

Development and application of graphene-based materials in memristors

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Abstract: Graphene-based materials are regarded as the key to building carbon-based platforms because of their unique two-dimensional material properties. In addition to graphene field effect tubes, graphene-based non-volatile memories are also an important part of building carbon-based platforms. Graphene-based materials have rich resistive switching mechanisms, excellent electronic properties, optical properties, and excellent impermeability, so that graphene-based materials can be used in various functional layers of memristors to prepare devices with low power consumption, long life, and Ultra-thin, ultra-transparent, flexible, high-stability large-scale integrated devices. This article reviews the different classifications of graphene-based memristors and the resistive switching principle of graphene-based materials. Taking the sandwich structure as an example, it introduces in detail the development status, working principle, and future development direction of graphene-based materials as the resistive layer, electrode layer, and modification layer of memristors. It provides researchers with ways to prepare graphene-based memristors.

Keywords: Graphene-based materials; Graphene; Memristor

1. Introduction

In order to break through the physical limits of traditional Si semiconductor devices, researchers have focused on carbon-based materials [1-3]. However, most current research on carbon-based electronic devices only focuses on field-effect transistors. If non-volatile memory can be implemented in carbon materials, logic circuits and memory devices can be integrated on the same carbon-based platform, achieving real industrial change. However, materials with resistance switching properties currently found cannot fully meet the requirements for memristor applications. Therefore, exploring new storage media is still a key project for the development of carbon-based memristors.

Since its discovery by Geim and Novoselov in 2004 [4], graphene (abbreviated as Gr) has garnered significant attention in micro-nano electronics research. Boasting ultra-high carrier mobility, exceptional mechanical strength and flexibility, remarkable thermal conductivity [5], a high specific surface area [6], and favorable optical properties [7], graphene stands out as a material with immense potential for technological innovations. Graphene oxide, comprising a graphene lattice interspersed with oxygen-rich functional groups including epoxides, hydroxyls (-OH), and carboxyls (-COOH), exhibits a substantially high resistivity due to the presence of these groups. The versatility of graphene oxide (GO) lies in its tunable energy band structure and electronic properties, which can be modulated by varying the concentration of these surface-attached chemical functional groups. Through standard photolithography techniques, deposited graphene oxide (GO) films can be further processed into functional devices with varying properties without compromising their film properties [8,9]. Consequently, graphene and its derivative materials are very suitable for use in making memristors.

From the perspective of graphene derivative materials, optimizing the memristive mechanism is crucial to promote the establishment of stable and highly integrated neural networks. This will not only build a more efficient and energy-saving computing architecture, but also promote the ability of memristors to adapt to diverse functional requirements in different environments.

2. Graphene-based memristor

2.1. Structure of graphene-based memristor

The architectures of conventional memristors can be broadly categorized into three configurations: two-terminal, three-terminal, and four-terminal structures^[10].

The two-terminal structure features a classic 'sandwich' architecture, comprising a top electrode, a functional layer, and a bottom electrode. This simplistic design ensures ease of manufacture and exhibits excellent scalability for production.

The three-terminal structure is similar to the structure of a commonly used triode, consisting of three terminals: gate, source, drain and an insulating layer. Upon the application of a certain electric field to the source and drain electrodes of the device, a low-resistance state is established between the two electrodes, resulting in a conductive pathway. When a negative bias voltage is applied to the gate, the atoms that form the conductive channel between the source and drain will move away from their original positions, causing the conductive channel to break, thereby disconnecting the path between the source and drain. When an adequate forward voltage is applied to the gate terminal, it facilitates the reformation of the conductive channel between the source and drain electrodes, thus reinstating electrical conductivity between the two terminals. The advantage of this structure lies in its facile manageability, whereby the device conduction can be modulated simply by varying the voltage at a single terminal.

The four-terminal structure solves the challenge of mitigating excessive power consumption that occurs during the transition between high and low resistance states. Contrasted with traditional two-terminal devices, this structure incorporates an additional pair of control electrodes alongside the working electrodes; the control electrodes serve to modulate the latter, thereby minimizing leakage current produced by the conductive channel and, consequently, improving the problem of excessive power consumption in the device.

