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Research Article

Environmentally Friendly Resistive Switching Memory Devices with DNA as the Active Layer and Bio-Based Polyethylene Furanoate as the Substrate

Jeun-Yan Lam, Guang-Way Jang, Cheng-Jyun Huang, Shih-Huang Tung,* and Wen-Chang Chen*

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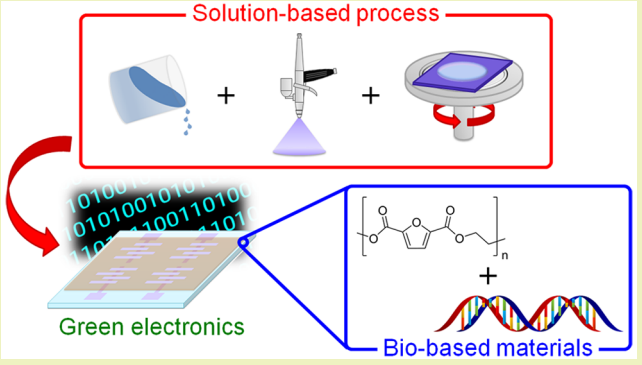
ABSTRACT: The development of flexible electronics for wearable or implantable devices has become an exciting research area in recent years. With the transition from rigid to flexible devices, polymeric materials, in particular the fossil-based PET, have been extensively used as the device substrate. For the environmental sustainability reason, biobased products have drawn much attention as a green replacement for fossil-based polymers. In this work, poly(ethylene furanoate) (PEF), a 100% biobased polyester, was utilized to replace PET as the substrate, and the biopolymer, deoxyribonucleic acid (DNA), was applied as the active layer to form the all-polymer resistive switching memory devices that are fully solution processable. The devices demonstrated the write-once-read-many-times (WORM) memory behavior with a low threshold voltage of approximately −2 V, an ON/OFF current ratio as high as 10^4, and a data retention time over 10^4 s. No noticeable degradation was observed under bending with various radius of curvature and after 1000 cycles of bending, suggesting an excellent endurance against severe and repeated deformation.



KEYWORDS: *Polyethylene furanoate (PEF), Biobased, Deoxyribonucleic acid (DNA), Solution processable, Flexible resistive switching device*

INTRODUCTION

The development of Internet of Things (IoT), big data, and cloud computing boosts the connection between cyberspace and the physical world, enabling seamless communication flows between diverse devices through digital networks at an unprecedented scale. The explosive growth of digital data has led to an urgent demand for high-density data storage components. With the trend of miniaturization of electronic components, the downscaling of transistors is inevitable to hit the physical limitation. Therefore, conventional transistor memory devices can no longer be satisfied in future memory applications.[1] Among the memories that have emerged as the next-generation nonvolatile information storage devices, resistive switching memory which possesses low power consumption and high data density in a simple device structure is considered as one of the most promising technologies.[2,3] At the same time, the rise of worldwide environmental consciousness promotes the establishment of a sustainable society. The large consumption of nonrenewable products, especially petrochemical-derived ones, has brought about the concerns of resource depletion and environmental pollutions. In this regard, the use of renewable and abundant biobased feedstocks to replace fossil fuel-based plastics is a solution to these increasingly serious issues. As a result, there is a growing demand for developing renewable materials to be employed in high-performance memory devices that are simultaneously environmentally friendly.

Several biobased materials, such as cellulose,[4] polylactide (PLA),[5] and polyethylene furanoate (PEF),[6] have been implanted into devices as a green replacement of petroleum-sourced substrates for flexible device applications. For instance, our previous work replaced one of most common petrochemical-derived polyesters, polyethylene terephthalate (PET), with PEF as a substrate for flexible optoelectronic devices.[6] As a substitute of PET, biobased PEF shows a higher glass transition and a lower melting point and has several advantages over PET, including improved mechanical properties and increased gas barrier capability.[7,8]

Apart from green substrates, biomolecules have opened new perspectives in electronic devices. Recently, interests have been

