Rectifying memory is an effective strategy to solve the cross-talk issues of crossbar switch architecture during the integration procedure for high-density data storage. Herein, a two-dimensional (2D) conjugated microporous polymer (CMP) is proposed as an ideal resistive switching medium for integrated rectifying memory devices. Large-area 2D CMP film is prepared using a surface-confined polymerization method, featuring rewritable nonvolatile memory with a low set voltage in a graphene/CMP/metal diode. Importantly, this film exhibits outstanding structural stability during solvent immersion and other polymer coating, being suitable for solution-processed multilayer polymer electronics. By directly stacking the CMP memristor with a poly(3-hexylthiophene) rectifier, the integrated multilayer device shows desirable rectifying memory effect, effectively reducing the misreading issue induced by the cross-talk in a crossbar array.

Resistive random access memory devices with synchronous rectifying effect, namely rectifying memory, are highly desirable for crossbar switch architecture toward high-density data storage, due to their ability to mitigate the cross-talk issue and reduce the misreading operation. Typically, rectifying memories are realized by integrating each memory cell with a complementary switching element such as rectifying diode and transistor to create one diode–one resistor (1D1R) structure and one transistor–one resistor (1T1R) structure. Among them, the former 1D1R system, especially based on polymer materials, presents the advanced features of simpler configuration, greater scalability and flexibility, easier procedure and low-cost potential than the 1T1R structure. To produce vertical stacked polymer-rectifying memory, the first polymer-active material should be processed into thin film to facilitate device fabrication, and this film must be immune to wet coating of the second successive layer from polymer solutions without erosion damage. The erosion in a polymer coating normally leads to a low yield, poor device performance, and reproducibility. The traditional solution is blending polymers with photoresist for photo-cross-linking to increase their solvent resistant. However, this approach is accompanied with complexity in the photolithography procedure. Consequently, using solvent-resistance polymers as active layers in polymer 1D1R devices is a charming approach in simplifying the fabrication procedures, whereas less attention has been paid in constructing polymer-active films with structural stability and electrical bistability for 1D1R device.

Two-dimensional (2D) conjugated microporous polymer (CMP) is a kind of emerging sheet-like nanomaterial with highly cross-linked porous structures. These cross-links grant CMPs excellent structural stability compared with conventional polymer films due to the network formation of covalent bond. Moreover, CMP films can be controllably prepared onto arbitrary substrates with wafer-scale size, nanoscale thickness, and patterned geometries. Furthermore, for organic porous frameworks, incorporating appropriate precursor units into frameworks can yield hybrid materials with desirable functions, for example, resistive switching induced by ions transport in porous structures under electrical field. This switching can also be achieved via filament mechanism in most sulfur-containing polymer films. Therefore, the combination of these factors can be used to design resistive-switching CMP for all-polymer integrated rectifying-memory devices, but their effects have not been studied systematically.
In this work, we propose that CMP is an attractive memristive switching medium for integrated polymer memory devices with rectifying function due to the combined merits of its cross-linked porous structure and versatile components. Large-area CMP nanofilms, featuring tunable nanoscale thickness, excellent structural stability, and desired elements, were successfully synthesized on arbitrary substrates, such as SiO$_2$/Si and reduced graphene oxide (rGO). The vertical rGO/CMP/Al device exhibits stable nonvolatile rewritable memory performance with low set and reset voltages, and long retention time. Due to its immune erosion in spin coating of second successive polymer layer, the vertically stacked all-polymer 1D1R crossbar arrays using poly(3-hexylthiophene) (P3HT) diodes show the expected self-rectifying memristor.

We commenced our study by dissolving 5,10,15,20-tetrakis(4-aminophenyl) porphyrin (Tapy) and thiophene-2,5-dicarbaldehyde (ThCHO) as monomer precursors in dimethylformamide (DMF) to create 2D CMP nanofilms on conductive rGO surface through Schiff-base reaction under ambient temperature (Figure 1a). The choice of porphyrin units is due to their novel electronic properties and unique chemical and thermal stability. In addition, the presence of heteroatoms (N and S) in polymer structures can coordinate with metal ions to form metal filament under external voltage, facilitating the electrical switching behaviors. In our typical experimental process, the rGO substrate was submerged in monomer solvents. These dissolved precursors were attached on rGO surface because of the π-π interaction or other weak interaction between monomers and rGO. Upon the addition of acetic acid catalyst, the imine condensation reaction was triggered and CMP films could be polymerized. The resulting films were fully washed with DMF to remove the residual monomers and oligomers. Then the residual DMF in the CMP films was exchanged with ethanol; a pristine CMP film was achieved by drying for further characterization and device applications.

