ISSN 2616-5880 Vol. 5, Issue 2: 64-69, DOI: 10.25236/AJMC.2024.050210

Research progress on the resistive switching materials in optoelectronic memristors

Kangfei Zhao[1], Qiang Wang[1,*], Di Chen[1]

[1]*School of Electronic Information and Artificial Intelligence, Shaanxi University of Science and Technology, Xi'an, Shaanxi Province, China*
[*]*Corresponding author: qiang.wang@sust.edu.cn*

Abstract: *Optoelectronic memristors, combining photoelectronic and resistive switching materials, have garnered considerable attention in recent years due to their unique characteristics. This article provides an overview of the latest research progress in optoelectronic memristors and the associated resistive switching materials. Emphasis is placed on discussing the performance and potential applications of various materials, including oxide materials, ferroelectric materials, perovskite materials, two-dimensional materials, and organic materials. The review also highlights the advantages and challenges encountered by optoelectronic memristors and provides insights into future development trends.*

Keywords: *Optoelectronic memristor, Resistive switching materials, Device performance*

1. Introduction

In the past few decades, the development of Moore's Law has significantly improved chip composition and storage density. However, as transistor sizes decrease and the number of transistors increases, the semiconductor process's complexity has grown, presenting challenges. Today, the demand for data processing capabilities is increasing, and computers based on von Neumann architecture face significant challenges. Therefore, researchers are exploring new materials, devices, and architectures to replace or improve device based on complementary metal oxide semiconductors (CMOS).

The memristor is considered a promising new type of storage component due to its ultra-high density storage, high switching speed, and reliability. It functions as a resistor with memory, exhibiting a non-linear relationship between charge and magnetic flux. Proposed by Chua [1] in 1971, the memristor is regarded as the fourth basic circuit component alongside resistors, capacitors, and inductors. The relationships among these components can be derived from the fundamental parameters of charge, current, voltage, and magnetic flux.

Optoelectronic memristor represents a novel category of optoelectronic devices that integrates light signals, electrical signals, and memory functions, holding significant research value and promising application prospects. Building upon traditional telecommunications signal memristors, the addition of synergistic optical signal modulation enhances its capabilities. This augmentation harnesses the advantages of light signals, including low power consumption, minimal crosstalk, high speed, and broad transmission bandwidth, allowing for flexible simulation of functions such as paired-pulse facilitation (PPF)/depression (PPD), spike timing-dependent plasticity (STDP), and the conversion from short-term memory (STM) to long-term memory (LTM). In recent years, propelled by advancements in optoelectronic technology, nanotechnology, and materials science, research on optoelectronic memristors has achieved significant strides.

Central to the functionality of optoelectronic memristors are resistive switching materials, whose performance directly impacts device operation. However, the stability and reliability of these materials require further enhancement to align with practical application demands. Recent research efforts have been dedicated to the exploration of high-performance resistive switching materials, encompassing oxide materials, ferroelectric materials, perovskite materials, two-dimensional materials, organic materials, among others. These materials exhibit diverse characteristics, such as high switching rates, excellent stability, and tunable band structures, thereby offering a rich array of options for advancing the study of optoelectronic memristors.

With the continuous development of science and technology, the research and application of optoelectronic memristor will make greater breakthroughs. The discovery and use of the new type of

ISSN 2616-5880 Vol. 5, Issue 2: 64-69, DOI: 10.25236/AJMC.2024.050210

resistive switching materials will provide new possibilities for the performance improvement of optoelectronic memristor. At the same time, the application of optoelectronic memristor in the field of optoelectronics will continue to expand, bringing new opportunities for the development of optical communication, optical computing and optoelectronics integrated circuits. This article describes the latest research progress of the preparation materials of optoelectronic memristor, summarizes the advantages of optical recollers and the main challenges facing development, and looks forward to its future development prospects.

2. Materials of memristors

Materials serve as the foundation of the device, with resistive switching materials playing a pivotal role in memristors. Different resistive switching materials have different resistance characteristics. The selection of an optimal optoelectronic material is crucial in the construction of high-performance optoelectronic memristors. Optoelectronic memristor not only requires electrical signals to modify the device block, but also the optical signal also has adjustment effect[2]. On the one hand, most optoelectronic resistive switching material itself has good photoelectronic characteristics, and can effectively use optical signals to modulate memory resistance, such as inorganic materials (a variety of semiconductor materials, perovskite materials, new two-dimensional materials, etc.) exhibit more stable, faster, and robust memristor behavior. On the other hand, some optoelectronic resistive switching material need to achieve its optoelectronic ductability through solutions such as organic materials, composite material systems, and device structure design with the advantages of high flexibility, simple preparation methods, and low cost.