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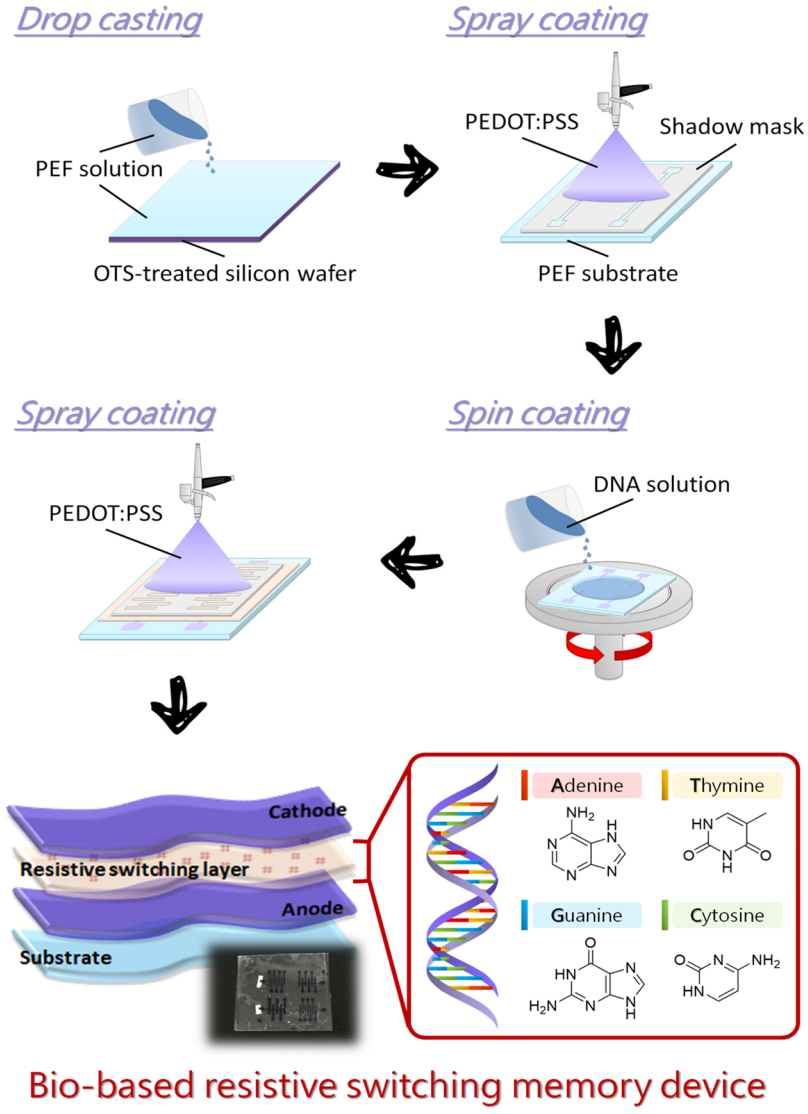
Scheme 1. Fully Solution-Processed Route toward a Biobased All-Polymer Resistive Switching Memory Device



aroused in the introduction of biomolecules into biocompatible bioelectronics as active layers. These electronic components are renewable and biodegradable, which can lower the negative environmental impacts from the greatly increasing electronic waste. Several natural biomaterials have been employed in resistive switching memory devices, including cellulose,[9] starch,[10] aloe vera,[11] chitosan,[12] silk,[13] pectin,[14] protein,[15] and enzyme.[16,17] In addition, the most important genetic material of living organisms, deoxyribonucleic acid (DNA), has been studied not only in genetic engineering and molecular nanotechnology[18] but also in the usage as a material in various applications due to its biocompatible and biodegradable nature.[19] DNAs are double-stranded helices in which the intertwined deoxyribose-phosphate backbones are hydrogen-bonded through nitrogenous bases. Although the mechanism of the carrier transport in its structure is a matter of controversy,[20] the advantages of DNA, including being mechanically solid, chemically stable, compatible with polymers, and processable into thin films,[21] have made it as a promising candidate for optoelectronics[22,23] and memory devices.[24,25]

Despite that the biomolecules perform satisfactorily as the active layer in information storage, the electrodes of the devices are usually metals and conducting oxides such as indium tin oxide (ITO). Generally, these kinds of devices are not competent in applications with large deformation due to the low flexibility of the inorganic electrodes under mechanical stress.[26] Furthermore, the inorganic electrodes are processed by relatively high-cost vacuum deposition. In contrast, all-polymer memory devices with conducting polymers as electrodes that can be fabricated by solution processes are competitive due to the high flexibility, tunable functionality, scalability, low-temperature process, and low cost.[27,28] In addition, for flexible electronic devices, especially for the bioelectronics that are wearable or implantable, not only the flexibility but the biocompatibility is often taken into consideration in order to minimize the possibility of undesired and potentially dangerous contact between electronics and living tissues.[29] The conducting polymer, poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS), has been widely used in organic electronics as interlayers or as transparent electrodes. It has been proven to be biocompatible[30,31] and therefore is suitable for manufactur-

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ing polymer-based electronic devices to be used in biorelated applications. With respect to resistive switching memory applications, although PEDOT:PSS has been studied as an active layer that shows a WORM memory behavior,[32,33] it is still relatively less studied as electrodes.