Figure 1b–e schematically shows the fabrication of CMP-based polymer 1R and 1D1R electronic devices (see details in Experimental Section). Briefly, after preparing the CMP films on patterned rGO electrodes (Figure 1b), Al interlayer with desired shape was deposited on CMP surface through shadow mask to prepare cross-point structure 1R device cells (Figure 1c). The solvent-resistant feature of CMP films benefits the subsequent preparation of P3HT film on CMP films by spin-coating method (Figure 1d). Then patterned Au electrodes were deposited onto P3HT film to fabricate 1D unit on 1R unit (Figure 1e). Finally, tandem structured all-polymer 1D-1R cell arrays were obtained via full solution process.

To explore the effect of initial monomer concentrations on film morphologies, the relationship between the monomer concentrations and film thicknesses was studied. It is found that the thickness of CMP films is precisely controlled by modulating the monomer concentration. When the original monomer concentrations (Tapy = 0.75 mmol L$^{-1}$, ThCHO = 1.5 mmol L$^{-1}$) were used, ultra-thin CMP films were formed on SiO$_2$/Si substrate and rGO (Figure S1a and S2a, Supporting Information). The thickness of CMP films on rGO surface is ≈6 nm (Figure S3a, Supporting Information). Upon double increasing the concentration, the thicknesses of CMP films were increased to 17 nm for 1.5 mmol L$^{-1}$ Tapy and 55 nm for 3.0 mmol L$^{-1}$ Tapy (Figure S3, Supporting Information). The color contrasts of CMP films strongly depend on the film thicknesses (Figure S1 and S2, Supporting Information). When we further increased the monomer concentrations (Tapy = 6.0 mmol L$^{-1}$, ThCHO = 12.0 mmol L$^{-1}$), the thickness of the as-prepared film only increased ≈3 nm (Figure S4, Supporting Information). A reasonable explanation for the aforementioned phenomenon

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**Figure 1.** Surface-confined preparation of large-area 2D CMP films and the fabrication of 1R and 1D1R devices. a) Schematic representation of the preparation of CMP films by the condensation of Tapy units (blue) and ThCHO units (red). b) CMP films grown on patterned rGO films covered substrates. c) The 1R device with sandwich architecture. d) Spin-coating of P3HT on the 1R device directly. e) The 1D1R device consists of a 1R device with the structure of rGO/CMP film/Al and a 1D device with the structure of Al/P3HT/Au.
is that the reaction was carried out at the rGO surface. In this case, only the monomers absorbed on the surface can form a CMP film. The reaction of the monomers in the solution was formed by solid powers (Figure S5, Supporting Information). Upon increasing the monomer concentration, more monomer would be absorbed on the rGO surface and thicker CMP films were formed. However, it is hard to absorb more monomer when the concentration of Tapy was higher than 3.0 mmol L\(^{-1}\). Therefore, the thickness of the film was only increased approximately 3 nm when we double increased the monomer concentration of Tapy to 6.0 mmol L\(^{-1}\). Interestingly, rGO template enables the growth of thicker CMP compared with traditional SiO\(_2\) substrate, with a thickness of approximately 55 and 43 nm under the same condition (Figure S6, Supporting Information), which might be attributed to the existence of \(\pi-\pi\) interaction force between the monomers and rGO. As a result, much more monomers could be absorbed onto rGO surface resulting in a thicker film.