Graphene-based memristors currently mainly use a two-terminal structure as it affords simplified fabrication, an uncomplicated architecture, and facilitates the observation and analysis of the memristive principle. This renders it an optimal choice for the foundational exploration of the memristive properties of two-dimensional materials, as shown in Figure 1.

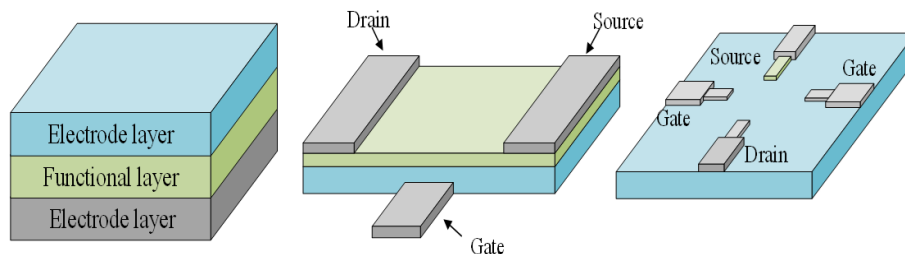


Figure 1: Schematic diagram of the basic structure of a memristor.

2.2. Response patterns of graphene-based memristors

Memristors can be categorized into two distinct types, digital and analog, contingent upon their mode of current response when subjected to voltage bias conditions. In a digital memristor, the current switches abruptly between a high resistance state (HRS) and a low resistance state (LRS) as the voltage increases. This binary mutation characteristic can simulate the fully open "1" or fully closed "0" behavior of neurons in a spiking neural network. The structure of the analog memristor is similar to that of a synapse, in which the two electrodes resemble the presynaptic and postsynaptic membranes, and the active layer corresponds to the synaptic cleft. The resistance of an analog memristor can change under various voltage simulations, similar to changes in synaptic weights. Therefore, analog memristors are suitable devices for simulating artificial synapses and neural networks.

Digital memristors can be classified into three operational modes: unipolar, bipolar, and write-once-read-many (WORM)^[11-12], as shown in Figure 2, unipolar devices can be switched off by either a forward or reverse voltage following a change in resistance state. The working principle of bipolar memristor is to change the resistance state by applying a forward or reverse voltage, and only when a voltage of opposite polarity is applied to the previous one, the device will change the resistance state again, thereby completing the storage of data. WORM-type devices, which stand for write-once read-many-times

memory, possess unique characteristics in the data storage domain with the capability of being written once and read repeatedly.

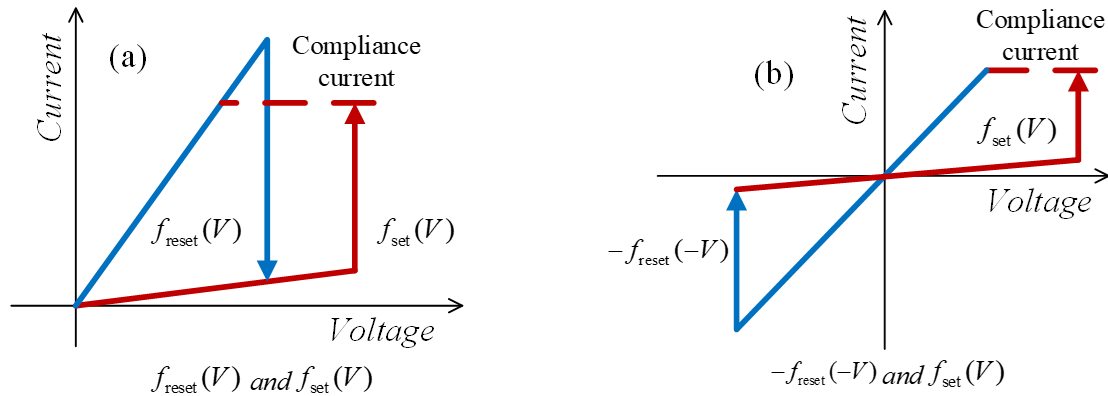


Figure 2: Unipolar/bipolar resistive switching behavior corresponding to I-V image.