In this work, we develop flexible biobased resistive switching memory by utilizing 100% biobased PEF substrate that performs comparably as petroleum-based PET. The all-polymer flexible resistive switching memory devices were fabricated with biomolecules, including DNA, ribonucleic acid (RNA), or polyamino acid (poly-L-proline, herein), as the active layers and PEDOT:PSS as the electrodes. All the components are solution-processable. Such memory devices show low threshold voltages, high ON/OFF current ratios, great data retention ability, and excellent durability upon repeated bending tests, demonstrating a promising potential for wearable and implantable bioelectronic applications.

EXPERIMENTAL SECTION

Materials. Polyethylene furanoate (PEF) powders were provided by the Industrial Technology Research Institute, Taiwan. PEDOT:PSS (PH1000) was purchased from Uni Region Biotech. Dimethyl sulfoxide (DMSO) and methanol were purchased from Sigma-Aldrich. The fluoro-surfactant (Zonyl FS-300) was purchased from Fluka Analytical. DNA sodium salt was purchased from MP Biomedicals, LLC. RNA and poly-L-proline (PLP) were purchased from Sigma-Aldrich. All materials and solvents were used as received.

Fabrication and Characterization of Flexible Resistive Switching Memory Devices. The memory devices were fabricated with a DNA film sandwiched between two electrodes. For the rigid device, a silicon wafer was first precleaned by ultrasonication with toluene, acetone, and isopropyl alcohol, each for 15 min successively, and dried under nitrogen. Prior to deposition of the bottom electrode, Si substrates were subjected to plasma cleaning. The bottom electrode is a patterned Al layer with a thickness of 100 nm deposited via thermal evaporation on the wafer. The DNA layer was deposited onto the bottom electrode by spin coating. DNA solutions were prepared as follows. First, 27 mg of DNA sodium salt was first dissolved in 1 mL of deionized water, and after the mixture stirred for 6 h, 2 mL of methanol and 1.5 μL of Zonyl were added to dilute the DNA concentration to ~9 mg mL^{-1}. The spin coating was operated at a speed of 800 rpm for 90 s. Finally, the top Al electrodes with 30 nm thickness were deposited through a patterned metal mask.

For the fabrication of the flexible devices, the cleaning process of the Si substrate, oxygen-plasma step, and thermal evaporation of Al electrodes were omitted. PEF films were drop-casted on the OTS-treated silicon wafer following the method described in a previous report[6] and were utilized as the substrate. The top and bottom electrodes were made by spray-coated PEDOT:PSS. First, 20 mL of PEDOT:PSS (PH1000) was pretreated by mixing it with 1 mL of DMSO to increase the conductivity. Then, 100 μL of Zonyl surfactant was also added into the solution, followed by stirring overnight at room temperature. The solution was loaded into the spray-coating machine after it was filtered with a PTFE membrane of 0.45 μm pore size. PEF substrate was placed 20 cm below the spray nozzle and was kept at 50 °C. The line PEDOT:PSS electrodes were then spray-coated on the PEF substrate using a shadow mask. Subsequently, a ~90 nm layer of DNA was spin-coated onto the PEDOT:PSS bottom electrode by the same procedures used in the rigid devices. The top PEDOT:PSS electrode lines aligned perpendicular to the bottom lines were then spray-coated onto the active layer with a shadow mask, leading to cross-point arrays of memory cells. The all-solution fabrication process of the devices is illustrated in Scheme 1. For the bending tests, the flexible devices were transferred onto poly(dimethylsiloxane) (PDMS) support. PDMS (Sylgard 184 silicone elastomer, Dow Corning) was cured at 60 °C for 12 h at a 20:1 base/cross-linker weight ratio. The PEF-based devices were then attached to PDMS. The separation of the PEF-based devices from PDMS support after testing caused no damage to the devices due to the weak interaction between PEF and PDMS.