Optical resistive switching materials exhibit changes in resistance under light exposure. Its working mechanism mainly involves the effects of photogenerated charge carriers on the lattice structure. Specifically, the photoelectronic resistive switching material will generate photogenerated charge carriers under the condition of light. These carrier will move in the material and interact with the lattice structure. This interaction can cause changes in the lattice structure, which changes the resistance of the material. Upon cessation of light exposure, the photoexcited carriers recombine and dissipate, but the lattice alterations persist, leading to a sustained change in resistance. This altered state can endure for an extended period, facilitating the development of devices capable of repeated resistance modulation.

2.1. Oxide materials

The common working mechanism of oxide optoelectronic memristors is to use optical signals to change the number of electrons captured by interface defect states, thereby modulating the Fermi level of the material or the potential barrier at the interface between the electrode and the dielectric layer, and thereby changing the resistance value of the optoelectronic memristor.

At present, binary oxides commonly used as resistive layer materials for memristors have the characteristics of simple composition, high stability, low cost, simple preparation process, and compatibility with traditional CMOS processes. Common binary oxides include TiOx, SiOx, AlOx, NiOx, CuOx, ZnOx, HfOx, TaOx, WOx, ZrOx, SnOx, etc. Among numerous resistive materials, HfOx and TaOx have greater potential compared to other oxide memory devices with high power consumption or low uniformity, as they have operating speeds below ns and ultimate durability exceeding 10^{10} cycles. HfOx has an ultra-high interruption ratio greater than 10^{10}, and TaOx has an ultimate endurance greater than 10^{12} cycles [3]. Although there are many potential candidate materials to prepare memristors, due to the compatibility issue of COMS process, many materials cannot achieve superior device performance.

During the Set process, oxide monolayer memristors typically require current limiting to protect the oxide layer in the memristor from permanent breakdown. The dual dielectric layer structure is a memristor that can achieve self limiting and self rectifying functions by adjusting the properties of the functional layer material and properly matching the energy band structure between the electrode and the functional layer. Zhang[4] et al. prepared a resistance random access memory of ITO/TiO_2/HfO_2/Pt structure, and after 500 mechanical bending cycles, the coefficient of variation of the high and low resistance states were 3.2% and 3%, respectively. The double-layer oxide HfO_2/TiO_2 interface plays a crucial role in the performance of this flexible device.

Polymetallic oxides refer to perovskite structured metal oxides and their dopants, which generally have high dielectric constants, ferromagnetism, and other characteristics. This type of oxide has rich chemical composition and crystal structure, diverse electronic structures and optoelectronic properties,

ISSN 2616-5880 Vol. 5, Issue 2: 64-69, DOI: 10.25236/AJMC.2024.050210

making it an ideal material for preparing optoelectronic memristors. Memristors based on multiple oxides have advantages such as high speed and low energy consumption.

2.2. Perovskite Materials

Perovskite materials are a class of crystalline materials with unique optoelectronic properties. They exhibit changes in electrical conductivity upon absorbing light, enabling information storage and retrieval. Perovskite materials, due to their suitable tunable band gap, high optical absorption coefficient, high photoluminescence quantum yield, narrow-band emission, relatively short absorption wavelength, long diffusion length, and low cost, offer high photoconversion efficiency, wide spectral absorption, and good stability, making them ideal materials for optoelectronic memristors.

Common perovskites include $CH_3NH_3PbI_3$ ($MAPbI_3$), $MAPbBr_3$, $(C_4H_9NH_3)_2PbBr_4$, $LaFeO_3$, etc. Due to their novel optoelectronic properties such as mixed ion and electron conductivity, switchable majority carrier concentration, and slow photocurrent decay, perovskites hold promise for designing next-generation neuromorphic memory resistors. Poddar [8] et al. fabricated RRAMs using perovskite quantum nanowire / well (NW) arrays as switch matrices, achieving a switch ratio of 10^7 with strong storage capability and ultra-fast programming speed.

