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Non-volatile resistive memory devices based on solution-processed natural DNA biomaterial

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ABSTRACT

Biomaterial-based devices have demonstrated versatility in various applications and have drawn recent considerable attention. In this study, we present non-volatile resistive switching memory devices based on a natural-derived DNA biomaterial. The structure consists of a DNA-based biomaterial layer sandwiched between two electrodes, where the DNA-based biomaterial is solution-processed without sequence control or external doping of nanoparticles. The fabricated device was tested at room temperature without encapsulation and exhibited a reliable resistive switching behavior and multi-level operation with low switching voltages, 10^4 s retention time, and more than 200 cycles in memory endurance testing. Our demonstration shows that reliable memory devices can be realized using only one layer of natural DNA biomaterial, without the need for composite layers. Our characterization also provides underlying physics that may be exploited for the design and fabrication of natural DNA-based optoelectronics.

1. Introduction

Motivated by the demand for ever larger storage capacity in the information era, research efforts have been devoted to the development of more efficient and cost-effective memory elements. Many digital data storage infrastructures are constructed based on the building block with a resistive switching (RS) behavior, where resistances can be reversibly changed by applying different voltages. So far, the RS effect has been demonstrated in many metal oxide and organic materials, such as SiO_2, HfO_2, P3HT, PVK, etc. [1–3] The use of biomaterials in electronic devices has also recently drawn considerable attention, driven by the rapid development of technology coupled with the growing interest toward green electronics [4]. Biomaterials are abundant, eco-friendly, and suitable for large-area implementation, which are of great interest for applications such as flexible displays and wearable technologies [5]. As green electronics continue to advance, the development of a facile approach to fabricate a biomaterial-based memory device becomes critical to pave the way for implementation of low-cost, green electronic devices. Previously there have been several studies that present the use of biomaterials for resistive memory devices [6–10]. In these approaches, some involve DNA sequence control, while others may require external doping of guest components. The addition of nanoparticles, for example, provides an easy route to tune the electrical properties of the composites. However, uniform dispersion and compatibility of the hybrid composites may be difficult to control. Furthermore, some reported devices need to be operated under controlled environmental conditions. These features increase complexity when it comes to practical implementation and device integration.

In this study, we report the fabrication of non-volatile resistive switching devices based on natural DNA biomaterial. The raw DNA material is isolated from salmon milt, which is randomly sequenced with a wide range of distribution of base pairs. Such DNA can be readily extracted from biological species and is abundant in nature. In the present device, the structure consists of a spin-coated DNA layer sandwiched by two electrodes without DNA sequence control or external doping of nanoparticles. The fabricated devices show a reliable resistive switching behavior with low switching voltages, data retention longer than 10^4 s, and more than 200 times in memory endurance under ambient conditions. The devices also show multi-level memory characteristics, in which the current levels can be controlled by reset voltages. To study the underlying switching mechanisms, the electrical properties are examined under different fabrication parameters and measurement conditions. Our demonstration shows that reliable memory devices operated under ambient conditions can be realized

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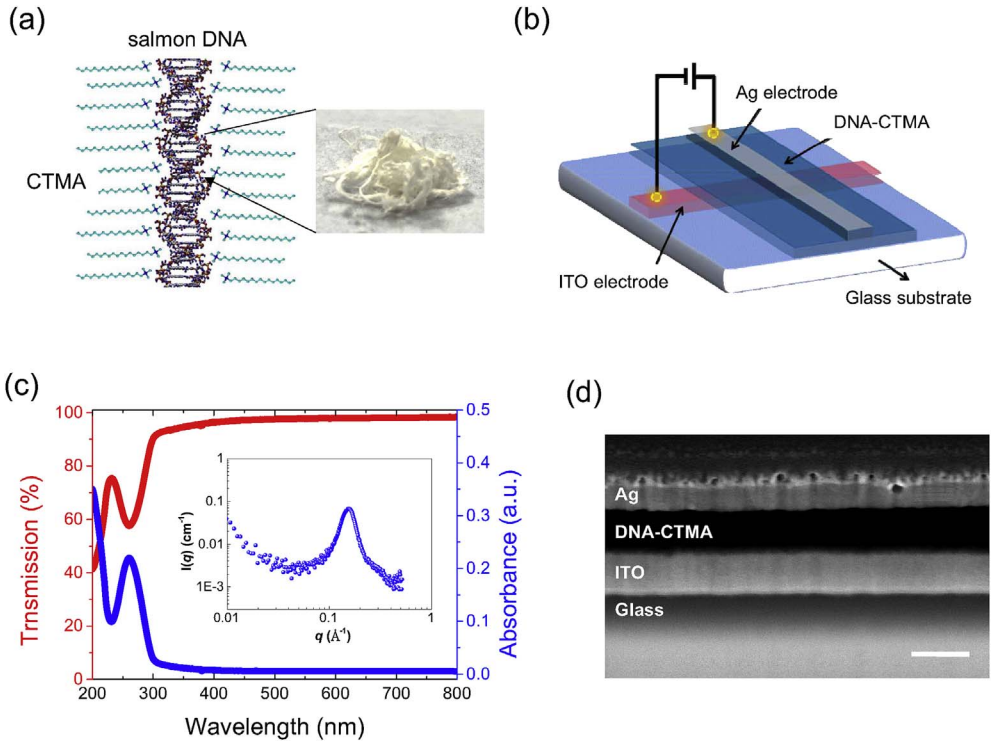