All the measurements of the electrical characteristics for the resistive memory devices were carried out with a Keithley 4200-SCS semiconductor parameter analyzer and a probe station in a N_2-filled glovebox at room temperature.

RESULTS AND DISCUSSION

Figure 1a and S1a show the current−voltage (I−V) curves in a semilogarithmic scale of the memory devices on PEF flexible

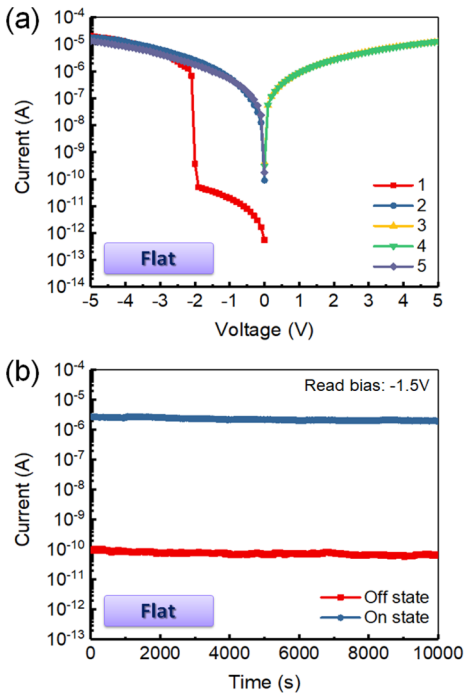

Figure 1. (a) I−V characteristics and (b) data retention test of PEF-based DNA resistive switching memory devices under flat conditions.

substrates (PEDOT:PSS electrodes) and rigid SiO_2/Si wafers (Al electrodes) with DNA as the active layers, respectively. While applying a voltage sweep in the negative direction (sweep 1), the current sharply increases at approximately −2 V, which is the threshold voltage where the high resistance state (HRS), i.e., OFF state, is switched to the low resistance state (LRS), i.e., ON state. This is referred to as the writing process in the memory devices. Note that the writing process could also be realized in the positive sweep (Figure S2). Once the resistance is switched, the ON state remains during the following voltage sweeps (sweep 2 and 3) and cannot be restored to the OFF state by applying voltages in either direction. The ON state can be preserved even after the electrical power is discontinued (sweep 4 and 5). The switch process between the OFF and ON state is irreversible and nonvolatile, with an ON/OFF ratio up to 10^4, demonstrating the feasibility of the DNA film as the active layer and PEDOT:PSS as the electrodes in all-polymer resistive switching memory devices that exhibit WORM memory behaviors.

The data retention performance of the PEF-based memory device was evaluated by measuring the currents in OFF and ON states with time at room temperature, as shown in Figure 1b. The retention of the rigid wafer-based memory device is shown in Figure S1b. No electrical power is required to retain the resistances in the OFF and ON states. The currents of the

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two states obtained at a constant reading voltage of −1.5 V are nearly unchanged during the test for at least 10^4 s, indicating a good retention capability of the memory devices. The data storage is expected to last even longer, considering that the currents in Figure 1b are essentially time independent.

Figure 2a shows the energy band diagram of the carrier transitions for the PEF-based flexible memory device. Under an external electric field, the charges could be trapped and transported in the DNA active layer through the nucleobase moieties on the DNA chains.[34] The ON−OFF state transition can be explained by comparing the energy levels of DNA and the PEDOT:PSS electrodes. The highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO) energy levels of natural DNA are around −5.30 and −1.08 eV, while those of PEDOT:PSS are −5.20 and −3.20 eV, respectively.[35−37] The HOMO levels of DNA and PEDOT:PSS are rather close, implying a lower energy barrier for the holes to inject from the electrode into the active layer. In contrast, the energy gap between the LUMO levels of DNA and PEDOT:PSS is higher, so that the electrons are less probable to inject through the interface compared with the hole injection. Therefore, it is the hole injection that triggers the electrical conduction processes in the DNA memory devices. Since the electron injection is limited by the large energy barrier, the ON state caused by the hole injection is hardly erased through the charge recombination, which explains the WORM behavior of the memory devices.

