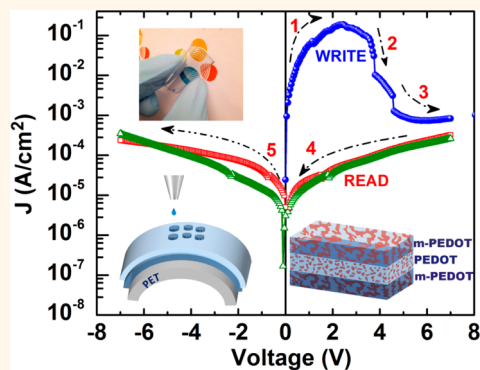


Metal-Free, Single-Polymer Device Exhibits Resistive Memory Effect

Unnat S. Bhansali,[†] Mohd A. Khan,[†] Dongkyu Cha,[†] Mahmoud N. AlMadhoun,[‡] Ruipeng Li,[†] Long Chen,[†] Aram Amassian,[†] Ihab N. Odeh,[‡] and Husam N. Alshareef^{†,*}

[†]Materials Science & Engineering, King Abdullah University of Science and Technology (KAUST), Thuwal 23955-6900, Saudi Arabia, and [‡]Corporate Research and Innovation Center, Saudi Basic Industries Corporation (SABIC), Thuwal 23955-6900, Saudi Arabia

ABSTRACT All-polymer, write-once-read-many times resistive memory devices have been fabricated on flexible substrates using a single polymer, poly(3,4-ethylenedioxythiophene):polystyrene sulfonate (PEDOT:PSS). Spin-cast or inkjet-printed films of solvent-modified PEDOT:PSS are used as electrodes, while the unmodified or as-is PEDOT:PSS is used as the semiconducting active layer. The all-polymer devices exhibit an irreversible but stable transition from a low resistance state (ON) to a high resistance state (OFF) at low voltages caused by an electric-field-induced morphological rearrangement of PEDOT and PSS at the electrode interface. However, in the metal–PEDOT:PSS–metal devices, we have shown a metal filament formation switching the device from an initial high resistance state (OFF) to the low resistance state (ON). The all-PEDOT:PSS memory device has low write voltages (<3 V), high ON/OFF ratio (>10³), good retention characteristics (>10 000 s), and stability in ambient storage (>3 months).



KEYWORDS: PEDOT:PSS · resistive memory · flexible substrates · polymer electrodes · metal filament · solution process

Polymer-based resistive random access memory (RRAM) is rapidly emerging as the technology for nonvolatile memory applications particularly for large-area, flexible, and printed electronics.¹ Write-once-read-many times (WORM) memory devices based on the resistive switching (RS) mechanism are attractive for many applications. They can be used for reliable and long-term storage of archival standards and tamper-proof, secure databases, where data can only be physically written once but read many times. WORM memory can function as the conventional CDR, DVD \pm R, or PROMs. WORM memories can also be used as disposable memories in some niche areas, such as electronic labels and RFID tags. Polymeric memories can offer an easy-to-process, low-cost alternative to conventional inorganic memory devices. A number of organic donor–acceptor complexes,² nonconjugated metal–organic complex,³ conjugated polymers such as regiorandom and regioregular poly(3-hexylthiophene) (P3HT), polyaniline (PANI), and poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) have been

shown to exhibit bistability and nonvolatility for WORM memory applications.^{4–9} Although polymer RRAM devices based on PEDOT:PSS have been extensively studied since the first demonstration of resistive switching behavior in PEDOT:PSS in a WORM memory architecture,⁹ the behavior and switching mechanisms remain debatable as evident from the large variation in reported results. There have been numerous reports of nonpolar, unipolar, and bipolar switching in PEDOT:PSS-based RRAM systems,^{5–7,10,11} and the switching behavior of PEDOT:PSS is reported to depend strongly on the bottom electrodes and the method of operation.^{11–13} Typically, formation and rupture of the conducting PEDOT+ filaments through oxidation and reduction of PEDOT:PSS is reported as the switching mechanism. However, formation of a metal filament through polymer thin films has also been reported as the origin of electrical bistability.^{4,14,15}

In this study, we report an all-polymer resistive memory device made from a single polymer PEDOT:PSS. Replacing the metal or oxide electrodes in PEDOT:PSS memory devices by polymer electrodes can help us

* Address correspondence to husam.alshareef@kaust.edu.sa.

Received for review May 2, 2013 and accepted November 9, 2013.