A significant drawback of perovskite materials is their lack of environmental stability. To address this, Cheng [9]et al. first utilized lead-free double perovskite $Cs_2AgBiBr_6$ for environmentally robust memristors. The ITO/Cs_2AgBiBr/Au structured optoelectronic memristors exhibited retention of 10^5 seconds and mechanical bendability of 10^4 times. Importantly, even after alcohol combustion for 10s at 180°C, the memristor device's performance remained stable, indicating resilience in harsh environments.

Despite extensive research on using perovskite materials in optoelectronic memristors, perovskite optoelectronic memristors still face drawbacks such as incompatibility with CMOS processes, limiting their practical applications.

2.3. 2D materials

In optoelectronic memristors, two-dimensional materials play a crucial role. Firstly, they serve as the photosensitive layer, enabling the detection of light signals. Two-dimensional materials like graphene exhibit excellent photoconversion efficiency, facilitating the transfer of photoexcited electrons to the functional layer of the optoelectronic memristor, thereby triggering the memristive effect. Wang [5] et al. fabricated graphene/molybdenum disulfide/graphene (GMG) vertical structure memristors. The GMG device exhibited typical bipolar resistance switching behavior. Importantly, the device demonstrated over 2×10^7 cycles of endurance and a 100 ns ultrafast switching time, displaying reliable resistive switching characteristics. Additionally, the prepared memristors exhibited good thermal stability, operating normally in the temperature range of 20°C to 340°C. This capability positions optoelectronic memristors with broad application prospects in optoelectronics, such as optical storage, image recognition, and sensor technologies.

Secondly, two-dimensional materials can also serve as the active layer in optoelectronic memristors, where the behavior of electrons can be modulated through light excitation. Wang's research group [6] constructed multi-color light-modulated transparent memristors based on polystyrene and black phosphorus (PB) materials. When applying near-infrared light (785nm), green light (500nm), and ultraviolet light (380nm) to the device, the device's turn-off voltage gradually decreased, and the switch ratio increased. This enhancement in resistive switching performance can be attributed to the elevation of the Schottky barrier caused by the capture of trapping sites capturing photoinduced electrons. These characteristics of two-dimensional materials enable optoelectronic memristors to exhibit more autonomous learning and plasticity behaviors akin to biological synapses. By adjusting the intensity and duration of light, controlled variations in the resistance values of optoelectronic memristors can be achieved, enabling the storage and processing of information to accomplish complex computational tasks.

The application of two-dimensional materials in optoelectronic memristors is made possible by their array of key characteristics. Firstly, they possess excellent conductivity, allowing electrons to freely move on their surface, thereby facilitating efficient electron transport. Secondly, two-dimensional materials exhibit extremely high light absorption rates, effectively converting light energy into electron energy, providing robust support for optoelectronic conversion. Additionally, their mechanical flexibility and chemical stability enable them to operate stably under various environmental conditions, enhancing the stability and reliability of optoelectronic memristors. Xiao [7] et al. proposed flexible memristors with vertical structures based on polyvinyl alcohol-graphene oxide hybrid nanocomposites. These devices exhibit excellent optical and electrical properties (a narrow band gap (0.2 − 2eV) and respond to light spanning from the visible to infrared regimes) with high stability against mechanical stress at voltages below 0.5 V.

The potential applications of two-dimensional materials in optoelectronic memristors are vast. Firstly, they can be utilized in the field of optical storage to achieve high-capacity and high-speed data storage and retrieval. Secondly, the optoelectronic properties of two-dimensional materials make them ideal materials for photosensitive sensors, enabling the detection of light signals, image recognition, and optoelectronic measurements. Furthermore, they can be used to simulate human vision, providing higher-level functionalities for optoelectronic systems, such as facial recognition and gesture control.

2.4. Ferroelectric Memristors

Ferroelectric materials, due to their nonlinear optoelectronic characteristics and controllable resistance states, have been widely used in preparing optoelectronic memristors. Ferroelectric materials possess special crystal structures exhibiting polarization controllable by an external electric field. Typical ferroelectric materials include ferroelectric oxides (such as barium titanate, zirconium titanate) and ferroelectric polymers (such as polyvinylidene fluoride). These materials, known for their excellent electro-optic coefficients, high charge storage capacity, good stability, and ease of integration, are considered ideal for preparing optoelectronic memristors.