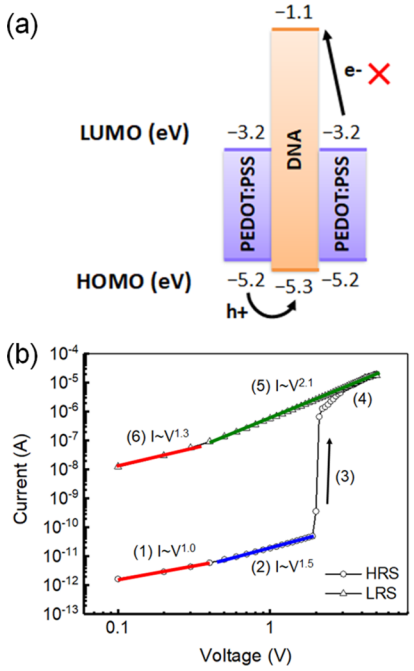

Figure 2. (a) Energy band diagram of the carrier transitions and (b) double-logarithmic plot of $I-V$ characteristics corresponding to the operating mechanisms for the PEF-based flexible memory device.

To further understand the conduction mechanism of the memory devices, the $I-V$ curves are replotted on double-logarithmic scales and the linear fits of the curves are depicted in Figure 2b. The fitting of the $I-V$ curves suggests that the space-charge-limited current (SCLC) is likely the dominant conduction mechanism in the HRS and LRS. At low applied voltage in HRS (regime 1) where the free charge carriers are thermally generated, the current is linearly proportional to the voltage ($I \propto V$), following Ohm's law. The current density J can be described by[38,39]

$$J = \frac{qn_0\mu V}{d}$$

where q is the elementary charge of the carrier, n_0 is the free charge carrier density, μ is the charge carrier mobility, V is the voltage, and d is the layer thickness. As the applied voltage is

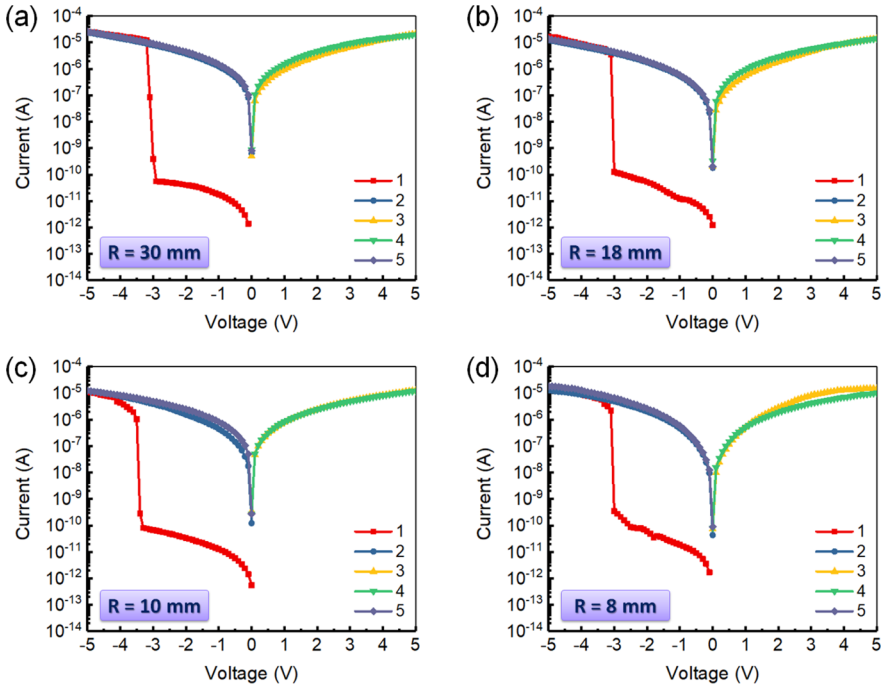

Figure 3. $I-V$ characteristics of PEF-based DNA memory device at various bending conditions.

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increased, SCLC arises at the intermediate bias region (2) when the current through the insulator becomes limited by the build-up of the charges injected from the source electrode. In this regime, the current is determined by the traps of the charge carriers in the active layer. According to the SCLC theory, a typical space-charge effect leads a square law dependence of the current on the voltage ($I \propto V^2$) after the initial Ohmic region. However, the $I-V$ characteristics depend on the types of the charge carrier traps. For instance, the slope ≈2 might not be observed in this regime for deep traps, in contrast to trap-free or shallow trap SCLC.[40,41] When a large percentage of carriers are injected into the unfilled deep traps, they cannot contribute to the conduction. Thus, the slope ∼1.5 in this regime could be a result of the discrete deep-level traps in the DNA layer. The trapping sites below the conduction band could be formed by the defects in the DNA layer or may be related to the chemical composition of the DNA chains, such as the carboxyl group and the amino linkage that have been shown to act as the nucleophilic or electrophilic sites.[42]