Published online November 09, 2013
10.1021/nn403873c

© 2013 American Chemical Society

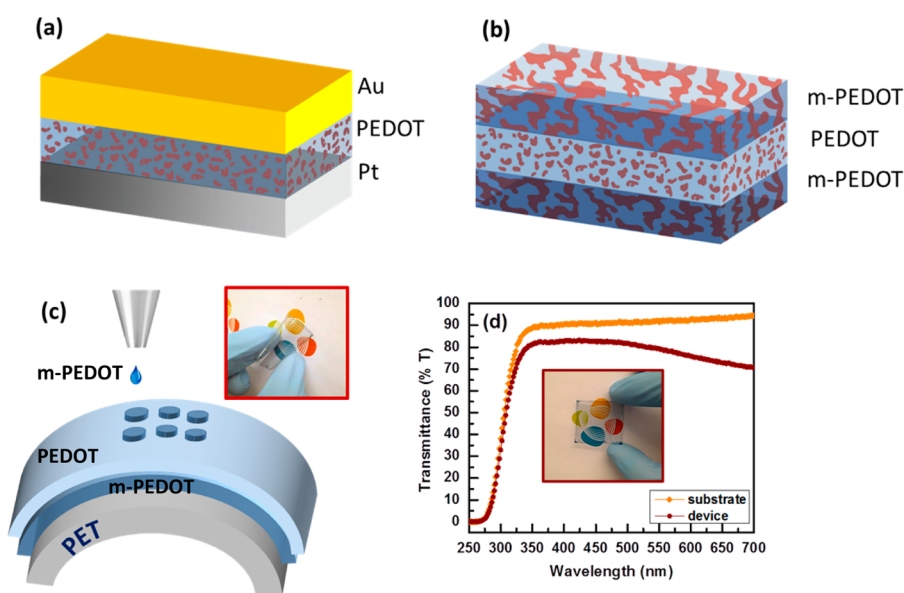


Figure 1. Schematic illustrations of the nonvolatile, PEDOT:PSS-based resistive memory devices with (a) Pt and Au as bottom and top electrodes, respectively, and (b) highly conducting PEDOT:PSS (m-PEDOT) as the bottom and top electrode (c) all-PEDOT memory device with an inkjet-printed array of top electrodes on the flexible PET substrate. The inset shows an actual photograph of the flexible memory device. (d) UV-vis optical transmission spectra of the flexible device compared to the bare substrate in the range of 250–700 nm. The photograph in the inset demonstrates the transparency of the final device.

determine the real switching mechanism by eliminating the potential for metallic atom diffusion into the polymer. In addition to the resistive switching behavior, PEDOT:PSS also has a unique set of properties ideally suited for fabricating memory devices on large, flexible substrates. For example, PEDOT:PSS is highly stable, easy-to-process, forms transparent thin films, and can be made highly conductive by the use of additives.^{16–18} Moreover, its ability to form continuous, smooth films from an aqueous dispersion by simple techniques such as spin-coating, doctor-blading, or inkjet-printing is an attractive feature for large-area, high-throughput, flexible electronics.

In this work, we have fabricated two types of resistive WORM memories and compared their switching behavior and mechanism. One memory is a metal/PEDOT:PSS/metal resistive memory device, in which Au and Pt were used for the top and bottom electrodes, respectively; the other is a single polymer (all-PEDOT:PSS) resistive memory device on a flexible substrate, which consists of a thin film made from as-received PEDOT:PSS dispersion to form the active layer, sandwiched between two highly conducting PEDOT:PSS (m-PEDOT) electrodes. The all-polymer devices can be very attractive to the flexible printed electronics community. The devices are extremely low-cost, solution-processed, scalable, highly transparent, flexible, and fabricated at very low temperatures of ~ 120 °C.

RESULTS AND DISCUSSION

Figure 1a,b shows the schematic illustration of PEDOT:PSS (referred to as PEDOT)-based resistive memory

devices with metal electrodes and polymer electrodes, respectively. The active PEDOT:PSS layer is the same in both structures. However, in the case of the all-polymer device, the metal electrodes have been replaced with modified PEDOT:PSS (m-PEDOT) to which 4 wt % dimethyl sulfoxide (DMSO) has been added, and hence the different texture in the schematic in Figure 1b. The device cross section of an all-PEDOT:PSS memory device on a PET substrate is illustrated in Figure 1c, with the inset showing the real finished device. To make the all-polymer device, a single layer of m-PEDOT was spun-cast on pre-cleaned substrates, followed by annealing on a hot plate at 120 °C for 1 h to form the bottom electrode. Once completely baked, the layer becomes insoluble, allowing spin-casting of the subsequent active polymer layer, PEDOT. To confirm that there is no redissolution of the underlying PEDOT:PSS layer, we immersed a thin film of PEDOT:PSS spun on a Si wafer in deionized (DI) water for 30 min. PEDOT:PSS is used as an aqueous solution; therefore, if at all, it should dissolve in water. We found that there is a negligible difference in the thickness of the PEDOT:PSS film measured before and after immersion, shown in the Supporting Information. After the active layer, a formulated m-PEDOT:PSS-based ink was used to inkjet-print an array of top electrodes. The thicknesses measured using a Dektak profilometer for the bottom electrode, active layer, and the top electrode are 30 ± 2 , 40 ± 2 , and 90 ± 5 nm, respectively, confirmed by cross-sectional transmission electron microscopy (TEM).