2.3. Resistive switching mechanism of graphene-based memristor

Memristor is a storage element with rich connotations and diverse mechanisms. The most common resistance-changing phenomenon in graphene-based memristors is mainly the change in resistance caused by oxidation-reduction reactions. This change is reflected in the dynamic migration of oxygen-containing groups in the resistive layer, the conductive filaments formed by oxygen vacancies, and the metal conductive filaments formed by metal electrodes. These characteristics jointly determine the resistance control mechanism of memristor, which is an important basis for its wide application in the field of storage technology.

In the resistive switching process of graphene memristor, graphene oxide (GO) plays a central role as the resistive switching layer, especially the dynamic migration of oxygen-containing functional groups plays a key role in the resistive switching. The GO layer contains a variety of oxygen-containing functional groups such as epoxy groups, hydroxyl groups, and carboxyl groups, and most of the functional groups are aggregated on the GO surface through sp^3 hybrid orbitals. Under the action of an external electric field or ultraviolet light, these functional groups will be removed, resulting in the formation of sp^2 hybrid orbitals, thus significantly improving the conductivity of the device. When a device transitions from a high-resistance state (HRS) to a low-resistance state (LRS), it signifies that the device has switched to an operational mode. During operation, oxygen ions migrate from the graphene oxide to the reduced graphene oxide (rGO), triggering a reduction reaction within the rGO material. Subsequently, when these oxygen ions reverse their migration, the reduced graphene oxide is re-oxidized to graphene oxide, thus returning the device to its initial high-resistance state (HRS), effectively turning the device off.

Secondly, the employment of active metals as electrode materials can produce memristors. Upon application of a forward bias to the active metal electrode, a redox reaction is initiated, resulting in the generation of metal cations. Driven by the electric field, these cations move toward the cathode and combine with electrons to return to their atomic form. With continued voltage application, these metal atoms progressively conglomerate, establishing metallic conductive filaments; as a consequence, the device undergoes a transition from a high-resistance state (HRS) to a low-resistance state (LRS). Conversely, when a reverse bias is applied to the active electrode, these metal filaments begin to disintegrate, spurred by both Joule heating and redox reactions. This dissolution culminates in the disconnection of the conductive paths, thereby restoring the device to its original high-resistance state.

Moreover, the operational principle of graphene-based memristors often hinges upon the electric field-induced migration of oxygen ions. This migration process plays a decisive role in the resistive switching characteristics of the device. Under the influence of an applied electric field, oxygen ions are prompted to move towards the anode, leaving behind positively charged oxygen vacancies where they were once situated. These vacancies then experience a redistribution under the continued influence of the electric field, resulting in the formation of conductive pathways. The conductive filaments formed by this oxygen vacancy are repeatedly disconnected and connected under the action of electrochemical redox reactions, causing the device to cycle between high and low resistance states.

A resistive switching mechanism dominated by electronic effects also appears in some hybrid

composite graphene-based memristors. This is because the charge traps inside the material or at the interface will reduce the mobility of electrons. When a voltage is applied to the electrode, electrons will continue to fill the traps. When the traps are filled, the electrons will not be affected by the traps, so the current suddenly increases. It becomes a low-resistance state. Then, driven by a reverse applied voltage, the electrons will be separated from the trap and return to a high-resistance state. In this way, the cycle is repeated to realize the resistive switching function of the memristor.

The resistive switching mechanisms of graphene-based memristors are manifold and often do not operate in isolation. Influences such as the fabrication process, the thickness of the resistive switching layer, electrode material choice, and extrinsic environmental conditions collectively impact the resistive switching behaviors exhibited by these devices. Consequently, ongoing research into the fundamental principles underpinning the resistive switching phenomena in graphene-based memristors remains paramount.

3. Graphene-based resistive layer

The resistive switching layer of memristor is the key to realizing resistive switching. A variety of graphene-like materials can be used as the resistive switching layer of memristor. Such as graphene oxide, reduced graphene oxide, graphene quantum dots, etc. Since graphene-like materials have special energy band structures and electronic properties, the materials can be modified by changing the chemical functional groups or resistive layers attached to the surface and doping, so that the graphene-like materials can meet different memristive requirements. Therefore, oxide graphene-like materials are a promising material for microelectronic devices.