The chemical structures of as-synthesized 55 nm thick CMP films were further characterized symmetrically. In the subsequent section, except for special emphasis, the CMP films with the same thickness were used for characterizations and applications. According to the photographs, the color variation on rGO surface allows for the differentiation between large-area CMP films covered region and bare region (Figure 2a). Impressively, the CMP film could be detached freely from substrate and transferred onto a transmission electron microscope (TEM) copper grid, indicating the excellent mechanical strength induced by the cross-linked microporous nature (Figure S7, Supporting Information). Atomic force microscopy (AFM) studies reveal homogeneous surface of CMP film on rGO template (Figure 2b). The CMP structures at a molecular level were studied by Fourier transform Infrared (FT-IR) spectral (Figure 2c). The disappearing of C–O band (1667 cm\(^{-1}\)) and N–H band (1620, 1606 cm\(^{-1}\)) in the two monomers while a new peak was formed at 1616 cm\(^{-1}\) in the CMP films revealed that the formation of C=N band, indicating the coupling reaction between the two monomers occurred successfully.\(^{[22]}\) In the UV–vis absorption spectra, the two monomers show absorption peaks at 296 and 436, 529, and 574 nm, respectively (Figure 2d). In comparison with the monomers, the absorption peak at 296 nm vanishes, yet a broader absorption peak at 446 nm is observed for CMP film, indicative of an increase in conjugation length of the polymer film.

To determine whether the CMP film work properly in facile solution process for the fabrication of vertically multilayer-stacked polymer architecture for integrated electronics, the chemical stability of porous structure was performed. The as-prepared CMP films were submerged in different common solvents to characterize their solvent resistant property. It is found that these films are insoluble in water, ethanol and most organic solvents, such as DMF, dichlorobenzene (DCB), dichloromethane (DCM), and toluene. The same physical appearance and SEM images indicate negligible alteration in film structure (Figure 2e,f). In addition, the effect of conjugated polymer coating on the structural integrity of CMP film was also investigated. A P3HT thin film was first spin-coated onto CMP films and then washed with organic solvent. It was found that the morphology of CMP films almost remains the same as the original film, indicating their outstanding immune ability of erosion during multilayer stacking (Figure 2g). Moreover, the UV–vis peaks of the submersed CMP films are also the same as the original one, confirming the excellent stability of the conjugated structure in soaking and rinsing process (Figure 2h). Thermogravimetric analysis double- indicated the thermal stability of the target materials. The
residual weight percentages were 96%, 93%, and 85% after heat treatment at 300, 400, and 500 °C, respectively, under nitrogen protective gas (Figure S8, Supporting Information). Thus, the solvent-treated CMP also exhibited high thermal stability. Therefore, we consider the CMP films are capable of high chemical and thermal stability, dictating its potentiality for integrated polymer electronics.

To explore the potential use of CMP films in the field of data storage, a device with a simple structure of CMP film sandwiched between bottom rGO electrode and top Al electrode was fabricated (Figure 3a). The memory performance has been investigated from the current-voltage (I–V) behaviors of fabricated device (Figure 3b). The I–V curves show a typical flash memory feature. In the first sweep, the current increased consecutively as the bias from 0 to 1.60 V and abruptly at a set voltage (≈1.65 V) (stages 1, 2). This process demonstrates the transition from a high-resistance state (HRS), denoted as “OFF state”, to low-resistance state (LRS), denoted as “ON state”, and subsequently maintain the ON state in a backward sweep from 3 to 0 V (stage 3), which is equivalent to the “Write” operation. Impressively, the device remained in the LRS during a negative sweep until the voltage approached −2.0 V and returned to initial OFF state (stages 4, 5), which is defined as the “Erase” process (stage 6). The observation of nonvolatile rewritable-resistive switching was highly stable in both ON and OFF states (Figure 3b,c). It is noted that the memory device show low power consumption; the statistical distribution of set and reset voltages ranged from 1.35 to 2.55 V and −1.20 to −2.30 V, with the center at 1.80 and −1.75 V, separately (Figure 3d). Moreover, the device poses negligible fluctuations in OFF and ON current during a test period of 10⁴ s under a read bias of 0.2 V, indicating its superior retention stability (Figure 3e). To study the carrier transport mechanism in each conductive state, the I–V curve in the positive sweep region was analyzed using log I-log V plots. The slopes of fitted lines are about 1.08 in ON state and 1.10 in OFF state, indicating the carrier transports were dominated by the ohmic conduction model (Figure 3f).²⁷