An actual photograph of the device is shown as an inset in Figure 1c, where the flexibility of the substrate

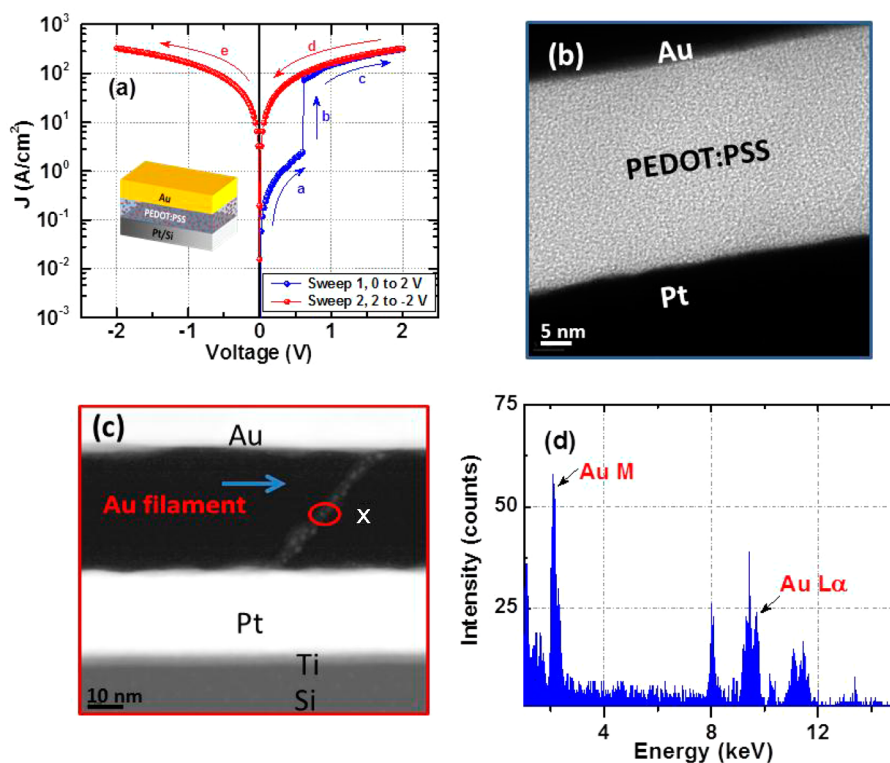


Figure 2. (a) Typical J – V curve for Pt/PEDOT:PSS/Au memory device measured in the voltage range of +2 to –2 V and compliance set at 10 mA. The sweep direction is indicated by the labeled arrows. (b) Cross-sectional TEM image of a pristine device depicting the amorphous PEDOT:PSS layer with a thickness of ~ 40 nm. (c) Cross-sectional STEM image showing the filament composed of Au atoms connecting the top and bottom electrodes. The encircled area “x” denotes the spot for EDX analysis. (d) Spot EDX spectrum obtained from the encircled area showing Au M and Au L α peaks at 2.12 and 9.7 keV, respectively.

is demonstrated. Figure 1d shows the UV–visible optical transmission spectra of the bare plastic substrate and the full all-PEDOT:PSS device stack from wavelength, $\lambda = 250$ –700 nm. The optical transmission only drops from 82 to 76% in the wavelength range of 350 to 600 nm. The excellent transparency of the final device stack is demonstrated in the actual photograph shown as an inset in Figure 1d.

Figure 2a shows a characteristic current density–voltage (J – V) plot for a unipolar, bistable WORM memory device with metal electrodes. The electrical characterization of these memory devices was done with the bottom electrode grounded and a sweeping bias applied to the top electrode. For all virgin devices, the current in the active PEDOT:PSS layer under positive bias increases slowly at low voltage followed by an abrupt increase in current density to 100 A/cm 2 at around 0.6 V, beyond which a saturation in the leakage current density is reached. This is shown in the sequence denoted by a \rightarrow b \rightarrow c in Figure 2a. This sharp transition from a high resistance state (HRS) to a low resistance state (LRS) can be considered as the “WRITE” step in such memory devices. Subsequent voltage sweeps from 2 to –2 V, denoted by d \rightarrow e, do not change the resistance state of the cell, implying that the data can only be “READ” once it has been written. It has been suggested that such a behavior in PEDOT:PSS

resistive memory is due to the formation of conducting PEDOT $^+$ filament through charge-induced oxidation.^{6,7} However, in this case, the formation of a conducting PEDOT $^+$ filament through charge-induced oxidation is unlikely because (a) it cannot explain the sharp increase in current density to 100 A/cm 2 , and (b) despite voltage sweeps in the opposite polarity, the resistance state could not be changed. The irreversibility of the process indicates that the sudden rise in current at point b is due to the formation of a metal filament, likely caused by the diffusion of Au atoms under the applied field, from the top electrode to the bottom electrode. Figure 2b shows a cross-sectional TEM image of the device Au/PEDOT:PSS/Pt stack before the switching voltages were applied (HRS state). Note that the PEDOT:PSS film (38 nm) shows an amorphous nature and no evidence of any metal diffusion. In contrast, Figure 2c shows a device which has been written to the low resistance state by application of a 2 V positive bias. Note that a filament of metal is clearly evident in the device after it has been written to the LRS. The diffused Au atoms in the amorphous polymer film form a bridge connecting the top and bottom electrode, resulting in the Ohmic behavior of the memory in the LRS state. To investigate the composition of this filament, cross-sectional scanning tunneling electron microscopy (STEM) was performed for