Realizing the resistive switching function by utilizing the redox properties of oxygen-containing functional groups in graphene oxide is currently the most common and widely used graphene-based memristor design method. Sung Kyu Kim et al. [13] prepared Au/GO/Al devices with a bottom interface layer and showed stable resistive switching behavior. And each resistive state is maintained without degradation after 10^4 s. The author used low-pressure spherical aberration-corrected transmission electron microscopy to prove that the conduction of this device is due to the nanoscale conductive graphite channels in the graphene oxide film. Figure 3 shows the cross-sectional BFTEM image of the ON-state device obtained after applying a negative bias to the upper electrode. The middle region of GO shows a dark contrast compared to the left and right regions. This contrast is closely related to the large increase in crystallinity. Therefore, it is explained that the conductive graphite channel is formed by the migration of oxygen ions under the action of a strong electric field. However, it is not easy to generate a local electric field in a specific area because the graphene oxide film has uniform thickness and roughness, Sung Kyu Kim et al. used Al conductive filaments as local current paths for concentrated electric fields, and directly formed stable conductive channels on the graphene oxide film through oxygen diffusion outwards.

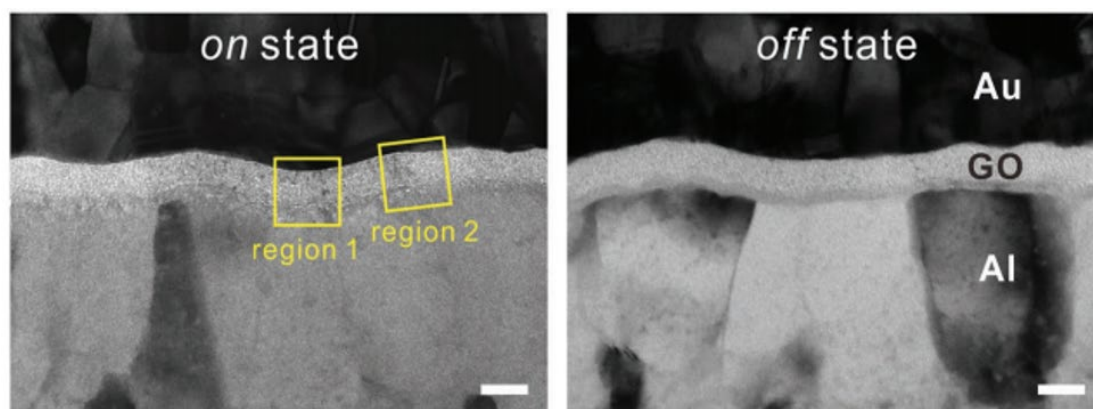


Figure 3: Cross-sectional BFTEM images of the device in the ON and OFF states of the device.

In order to modulate the effects of oxidation-reduction, researchers have made numerous attempts, addressing the issue through material selection as well as through optimization of the fabrication processes, controlling annealing temperatures to enhance a set of properties. Mingdong Yi et al. [14] fabricated an indium tin oxide (ITO)/graphene oxide (GO)/Ag structured memristor, and by controlling the annealing temperature of the graphene oxide film at 20°C , the device exhibited a typical write-once, read-many-times (WORM) memory behavior. It demonstrated a relatively high ON/OFF current ratio

($\sim 10^4$), a high high-resistance state (HRS) to low-resistance state (LRS) ratio ($\sim 10^5$), and stable retention characteristics ($>10^3$ s) at low programming voltages of -1 V and -0.5 V.

The conduction mechanism of the device is analyzed through the I-V curves of the on and OFF states. The charge transport in the OFF stage is controlled by the Ohmic model and the SCLC model, and the charge transport in the ON stage is controlled by the Ohmic model, as shown in Figure 4.

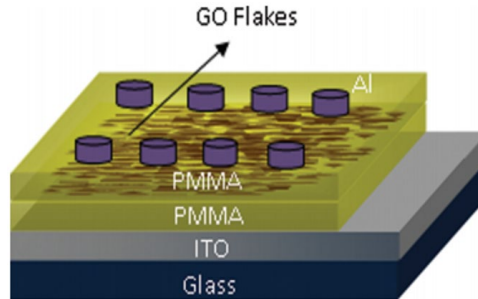


Figure 4: ITO/PMMA/GO/PMMA/Al device structure diagram.