Once the traps are almost filled, a sharp rise in the current of a slope much larger than 2 occurs in the trap-filled limit (TFL) regime (3) and the device is switched from the HRS to LRS. After the abrupt transition, the current in the trap-free SCLC regime (4) shows a square law dependence on voltage, and it is not affected by the traps in the active layer. During the back voltage sweep in the LRS, the current is controlled by SCLC with shallow traps, and the slopes are 2.1 and 1.3 in the high (5) and low (6) bias region, respectively. The nonlinear characteristics suggest that the conduction is bulk-limited SCLC, different from the conduction caused by the metallic filament which exhibits linear $I-V$ characteristics.[43] In contrast, the rigid device with Al as the electrodes shows a linear-like characteristics with slope close to 1 in the LRS (inset of Figure S1a), indicating that the current conduction might be dominated by different mechanisms such as conductive filament model.[44−46] Note that the conduction mechanism of the flexible devices using PEDOT:PSS as electrodes could be complicated since PEDOT:PSS itself also exhibits resistive switching effect.[33]

To confirm the feasibility of the all-polymer memory devices in flexible electronic applications, the memory behaviors under the bending condition were investigated. PDMS was used as the supporting substrate for the bending tests. The samples were manually bent into curves with the radii (R) of the devices at 30, 18, 10, 8, and 5 mm (Figure S3), and no cracks occurred for all the devices upon bending. The corresponding resistive switching characteristics of top-performance devices are shown in Figure 3a-d and 4a. The devices under different bending radii exhibit similar WORM-type memory behaviors and their threshold voltages are statistically comparable to that in the flat condition. The data retention time of the device bent at R = 5 mm (Figure 4b) is also similar to that of the flat device. Note that since the LRS cannot be recovered to the HRS for the devices, the $I-V$ characteristics were taken from different devices under desired conditions. Thus, the performance might be affected by the variation of the film uniformity between the devices.

We also fabricated the devices utilizing RNA and poly-L-proline as the active layers on PEF substrate, and the characteristics of the memory devices are shown in Figures S4 and S5, respectively. In the flat condition, a similar resistive switching behavior could be observed by using RNA and poly-L-proline as the active layers. However, the HRS current of the

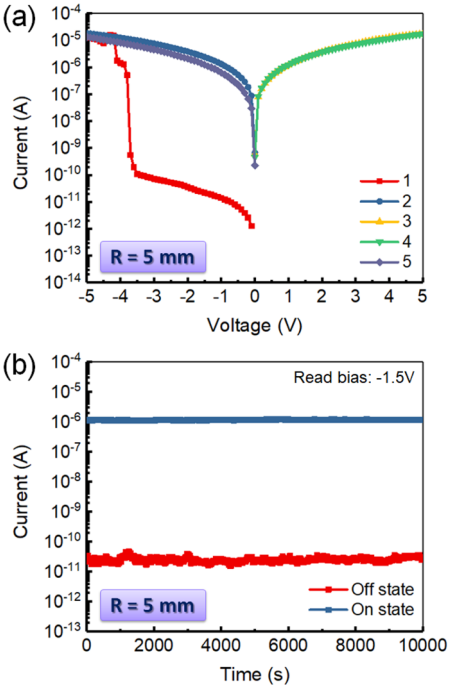

Figure 4. (a) $I-V$ characteristics and (b) data retention test of PEF-based DNA resistive switching memory devices under R = 5 mm condition.

RNA device bent at R = 5 mm shown in Figure S4c is slightly higher than that under the flat condition. This might be due to the decrease in the thickness of the active layer when it is severely bent. The mechanical properties of the single-stranded RNA would be lower in comparison to DNA and a nonuniform deformation may occur for RNA layer upon bending. A larger threshold voltage in poly-L-proline-based devices with a slight fluctuation of $I-V$ curve during the voltage sweep in the OFF state are possibly caused by the more hydrophobic nature of poly-L-proline that suppresses the contact with the hydrophilic electrodes and the lower water solubility of poly-L-proline that affects the uniformity of the spin-coated active layer. Both RNA and poly-L-proline yield similar performance in data retention test, reflecting the merit of using them as the active layers for long-term data storage devices.