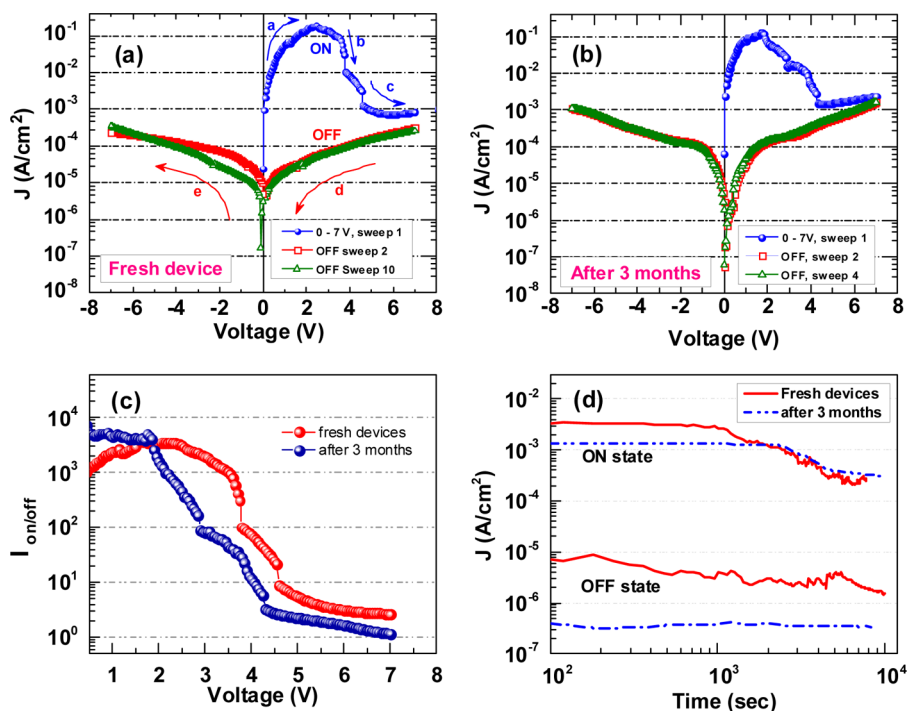


Figure 3. (a) Typical J - V characteristics of all-PEDOT resistive memory showing WORM behavior. The devices were swept from $+7$ to -7 V. A normally ON state (LRS) is maintained until 3 V, beyond which the devices are switched to an OFF state (HRS) denoted by $a \rightarrow b \rightarrow c$. (b) J - V behavior of the devices measured after 3 months of storage in ambient air, demonstrating negligible deterioration in device performance. (c) Comparison of the ON/OFF ratio for fresh and stored devices and (d) retention test measurement for fresh and stored devices. For the ON state current response, a continuous read voltage of 0.5 V was applied across the virgin devices. The OFF state current response was also measured at 0.5 V after sweeping the voltage from 0 to $+7$ V, switching the device to the HRS.

a memory cell in the LRS state and is shown in Figure 2c. High-angle annular dark-field (HAADF) imaging using STEM mode provides sensitive imaging of heavy elements due to Z -contrast. The analysis in Figure 2c clearly shows a continuous filament, about 45 nm long and 4–5 nm wide, within the PEDOT:PSS film. Several instances of such connecting metal filaments were found within the same device. A spot energy-dispersive X-ray (EDX) analysis of the metal filament, shown in Figure 2d, further confirms the presence of Au atoms. Under the STEM mode, the spot size can be controlled to very small dimensions, typically 0.5–1 nm, which rules out the possibility of collecting signals from the top Au electrode. The peaks occurring at 2.12 and 9.71 keV are identified as Au M and Au $L\alpha$, respectively. Simultaneously, we performed an EDX analysis of the PEDOT:PSS adjacent to the Au filament which showed no Au signal as shown in Figure S1 of the Supporting Information. This further confirmed that no signal was coming for the Au electrode. It is important to mention here that though other groups have reported “fusing-out” or “rupturing” of the metal filament under opposite voltage or high current, we do not observe this reversible behavior. We also measured the current–voltage (J - V) characteristics of Au/PEDOT:PSS/Pt devices under a sweeping bias of ± 4 V, shown in the Supporting Information. The current levels remain high and saturate at the

compliance level of the measurement instrument, indicating that the devices do not switch to the HRS (high resistance state) or turn OFF even at higher bias or larger current flow. Instead, we observed that the Au electrode delaminates at higher bias (~ 4 V), possibly due to Joule heating. This is consistent with our suggestion that the memory effect in the case of our device with gold electrode is not real, and devices cannot be used for practical applications. Though these devices exhibit nonvolatility, bistability, and a WORM behavior, it can be argued that the switching mechanism (metal filament formation) is an artifact of a Au diffusion process rather than a characteristic of PEDOT:PSS itself. This observation may also explain why there are large variations in the reported behavior of PEDOT:PSS resistive memories that use metal electrodes, particularly Au. Partial or full diffusion of the Au electrode through the active polymer layer can mask the true behavior of the device and produce such variation.^{4,7,11,14,20} To study the actual memory behavior of PEDOT:PSS, one must eliminate the possibility that metal filament formation will mask the true resistive switching behavior of the PEDOT:PSS active layer itself. Therefore, we have fabricated all-polymer, metal-free PEDOT:PSS resistive memory devices, where metals were replaced with conductive polymer contacts. As shown in Figure 1b, conducting electrodes were made using modified PEDOT:PSS (m-PEDOT) instead of metals.