On the basis of redox, researchers have also tried to introduce light signals to regulate the resistive switching process. Xiaoning Zhao et al. [15-16] proposed a photocatalytic reduction method for local control of graphene oxide domains. The redox process of graphene oxide can be understood as the electric field-induced connection/disconnection of nanoscale reduced graphene oxide conductive filaments. The electroformation process usually results in current overshoot resulting in high randomness of rGO CFs, so it uses a TiO_2 -assisted photocatalytic reduction method to locally generate rGO domains by controlling the UV irradiation time and TiO_2 concentration. The formation process was eliminated and the overgrowth of rGO was successfully suppressed and improved RS memory properties were achieved in graphene oxide- TiO_2 (GO- TiO_2) nanocomposites, including reduced SET voltage, improved switching variability, and improved the switching speed as shown in Figure 5. This is because TiO_2 nanoparticles catalyze the reduction of GO under the action of ultraviolet illumination, which will form local clusters of sp^2 in the film, causing the device to increase the surrounding local electric field during the conversion process of sp^3 and sp^2 .

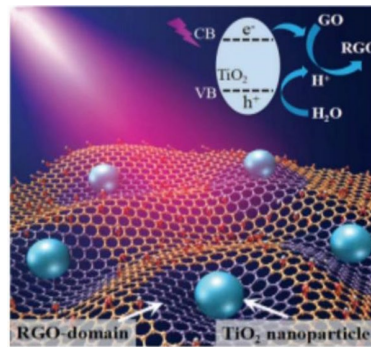


Figure 5: Schematic diagram of TiO_2 -assisted photocatalytic reduction of GO.

Doping usually refers to the incorporation of small amounts of other elements or compounds into a material or matrix in order to improve performance. Doping can produce specific electrical, magnetic and optical properties in materials and matrices, thereby making them have specific value or use. In the preparation of graphene-based memristors, our common doping is doping metal particles in the resistive layer. Doped metal particles can eliminate part of the energy of the initial oxide during the electrochemical process, improve the uniformity of the conductive filaments, and reduce energy consumption. Marina Sparvoli et al. [17] produced a silver-doped graphene oxide (GO) memristor to simulate neuron membrane discharge. This structure is RRAM rGO + 1% Ag/GO + 1% Ag/Al. According to the doping with different concentrations of Ag, memristor devices have different threshold switching characteristics. Graphene quantum dots can also be introduced into the resistive switching layer. Graphene oxide quantum dots (GOQDs) are obtained by oxidation of graphene quantum dots with a size less than 100 nm. They can be embedded in the resistive switching functional layer to guide the nucleation of carbon filaments. And growth direction, improving the uniformity of conductive filaments and improving the dispersion of switching voltage.

Xiaobing Yan et al. [18] introduced goqd into the ZHO layer to prepare memristor, which has lower

VS and VR threshold voltages than memristor without GOQD. In addition, switching speed and hold-up performance are significantly improved, and low power consumption is achieved. The reason for achieving good RS is that GOQDs enhance the local electric field and guide the formation direction of cf. This work provides a new way to reduce device power consumption and improve the stability and uniformity of oxide-based memristors. On this basis, Xiaobing Yan et al. [19] produced a memristor device based on goqds with bidirectional fine-grained conduction tuning characteristics. The device resembles many functions of biological systems, including nonlinear transmission, STDP, PPF, and "learning experience" behavior. This device provides a promising option for future applications in neural computing systems with low power consumption and ultra-fast switching speeds.

Inserting homogeneous carbon-based materials is also an effective method to improve the RS reliability of graphene oxide-based memristor devices. Ye Tao et al. [20] prepared an Al/GO/LGAC/Ni nanocrystal/p-type si device, in which the LGAC (locally graphitized amorphous carbon) layer was used to improve RS reliability. The optimized memory device also has good cycle durability and high temperature retention characteristics. This is due to the LGAC insertion layer, so that the migration of oxygen ions during the SET and RESET processes is well restricted within a narrow conductive region.