Fig. 1. (a) The chemical structure of DNA-CTMA. The inset is a photo of the raw DNA material extracted from salmon. (b) A schematic illustration of the device structure. The DNA layer is sandwiched between the ITO and Ag electrode. (c) Transmission and absorbance of the DNA film. Inset shows the SAXS profile of the DNA film. A broad scattering peak is located at $q = 0.154 Å^{-1}$. (d) Cross-sectional SEM image of the fabricated device. The laminated structure can be observed. The scale bar is 200 nm.

based on one layer of solution-processed natural DNA biomaterial, without the use of composites or hybrid schemes. The ease of material handling and device fabrication may lead to future development for biomaterial-based multifunctional devices or green electronic devices.

2. Experimental

DNA is hydrophilic due to the sugar-phosphate backbone. A facile and efficient approach has been developed to adapt natural DNA materials to be compatible with solvent-based thin film technology. In this approach, a cationic surfactant, cetyltrimethylammonium (CTMA) chloride, is used to form DNA-CTMA macromolecules as illustrated in Fig. 1 (a), rendering DNA-CTMA soluble in organic solvents and more robust for thin film applications [11]. The inset is a photo of the fiber-like purified salmon DNA, purchased from GEM. DNA was processed following the sonication dialysis/Soxhlet rinsing procedures provided in detail in a previous publication [12]. The dialysis/Soxhlet rinsed DNA-CTMA was dissolved in ethanol and mixed for one day at room temperature, using a magnetic stirrer. The DNA-CTMA solution was then spin-coated on a patterned indium tin oxide (ITO) substrate at 2000 rpm for 20 s. A top silver (Ag) electrode with a thickness of 100 nm was deposited by thermal evaporation at a vacuum pressure of 4×10^{-6} Torr. Fig. 1(b) is an illustration of the device structure. The optical properties of the film were measured by a UV-VIS spectrometer (Lambda 35, PerkinElmer). The SAXS measurement was performed at National Synchrotron Radiation Research Center (NSRRC). The cross-sectional SEM image of the device structure was prepared and measured by a focused ion beam system (600i system, FEI Helios NanoLab). The current-voltage characteristics were measured using a probe station by a source meter (ADCMT 6241A). All the electrical measurements were carried out under ambient conditions without encapsulation of the devices. For simplicity, the DNA-CTMA layer is denoted hereafter as the DNA layer.

3. Results and discussion

3.1. Material characterization

The optical properties of the DNA layer on a quartz substrate are shown in Fig. 1(c). The transmission spectrum shows that the film exhibits a high transparency (>90%) over a broad wavelength range above 300 nm. An absorption peak can be found at ~260 nm, indicating the characteristic feature of heterocyclic rings of DNA molecules. The inset is the SAXS measurement of the DNA film. One broad scattering peak at $q = 0.154 Å^{-1}$ can be observed without the higher-order peaks, which is consistent with the previous study of DNA-lipid complexes with a charge molar ratio of 1 [13]. This implies that DNA molecules undergo an irregular parallel packing with a characteristic nearest-neighbor distance [14]. From the peak position of the SAXS measurement, the nearest-neighbor distance between the DNA strands can be calculated to be 40.8 Å [15]. Fig. 1(d) shows a cross-sectional SEM image of the fabricated device. The laminated structure can be clearly seen, and the thicknesses of the Ag electrode, DNA layer, and ITO substrate are determined to be 100 nm, 200 nm, and 160 nm, respectively.

3.2. Electrical properties of the devices

3.2.1. I-V characteristics

A representative current-voltage (I-V) behavior of the device is plotted in Fig. 2, showing a bipolar resistive switching behavior. The pristine device is initially at a high resistance state (HRS, OFF-state). When the applied voltage increases (Sweep 1), an abrupt current change is displayed under a threshold voltage (V_{set}) of 0.65 V, indicating the device is switched from a HRS to a low resistance state (LRS, ON-state). When the sweeping voltage is back to zero, the device remains in the LRS (Sweep 2). For sweeping at negative bias, the device remains in the LRS for smaller voltages, followed by a section with negative differential resistance (NDR), at a reverse voltage (V_{reset}) of −1.25 V