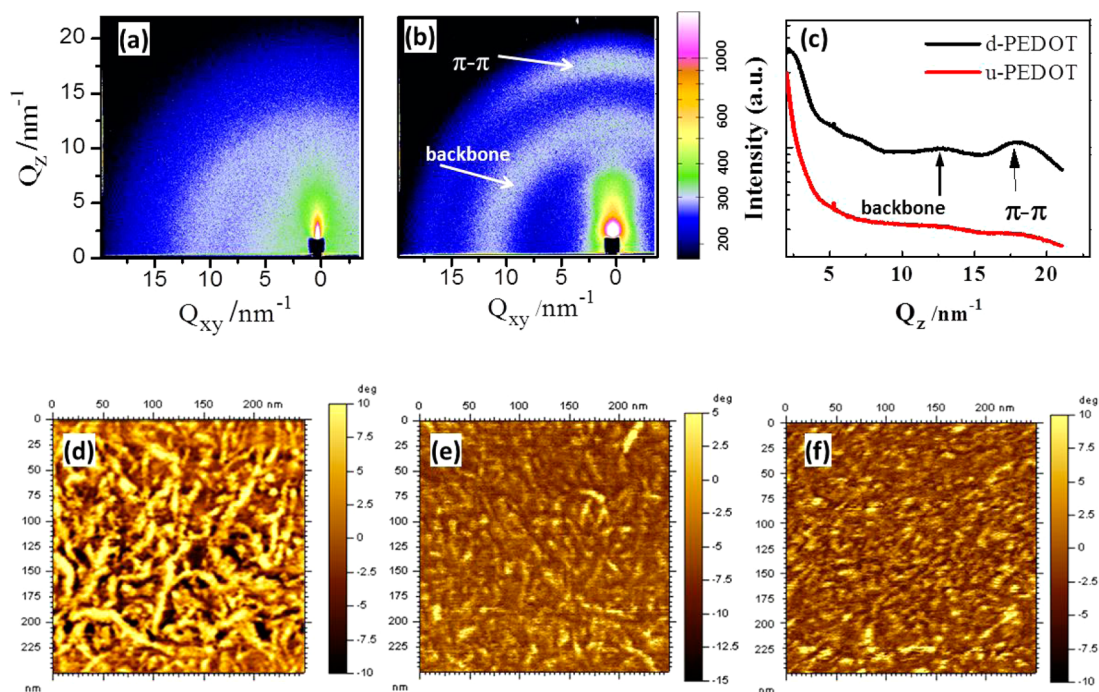


Figure 4. GIWAXS patterns of (a) PEDOT thin film and (b) m-PEDOT thin film with the scale bar shown on the side; (c) intensity integration along the Q_z direction for the two films indicating peaks originating from the π - π stacking and the interaction from the polymer backbone; $0.25 \mu\text{m} \times 0.25 \mu\text{m}$ AFM phase images of (d) m-PEDOT film spun on PET, (e) PEDOT spun on m-PEDOT showing conducting PEDOT domains as bright, elongated features, and (f) PEDOT spun on a metal substrate, Pt showing a granular morphology.

The J - V characteristics of freshly prepared all-PEDOT:PSS resistive memory devices on a flexible PET substrate are shown in Figure 3a. It is evident that no “forming” process was required to induce the memory behavior in these devices. The devices are typically in a low resistance state (LRS) at voltages below ~ 3 V. This state could be considered as the ON state of the memory. Applying a relatively high voltage, more than 3.5 V, writes the device into a HRS, which becomes the OFF state of the memory. After the writing step, the current from the devices is low ($< 10^{-3}$ A/cm 2) and remains low up to ± 7 V. At this stage, the devices are considered set and can no longer be rewritten, edited, or tampered with, emphasizing their potential use in security and data protection applications. Also, the high $I_{\text{ON}}/I_{\text{OFF}}$ ratio ($> 3 \times 10^3$) at low operating voltages suggests lower possibilities of data misread in such devices.