Figure 4a shows the schematic diagram of the integrated 1D1R device with rGO/CMP/Al/P3HT/Au vertically stacked structure, where Al/P3HT/Au is the Schottky diode and rGO/CMP/Al is the resistive memory cell. The P3HT diode was vertically integrated on top of the memory cells by directly spin-coating a P3HT layer over CMP film, benefiting from the resistance to chemical solvents of CMP film. The integrated diode can eliminate cross-talk issue between memory cells, which is caused by parasitic current paths in a cross-bar array architecture.³⁹ The I–V curve of Schottky diode exhibited a typical rectifying behavior with a rectification ratio of 4.6*10² at ±3 V (Figure 4b). The I–V characteristics of each memory cell also exhibit typical flash memory-switching behavior (Figure 4c). As the applied bias increased from 0 to 4 V, the device was switched from an initial HRS to LRS at the set voltage (≈2.75 V). The device maintained the ON state during the subsequent reverse sweep and returned to OFF state as negative bias reached −2.15 V. The I–V curves of memory cell in 1D1R device is almost the same as the 1R-only cell in Figure 3b, indicating that vertically integrated, a Schottky diode on the memory cell did not sacrifice the electrical properties of the memory device. The I–V characteristics of integrated 1D1R device clearly show desired rectifying memory effect via the cooperation of the rectifying diode and memory cells (Figure 4d).

Similar to the 1R cell, the I–V curve of memory cell in integrated device retained its resistive-switching memory performances in

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positive bias regime due to the ohmic contact between the P3HT layer (Highest Occupied Molecular Orbital, HOMO: 4.9 eV) and Au electrode (work function: 5.1 eV). The current increased sharply when the forward voltage was increased to 3.6 V, indicating a shift from HRS to LRS. In comparison with the 1R-only cell, the 1D1R cell showed a larger set voltage, which might be attributed to the vertical integration of the 1R and 1D devices; the existence of inherent resistance increases the series resistance of each 1D–1R device. On the other hand, the memory cell in negative-bias regime exhibited limited current flow due to the high Schottky barrier between the P3HT layer (HOMO: 4.9 eV) and Al electrode (work function: 4.3 eV). To certify the vertical integrated diode can well suppress the sneak paths, the read operation tests for 2 × 2 1R and 1D1R arrays were conducted (Figure 4e). In the 1R-only array, the selected three neighboring cells were programmed into LRS (defined as the “1” state), meanwhile the unselected cell (defined as the “0” state) also show LRS, indicating the existence of misreading induced by cross-talk problem between the neighboring cells (Figure 4e,f). In contrast, for the 1D1R array, when the selected three neighboring cells were programmed into LRS, the unselected cell also exhibited HRS (defined as the “0” state) (Figure 4e,f). Therefore, the HRS of unselected 1D1R device was read correctly, confirming the effective suppressing of sneak paths by the integrated 1D1R all-polymer device in cross-bar architecture.

In summary, for the first time, we demonstrated the solution-processed all-polymer 1D1R cross-bar memory array consisting of a vertically stacked P3HT diode and a 2D CMP memory cell. The 2D CMP layers were prepared via surface-confined Schiff-base reaction at room temperature, with the merits of large-area, tunable nanoscale thicknesses, excellent structural stability, and multi-element designs. With the CMP layer obtained as resistive switching medium, the sandwichstructured CMP device exhibits nonvolatile flash performance, with the features of low set voltage (~1.65 V), stable cycling endurance (>25 cycles), and long retention time (10^5 s). More importantly, an all-polymer 1D1R array was fabricated by integrating a P3HT diode on the CMP memory device directly. The direct formation of bilayer structures consisting of 2D memristive polymer and conventional conjugated polymer materials provide intriguing opportunities in memory electronics because of its simpler process compared with the existing all-polymer 1D1R devices. In comparison with the 1R-only memory cell, the 1D1R device is capable of rectifying memory and suppressing the sneak paths effectively caused by the cross-talk issue in cross-bar memory architecture. Such advances in constructing 2D memristive polymers and emergent-stacked all-polymer devices present an effective strategy for the precise and facile preparation of bilayer heterostructure conjugated polymer films for integrated polymer memory electronics.