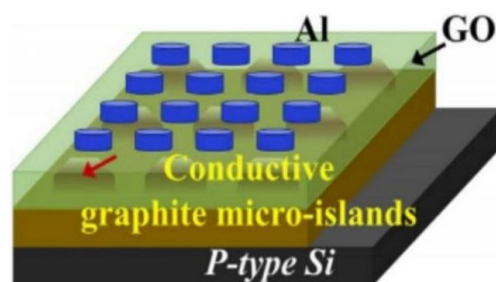


Figure 6: Schematic diagram of Al/GO/LGAC/Ni nanocrystal/p-type si structure.

Graphene-based composite materials are also the main materials for the resistive layer of graphene-based memristors. Huu Thoai Ngo et al. [21] produced and studied Ag/PVA-GO/FTO hybrid composite resistive layer memristive devices as shown in Figure 6. The unoccupied energy levels of graphene oxide can serve as electron traps, and the switching mechanism of the device is the trapping and detrapping process of injected electrons under the action of an external electric field. By comparing with Ag/PVA/FTO, Ag/GO/FTO and Ag/PVA-1.0 wt% GO/FTO devices, the authors found that, the power consumption of the Ag/PVA-0.5 wt% GO/FTO device (scan voltage -0.5 V -0.5 V, $V_{SET}=0.28$ V, $V_{RESET}=0.34$ V, switching ratio $I_{ON}/I_{OFF}=104$) is lower, which the author believes is due to There is a good interaction between the hydroxyl group (-OH) of PVA and the C=O of GO at this mixing concentration.

Enming Zhao et al [22] prepared cross arrays of flexible memory devices based on graphene oxide polymer nanocomposites. The resistive switching memory characteristics of the devices were found to be significantly responsive to bending. 2000 cycles of flexible bending could convert the resistive switching memory characteristics from WORM to SRAM. The authors suggested that the resistive switching mechanism is attributed to the formation of a carbon-rich conducting layer with non-volatile WORM characteristics, and that bending-induced micro-cracks lead to incomplete breakage of the conducting channels, resulting in volatile characteristics of the devices.

4. Graphene-based electrode layer

Two-dimensional (2D) graphene has excellent electronic properties, optical properties and chemical stability. Compared with metal electrodes, graphene can be used as an electrode not only to reduce the power consumption of the memristor, but also to improve the bending resistance of the flexible memristor because of its high electrical conductivity, special tunneling barrier effect and good flexibility.

Bin Han et al [23] successfully constructed chalcogenide PDs by plating graphene films on quartz substrates as electrodes. gftz -chalcogenide PDs have an ultra-low detection optical density ($0.5 \mu\text{W}/\text{cm}^2$) at a bias voltage of 1 V compared to PDs prepared with Au electrodes. gftz -chalcogenide PDs have higher maximum responsivity and defect rate (0.1 A/W) and D^* ($2.6 \mu\text{g} \times 10^{11}$ Jones) were higher than the values of Au - chalcogenide PDs. gftz - chalcogenide PDs presented higher switching ratios than PDs fabricated with Au electrodes, and the authors concluded that graphene electrodes could collect carriers more efficiently, and that the well-defined Schottky contacts contributed to the reduction of the

operating voltage.

Hyun Wook Shin et al ^[24] prepared graphene/NiO/HOPG by depositing a polycrystalline NiO film on a single-crystal HOPG substrate using the RF sputtering technique as shown in Figure 7. The graphene/NiO/HOPG RRAM memristors exhibited lower SET and RESET voltages than the Pt/NiO/Pt RRAM memristors, and the authors suggested that this was due to the graphene and the HOPG electrodes' low power function. In addition, it reduces the dispersion of the LRS and HRS states as well as the SET and RESET voltages for repeated switching cycles.

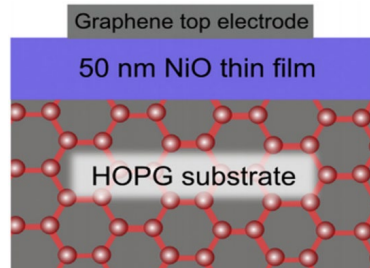


Figure 7: Schematic diagram of graphene/NiO/HOPG structure.