The information stored in these devices can be accessed or read many times using small voltages (e.g., 1 V or lower). After multiple voltage sweeps from 0 to ± 7 V (denoted by d \rightarrow e), the device still remains in the HRS or the OFF state, demonstrating good electrical bistability. All the devices measured exhibit an irreversible transition from a conducting (LRS) state to a nonconducting (HRS) state at higher voltages. This irreversible and reproducible switching process is essential for memory components in practical circuits. The J - V characteristics of these devices were remeasured after 3 months of storage in ambient air, as shown in Figure 3b. The switching characteristics and

device performance are maintained, suggesting excellent stability of these all-PEDOT:PSS memory devices. Figure 3c compares the $I_{\text{ON}}/I_{\text{OFF}}$ ratio of devices measured immediately after fabrication (fresh) and after 3 months of storage. The slightly higher $I_{\text{ON}}/I_{\text{OFF}}$ ratio for the aged devices in the low voltage regime (< 2 V) can be attributed to the lower leakage current in the OFF state of the aged device, while the ON state current remains similar. In comparison, at higher voltages, the $I_{\text{ON}}/I_{\text{OFF}}$ ratio for the aged devices declines at a faster rate than the fresh device, which could be attributed to the higher conductivity of the polymer thin film due to oxidation over time, which can also be seen in Figure 3b.

The retention characteristics of the PEDOT:PSS memory with polymer electrodes were also investigated. Figure 3d shows the magnitude of the ON and OFF state currents measured as a function of time for 10 000 s. The ON state (LRS) current was measured at 0.5 V, prior to switching the device to the OFF state. In comparison, the OFF current was measured after switching the device to OFF state (HRS) by applying a high voltage. The device shows an ON/OFF ratio of 100, even after 10 000 s, indicating excellent retention characteristics of these devices. For comparison, we also measured the retention properties of the all-polymer memory devices after 3 months of storage in ambient conditions, but no significant deterioration was observed for the current values in the ON state, as shown in Figure 3d. After 3 months, the OFF state

current is reduced by almost an order of magnitude, consistent with the J – V data shown in Figure 3b.

The most widely reported switching mechanism for bistable polymeric resistive memory devices is based on filamentary conduction. There are generally two types of conductive filaments for polymeric systems, particularly involving PEDOT:PSS as the active layer. One is a metal filament which involves either a redox reaction or the diffusion of the electrode metal atoms under an applied electric field, and the other is a molecular filament mainly related to the oxidation and reduction of the PEDOT:PSS thin film.¹⁴ We have investigated the morphology and crystallinity of m-PEDOT and PEDOT films using grazing incident wide-angle X-ray scattering (GIWAXS) and atomic force microscopy (AFM). Our results suggest that the ON to OFF switching behavior of devices with polymer electrodes may also be related to the morphological modifications of the PEDOT:PSS polymer films.

The GIWAXS patterns of PEDOT and m-PEDOT thin films are shown in Figure 4a,b, respectively. The m-PEDOT film shows better crystallinity, as indicated by the stronger diffraction intensity from π – π stacking and the polymer backbone, compared to the PEDOT films. Figure 4c compares the integrated intensity along the Q_z direction for both films. The two scattering peaks in the m-PEDOT film occurring at $Q_z = 12.4$ and 17.7 nm^{-1} (d spacing = 5.1 and 3.5 Å) correspond to the backbone interchain planar distance and the distance between the π -orbitals, respectively.^{21,22} At the molecular level, the increased π – π orbital interaction facilitates better charge transport, resulting in the higher conductivity of m-PEDOT films compared to the amorphous PEDOT films. The higher conductivity of m-PEDOT films can also be explained based on the morphology studied by tapping-mode AFM shown in Figure 4d. In m-PEDOT, addition of high boiling point solvents like DMSO or diethylene glycol (DEG) causes the excess PSS to phase-segregate into PSS-rich domains while the PEDOT grains merge together to form an interconnected network of conducting fibers (bright fibers in the phase image).²³ The presence of highly conducting PEDOT fibers through which the charges can move freely leads to the high conductivity ($>900 \text{ S cm}^{-1}$) of the m-PEDOT film, rendering it suitable for use as an electrical contact.

The phase images of the PEDOT layer spun-cast on m-PEDOT film and Pt substrate are shown in Figure 4e,f,

respectively. Unmodified PEDOT films spun on the m-PEDOT layer show elongated features or grains of PEDOT dispersed in the PSS matrix, similar to the underlying m-PEDOT film. This is in contrast to the granular morphology for PEDOT films grown on metal (Pt) substrates, as seen from Figure 4f. We believe that the fibrous morphology of the bottom m-PEDOT electrode film promotes preferential alignment of PEDOT chains at the interface, forming a conducting path for the charges to freely move from the bottom electrode to the PEDOT active layer. This can also explain the normally ON state of the memory devices at low voltages, as noticed in the J – V characteristics. However, it is known that in the presence of a large applied electric field, the insulating PSS chains migrate toward the interface and prevent charge transport from the bottom electrode to the PEDOT+ fibers.²⁴ This hindered charge transport results in a gradual drop in conductivity of the PEDOT:PSS film, and consequently, the current density is also reduced at high voltages. Further voltage sweeps result in low current response or the OFF state of these devices. We have characterized our WORM memory devices with high voltages ($\pm 12 \text{ V}$) to see the stability of the behavior. This was to see if the PSS diffusion can be reversed and whether the devices can be turned back to the ON state. The current from all-polymer memory devices remained low even at high voltages up to 12 V, as shown in the Supporting Information. Thus we do not observe any reversible behavior, further confirming the stable WORM behavior of our polymer devices. This irreversible, field-induced change in the morphology defines the WORM behavior in PEDOT:PSS resistive memory devices.