Figure 5a shows the current variation as a function of bending radius. The currents in the HRS and LRS can stably maintain at the bending radius down to 5 mm, revealing that the DNA memory device using PEF as a substrate is reliable even under severe bending. The device was repeatedly bent to ∼5 mm radius performed by a machine to evaluate the mechanical endurance of the flexible memory device. As seen in Figure 5b, even though the currents in the HRS and LRS slightly fluctuated, no noticeable change was observed up to 1000 times of bending, manifesting excellent reliability in endurance. Similar results could also be observed in the RNA and poly-L-proline devices as shown in Figures S6 and S7, respectively. The above results demonstrate that coupling with PEF as the substrate, the biopolymers, especially the DNA, are promising as the active layers for flexible nonvolatile memory applications.

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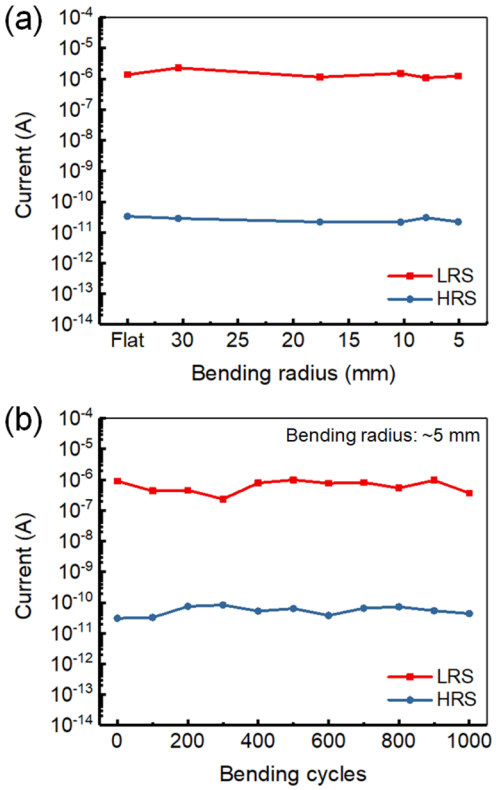

Figure 5. (a) Current variation as a function of bending radii and (b) bending endurance of PEF-based DNA memory device under $R \sim 5$ mm.

CONCLUSIONS

In this work, PEF film was utilized as the substrate and biopolymer DNA was applied as the active layer for the fabrication of flexible all-polymer resistive switching memory devices via a solution process. The devices show the write-once-read-many-times (WORM) memory behavior with a low threshold voltage, a high ON/OFF current ratio, and a long data retention trend. The analyses of the operation and conduction mechanism suggest that the currents of the flexible devices were dominated by the space charge limit current (SCLC) conduction. In addition, the devices exhibit excellent endurance against severe bending conditions and continuous bending operations. PEF and DNA are shown as promising materials for flexible resistive switching device applications that can simultaneously achieve an environmentally friendly purpose.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acssuschemeng.9b07168.

I–*V* characteristics and data retention test of rigid DNA-based, flexible RNA-based, and polyamino-based resistive switching memory devices (PDF)

AUTHOR INFORMATION

Corresponding Authors

Shih-Huang Tung − *Institute of Polymer Science and Engineering and Advanced Research Center for Green Materials Science and Technology, National Taiwan University, Taipei 10617, Taiwan;* orcid.org/0000-0002-6787-4955; Email: shtung@ntu.edu.tw

Wen-Chang Chen − *Institute of Polymer Science and Engineering, Advanced Research Center for Green Materials Science and Technology, and Department of Chemical Engineering, National Taiwan University, Taipei 10617, Taiwan;* orcid.org/0000-0003-3170-7220; Email: chenwc@ntu.edu.tw

Authors

Jeun-Yan Lam − *Institute of Polymer Science and Engineering, National Taiwan University, Taipei 10617, Taiwan*

Guang-Way Jang − *Material and Chemical Research Laboratories, Industrial Technology Research Institute, Hsinchu 30011, Taiwan*

Cheng-Jyun Huang − *Material and Chemical Research Laboratories, Industrial Technology Research Institute, Hsinchu 30011, Taiwan*

Complete contact information is available at: https://pubs.acs.org/10.1021/acssuschemeng.9b07168

Notes

The authors declare no competing financial interest.

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