Hongbin Zhao et al ^[25] demonstrated a highly transparent resistive random access memristor with a multilayer graphene (MLG)/Dy₂O₃/indium tin oxide (ITO) structure, which has faster switching speed, larger resistance ratio, lower threshold voltage, and lower power consumption relative to a conventional transparent memristor. The authors believe that these properties are derived from a combined model of graphene oxide and Dy₂O₃ fiber conductivity. Oxygen vacancies are created when oxygen ions (O²⁻) are retracted from the substrate by external fields and thermal effects. When a positive bias voltage is applied to the MLG electrode, a graphene oxide layer is formed at the MLG/Dy₂O₃ interface, and once a sufficient concentration of oxygen vacancies is created in the Dy₂O₃ film, a conductive filament is formed, which changes the device to a low-resistance state. Due to the formation of the graphene oxide layer, the total resistance of the memristor device in the LRS increases compared to the metal electrode device. Subsequently, the formation of a high resistance state when a positive bias voltage is again applied to the MLG is a result of the breakup of the conductive filaments by Joule heating.

In the above discussion we found that graphene-based materials can act as different functional layers of sandwich-structured memristors under appropriate conditions. For example, Ya lin et al. ^[26] developed an all-carbon memristor based on GO. As shown in the figure 8, graphene oxide (rGO) and graphene (graphene) layers are used as the top layer and BEs, graphene oxide and NCQDs nanocomposite film (GO-NCQDs) is used as a resistive switching layer to form a memristor with an rGO/GO-NCQDs/graphene sandwich structure. UV light irradiation induces the local reduction of graphene oxide near NCQDs, forming multiple weakly conductive filaments and exhibiting a simulated RS with continuous changes in conductance. The image recognition accuracy of the memory neural network prepared by this device is as high as 96.7%. And it can also be applied to different substrates, showing good flexibility and consistency as well as ultra-high thermal stability. This provides the possibility for us to realize all-carbon wearable electronic devices.

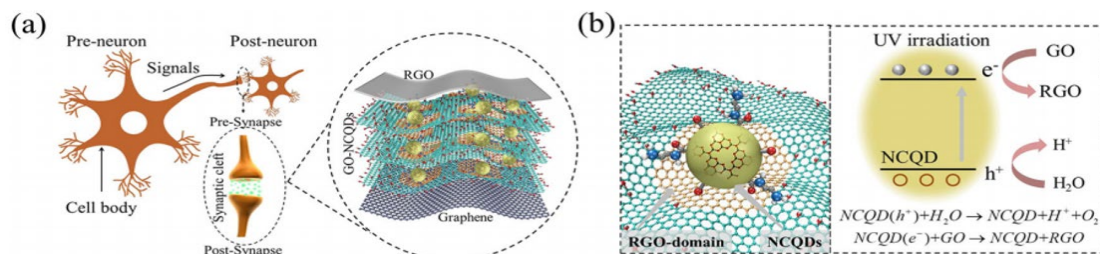


Figure 8: Schematic diagram of device switching mechanism of rGO/GO-NCQDs/G structure.

5. Graphene-based modified layers

Sen Liu et al ^[27] prepared Ag/ZrO₂/G/Pt, Cu/HfO₂/G/Pt and Ag/SiO₂/G/Pt devices by transferring a monolayer graphene barrier layer onto the surface of Pt electrode by wet chemical transfer process. It was found that the insertion of a single layer of graphene could suppress the unexpected negative set. Inserting a barrier layer at the interface between the RS layer and the Pt electrode is an effective method

to stop the diffusion of active metal ions or atoms. This is due to the fact that 2D graphene exhibits excellent impermeability in stopping molecular migration.^[28] In addition to the impermeability, graphene has high electronic and thermal conductivity as shown in Figure 9. Therefore, graphene would be an excellent ionic/atomic barrier layer embedded in the interface between the RS layer and the Pt layer. Based on this we conclude that there is competition for CF formation and dissolution during negative reset if an active metal source is embedded in the counter electrode. The competition will lead to the failure of RESET operation. When the graphene barrier layer prevents the CF from penetrating the counter electrode, it suppresses the competition, eliminates the RESET failure phenomenon, and enhances the stability of the device.