CONCLUSIONS

In summary, we have fabricated all-polymer resistive memory devices on transparent and flexible substrates using a single polymer (PEDOT:PSS). The devices use DMSO-modified, highly conducting PEDOT:PSS as bottom and top electrodes and an unmodified PEDOT:PSS film as the active layer. Highly stable and reproducible devices with an ON/OFF ratio $>10^3$, low writing voltage, and excellent retention characteristics have been maintained for 3 months so far. The switching behavior of PEDOT:PSS with metal electrodes may be susceptible to misinterpretation due to irreversible diffusion of metallic contacts, possibly forming filaments larger than the film thickness.

METHODS AND MATERIALS

The conductivity of PEDOT:PSS (Clevios PH-1000 from Heraeus) was enhanced up to $>900 \text{ S cm}^{-1}$ by chemically doping with 4 wt % dimethyl sulfoxide (DMSO). PEDOT:PSS from Sigma Aldrich, 1.3 wt % dispersion in water and conductivity ~ 1 – 3 S/cm , was used as the active layer. Solvent-doped PH-1000 was diluted with ethylene glycol (EG) and deionized

(DI) water to form the printing ink. A MicroFab JetLab II piezoelectric inkjet printer was used to print the top patterned electrodes.¹⁹ The substrate temperature was maintained at 40 °C during the printing process, followed by annealing on a hot plate at 100 °C for 1 h.

The morphology of doped and undoped PEDOT:PSS thin films was studied using an atomic force microscope (Agilent 5400).

Grazing incident wide-angle X-ray scattering measurements were performed at D-line, Cornell High Energy Synchrotron Source (CHESS) at Cornell University. A wide band-pass (1.47%) X-ray with a wavelength of 1.167 Å was shone on the samples with a grazing incidence angle of 0.15°. A 50 × 50 mm CCD detector (Medoptics) with a pixel size of 46.9 μm was placed at a distance of 95 mm from the samples. A 1.5 mm wide tantalum rod was used to block the intense scattering in the small-angle area. The exposure time was 1 s. The samples for cross-sectional TEM analysis were prepared by focused ion beam (FIB, Helios 400s, FEI) with lift-out method. The lamella was thinned with a Ga ion beam (30 kV, 0.28 nA) and cleaned at 2 kV and 47 pA. A Titan ST 300 kV was used for high-resolution transmission electron microscopy (HRTEM) and STEM imaging. The electrical characterization including retention tests was done in ambient air using a Keithley 4200 semiconductor parameter analyzer.

Conflict of Interest: The authors declare no competing financial interest.

Acknowledgment. The authors would like to thank Mrs. Supriya Chewle for her help with the artistic rendering of the images. H.N.A. acknowledges the financial support from the KAUST baseline fund and Saudi Basic Industries Corporation (SABIC) Grant No. 2000000015.

Supporting Information Available: The EDX spectrum and the high-field $I-V$ measurements of the single polymer resistive memory device is available in the Supporting Information. This material is available free of charge via the Internet at <http://pubs.acs.org>.