**Experimental Section**

Reagents and Materials: We purchased Tapy and ThCHO from Aladdin Chemicals. The organic solvents including DMF, toluene, DCM, and chlorobenzene in the experiment were supplied from Aladdin Chemicals. These above organic solvents were used without further purification, unless stated otherwise. SiO_2 (300 nm)/Si substrates were purchased from SZJXTech (SuZhou, China). The rGO/SiO_2 (300 nm)/Si and patterned rGO/SiO_2 (300 nm)/Si substrates were prepared according to our previous studies. Preparations of CMP Films: A typical experimental procedure is described as follows: Tapy and ThCHO were dissolved in DMF to prepare monomer...
solution. A mixed solution of the two monomers with molar ratio of 1:2 was added in a glass bottle. The monomer concentrations of Tapy and ThCHO were 0.75, 1.5, 3.0, or 6.0 mmol L\(^{-1}\), and the corresponding monomer concentrations of ThCHO were 1.5, 3.0, 6.0, or 12.0 mmol L\(^{-1}\), separately. The substrates (SiO\(_2\)/Si, rGO/SiO\(_2\)/Si, quartz) were submerged in the reaction solution. Then, 100 \(\mu\)L acetic acid was added into the solution. The glass bottle was fastened with a lid and kept in room temperature for 3 days. After that, the CMP thin films were formed onto the substrate. Then, the CMP films were washed with DMF and ethanol, and dried at 120 °C under vacuum overnight. Anal. Calcd. for CMP thin films: C, 76.89; H, 3.89; N, 10.89; S, 8.34; found: C, 76.83; H, 3.89; N, 11.87; S, 7.39.

**Fabrication of CMP-Based Memory Devices:** The memory devices were fabricated on SiO\(_2\)/Si substrates with the structure of rGO/CMP films/Al. CMP films were first grown on patterned rGO electrodes according to the aforementioned method. The monomer concentration of Tapy and ThCHO were 3 and 6 mmol L\(^{-1}\), separately. After the CMP films were cleaned and dried, Al electrodes were deposited on CMP films using shadow masks by thermal deposition, with a slow deposition rate, to fabricate cross-point or cross-bar sandwich structure memory devices.

**Fabrication of All-Polymer 1D1R Memory Devices:** The CMP films were prepared on patterned rGO electrodes as the aforementioned method. Square-type (bar-type) patterned Al electrodes were deposited on CMP films by a thermal evaporator in a direction perpendicular to the bottom rGO electrodes. The length of square structure Al electrodes is the same as the width of the rGO electrode. The Al electrodes were used as the top electrodes of the memory devices and the bottom electrodes of diode devices. Then, P3HT solution (50 mg mL\(^{-1}\)) was spin coated on the memory device at 1500 rpm for 60 s. Finally, as the top electrodes for the diode, bar-type (square-type) patterned Au electrodes (100 nm) were slowly deposited on P3HT film by a thermal evaporator.

**Characterization:** Elemental analyses were carried out on an Elemental Analyzer Vario EL III Instrument. Optical microscopy was performed using a Nikon optical microscope. The morphologies of these CMP films were recorded with an AFM instrument (Dimension ICON with NanoScope V controller, Bruker). SEM images were recorded by using a field-emission SEM (JEOL, Model JSM-7600F). FT-IR spectra were taken on a JASCO model FT IR-6100 IR spectrometer using KBr platelets. The absorption spectra of the monomers and CMP films were measured with a Shimadzu UV-3600 spectrophotometer. CMP films were directly prepared on the quartz plates. Thermogravimetric analysis was undertaken with a Shimadzu thermogravimetry at a heating rate of 10 °C min\(^{-1}\) under N\(_2\). The electrical characteristics of memory devices were characterized using a Keithley 4200-SCS semiconductor parameter analyzer. All electrical measurements were carried out under ambient conditions.

**Supporting Information**

Supporting Information is available from the Wiley Online Library or from the author.

**Acknowledgements**

The authors thank primary financial supports by the National Key R&D Program of China (2017YFB1002900), the National Natural Science Foundation of China (61622402, 51703093), and the Natural Science Foundation of Jiangsu (BK20171000).

**Conflict of Interest**

The authors declare no conflict of interest.

**Keywords**

cross-talk issues, multilayer polymer electronics, rectifying memory, rewritable nonvolatile memory, two-dimensional conjugated microporous polymer

Received: June 18, 2019  
Revised: July 3, 2019  
Published online: July 23, 2019

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