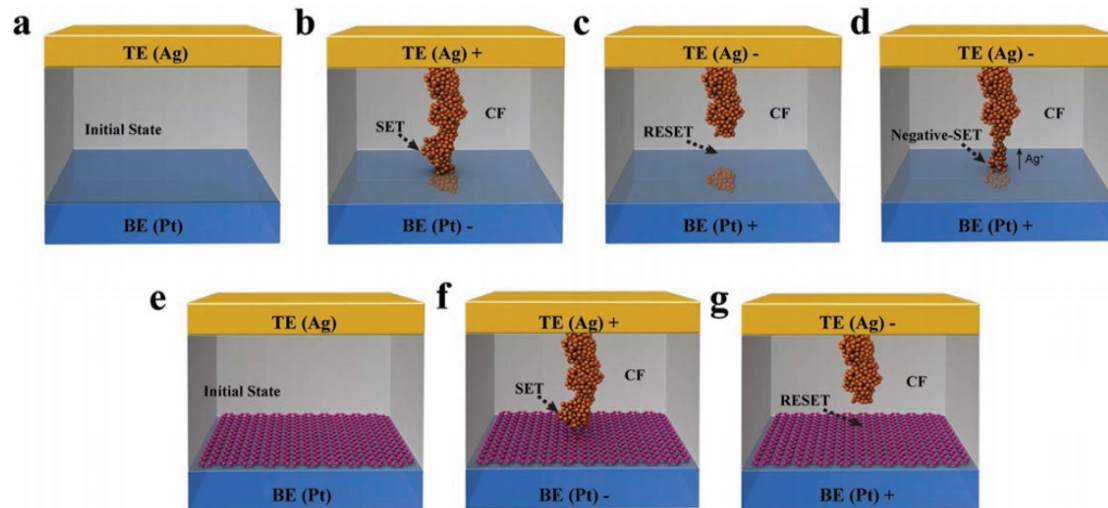


Figure 9: Schematic diagram comparing the switching mechanisms of Ag/ZrO₂/Pt and Ag/ZrO₂/G/Pt devices.

6. Conclusion

This paper reviews the application development of graphene and its derivatives in the field of memristors. When graphene is used as an electrode layer, it can effectively reduce operating current and energy consumption due to its high out-of-plane contact resistance and weak van der Waals forces on the surface. In some cases, graphene is not only an electrode material, but also participates in the redox reaction of the resistive layer, which not only improves device performance but also simplifies the preparation process. As a resistive switching layer, graphene and its derivatives have rich resistive switching principles, such as redox effect, metal thermal effect and other mixed memristive mechanisms, which can achieve a stable resistive switching effect. By changing the thickness, preparation process, contact electrode material, additive voltage and external environment control of the resistive switching layer, or doping the resistive switching layer with specific impurities, it can be adapted to different application scenarios. In addition, graphene-based materials can also be used as a modification layer in memristors to enhance their resistance to permeability, thus improving the stability and reliability of the entire device.

Graphene-based materials can be applied to various functional layers of memristors, and graphene-based memristors generally have good bending resistance, flexibility, non-volatility and transparency, and have unique light response characteristics. In the future, they can be produced with low power consumption, long life, ultra-thin and high Transparency, flexible, and highly stable large-scale integrated devices can be integrated on various curved surfaces, including human skin. Therefore, memristors made of graphene and its derivative materials have very broad development prospects. With the development and application of graphene-based memristors, graphene-based materials are expected to make the integration of logic circuits and memory devices on the same carbon-based platform a reality, achieving real industrial change.

The first is to continue to conduct in-depth research on the switching mechanism of graphene-based memristors. Due to the diversity of device structures, materials and preparation methods, as well as the limitations of research methods, the rules of device switching mechanisms are still unclear. It is necessary to use advanced research methods such as in-situ observation for different material systems and device structures to explore the rules followed by the device switching mechanism.

The second is material modification, optimizing the selection and ratio of doping materials to further improve switching performance and stability.

The third is to optimize the device preparation process. From raw material selection to preparation process, everything tends to be simple, environmentally friendly, and can be manufactured in batches.

Fourth, attention can be paid to the control of memristors by optical signals, acoustic signals, pressure signals, etc., and the development of coupled memristor functions will make memristors more practical.

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