REFERENCES AND NOTES

- Ling, Q.-D.; Liaw, D.-J.; Teo, E. Y.-H.; Zhu, C.; Chan, D. S.-H.; Kang, E.-T.; Neoh, K.-G. Polymer Memories: Bistable Electrical Switching and Device Performance. *Polymer* **2007**, *48*, 5182–5201.
- Liu, C.-L.; Chen, W.-C. Donor–Acceptor Polymers for Advanced Memory Device Applications. *Polym. Chem.* **2011**, *2*, 2169–2174.
- Potember, R. S.; Poehler, T. O.; Cowan, D. O. Electrical Switching and Memory Phenomena in Cu-TCNQ Thin Films. *Appl. Phys. Lett.* **1979**, *34*, 405–407.
- Joo, W.-J.; Choi, T.-L.; Lee, J.; Lee, S. K.; Jung, M.-S.; Kim, N.; Kim, J. M. Metal Filament Growth in Electrically Conductive Polymers for Nonvolatile Memory Application. *J. Phys. Chem. B* **2006**, *110*, 23812–23816.
- Ha, H.; Kim, O. Bipolar Switching Characteristics of Non-volatile Memory Devices Based on Poly(3,4-ethylenedioxythiophene):Poly(styrenesulfonate) Thin Film. *Appl. Phys. Lett.* **2008**, *93*, 033309.
- Lee, J.; Ohyun, K. Nonvolatile Resistive Memory Device Based on Poly(3,4-ethylenedioxythiophene):Poly(styrene sulfonate) Thin Film for Transparent and Flexible Applications. *Jpn. J. Appl. Phys.* **2011**, *50*, 06GF01.
- Liu, X.; Ji, Z.; Tu, D.; Shang, L.; Liu, J.; Liu, M.; Xie, C. Organic Nonpolar Nonvolatile Resistive Switching in Poly(3,4-ethylene-dioxythiophene):Polystyrenesulfonate Thin Film. *Org. Electron.* **2009**, *10*, 1191–1194.
- Wang, Z. S.; Zeng, F.; Yang, J.; Chen, C.; Yang, Y. C.; Pan, F. Reproducible and Controllable Organic Resistive Memory Based on Al/Poly(3,4-ethylene-dioxythiophene):Poly(styrenesulfonate)/Al Structure. *Appl. Phys. Lett.* **2010**, *97*, 253301.
- Moller, S.; Perlov, C.; Jackson, W.; Taussig, C.; Forrest, S. R. A Polymer/Semiconductor Write-Once Read-Many-Times Memory. *Nature* **2003**, *426*, 166–169.
- Jeong, H. Y.; Kim, J. Y.; Yoon, T. H.; Choi, S.-Y. Bipolar Resistive Switching Characteristics of Poly(3,4-ethylenedioxythiophene):Poly(styrenesulfonate) Thin Film. *Curr. Appl. Phys.* **2010**, *10*, e46–e49.
- Ha, H.; Kim, O. Electrode-Material-Dependent Switching Characteristics of Organic Nonvolatile Memory Devices Based on Poly(3,4-ethylenedioxythiophene):Poly(styrenesulfonate) Film. *IEEE Electron. Device Lett.* **2010**, *31*, 368–370.
- Dongjin, L.; Sungsik, B.; Moonhor, R.; Ohyun, K. Effect of the Electrode Material on the Electrical-Switching Characteristics of Nonvolatile Memory Devices Based on Poly(*o*-anthranilic acid) Thin Films. *IEEE Electron Device Lett.* **2008**, *29*, 694–697.
- Mukherjee, B.; Pal, A. J. Tuning of Electrical Bistability in Organic Devices through Electrochemical Potential of Metal Contacts. *Org. Electron.* **2006**, *7*, 249–255.
- Wang, Z.; Zeng, F.; Yang, J.; Chen, C.; Pan, F. Resistive Switching Induced by Metallic Filaments Formation through Poly(3,4-ethylene-dioxythiophene):Poly(styrenesulfonate). *ACS Appl. Mater. Interfaces* **2011**, *4*, 447–453.
- Joo, W.-J.; Choi, T.-L.; Lee, K.-H.; Chung, Y. Study on Threshold Behavior of Operation Voltage in Metal Filament-Based Polymer Memory. *J. Phys. Chem. B* **2007**, *111*, 7756–7760.
- Khan, M. A.; Bhansali, U. S.; Alshareef, H. N. High-Performance Non-volatile Organic Ferroelectric Memory on Banknotes. *Adv. Mater.* **2012**, *24*, 2165–2170.
- Bhansali, U. S.; Khan, M. A.; Alshareef, H. N. Electrical Performance of Polymer Ferroelectric Capacitors Fabricated on Plastic Substrate Using Transparent Electrodes. *Org. Electron.* **2012**, *13*, 1541–1545.
- Khan, M. A.; Bhansali, U. S.; Zhang, X. X.; Saleh, M. M.; Odeh, I.; Alshareef, H. N. Doped Polymer Electrodes for High Performance Ferroelectric Capacitors on Plastic Substrates. *Appl. Phys. Lett.* **2012**, *101*, 143303.
- Bhansali, U. S.; Khan, M. A.; Alshareef, H. N. Organic Ferroelectric Memory Devices with Inkjet-Printed Polymer Electrodes on Flexible Substrates. *Microelectron. Eng.* **2013**, *105*, 68–73.
- Cölle, M.; Büchel, M.; de Leeuw, D. M. Switching and Filamentary Conduction in Non-volatile Organic Memories. *Org. Electron.* **2006**, *7*, 305–312.
- Choi, J. W.; Han, M. G.; Kim, S. Y.; Oh, S. G.; Im, S. S. Poly(3,4-ethylenedioxythiophene) Nanoparticles Prepared in Aqueous DBSA Solutions. *Synth. Met.* **2004**, *141*, 293–299.
- Zhang, X.; Chang, D.; Liu, J.; Luo, Y. Conducting Polymer Aerogels from Supercritical CO₂ Drying PEDOT-PSS Hydrogels. *J. Mater. Chem.* **2010**, *20*, 5080–5085.
- Crispin, X.; Jakobsson, F. L. E.; Crispin, A.; Grim, P. C. M.; Andersson, P.; Volodin, A.; van Haesendonck, C.; Van der Auweraer, M.; Salaneck, W. R.; Berggren, M. The Origin of the High Conductivity of Poly(3,4-ethylenedioxythiophene)–Poly(styrenesulfonate) (PEDOT–PSS) Plastic Electrodes. *Chem. Mater.* **2006**, *18*, 4354–4360.
- Xu, X.; Register, R. A.; Forrest, S. R. Mechanisms for Current-Induced Conductivity Changes in a Conducting Polymer. *Appl. Phys. Lett.* **2006**, *89*, 142109.