Research directions for ferroelectric materials include one-dimensional nanowires, two-dimensional thin films, and heterostructures. These structures not only provide high charge storage capacity but also effectively improve charge injection and transmission efficiency. Optoelectronic memristors prepared from ferroelectric materials have broad application prospects in optoelectronics, photonics, and intelligent sensors. For example, Sun [10] et al. fabricated an Au/BFO/SRO/BTO/mica structure on mica substrates, where after 10^4 bending tests with a bending radius of 5mm, the ferroelectric properties showed no significant degradation. They also achieved plasticity functions correlated with peak time and approximately 90% recognition accuracy for handwritten digits in artificial neural network simulations, indicating potential applications in wearable data storage and computation.

Zhong[11] present the Bi_2FeMnO_6 ferroelectric artificial synapse with optoelectronic response, capable of generating an excitatory post-synaptic current through ultraviolet light irradiation and subsequently modulating it via ferroelectric polarization. This enables the realization of a photoferroelectric dual-control synapse. They propose a training method based on ultraviolet excitation and ferroelectric enhancement, which significantly reduces the number of required training pulses while enhancing synaptic plasticity. Additionally, they introduce an optimized spike scheme based on ferroelectricity to endow the synapse with highly linear weight adjustment functionality. By employing this synapse in a convolutional neural network architecture, they achieve an impressive recognition accuracy of 98.80% on the MNIST dataset.

In conclusion, significant progress has been made in the preparation of optoelectronic memristors using ferroelectric materials, but challenges remain, such as improving device stability and lifespan, achieving high-performance optoelectronic conversion, and gaining deeper insights into memristor mechanisms. Future research will continue to explore novel ferroelectric materials and structural designs to achieve the preparation and application of high-performance optoelectronic memristors.

2.5. Perovskite Materials

Organic materials typically have simple fabrication processes and low production costs, while also exhibiting good flexibility. So far, resistance switching effects and switch characteristics have been observed in many devices based on organic materials, such as organic small molecule materials, organic photochromic materials, polymer materials (polyaniline, polystyrene), and composite materials.

Subramanian [12] et al. utilized the Vacuum Pressure Impregnation (VPI) method to incorporate AlOx molecules into organic small molecule SU-8 to prepare Ag/SU-8Ag/Pt memristors. By altering the permeation of AlOx, they not only achieved active control over the device switching parameters such as V_{SET}, V_{RESET}, and I_{OFF}, resulting in reduced amplitude and increased switch ratio, but also highlighted the potential of combining traditional polymer resist materials with the VPI process.

Organic photochromic materials are also a class of materials systems that exhibit photonic memristive behavior, where the mechanism involves light-induced changes in the organic molecular structure.

ISSN 2616-5880 Vol. 5, Issue 2: 64-69, DOI: 10.25236/AJMC.2024.050210

Kemp's[13] research team utilized organic photochromic material, the photoactive azobenzene polymer PDR1A, combined with ZnO nanorods to fabricate reversible photo memristors. The device structure consisted of an Al top electrode, memristive layer, and ITO bottom electrode. Experimental results demonstrated that under alternate irradiation of circularly polarized light and linearly polarized light, PDR1A polymer could undergo continuous reversible photochemical isomerization. Under circularly polarized light, the PDR1A material would expand, increasing the overall film thickness by 30%, thereby increasing the device resistance. Conversely, when subjected to linearly polarized light, the polymer material would contract, reducing the film thickness and returning to its initial state, resulting in decreased overall resistance. This photochemical reaction enabled reversible light modulation of the device's high-resistance state (HRS) and low-resistance state (LRS).

Arunagirinathan et al. utilized a single-layer polymer PFO-NPN film to prepare an Al/PFO-NPN/ITO memristor structure. This device exhibited bistable switching behavior and extremely high switch current ratio. By fitting the I-V curves of different conduction models, the charge transport of the polymer was studied to achieve trap charge-limited current (TCLC), contributing to the switch and demonstrating the bistable characteristics of the memory device. The polymer was theoretically calculated, revealing the existence of traps on the carbonyl oxygen atoms of the NPN portion.

Ren et al. [14]constructed Ag/PFTBDD-LrTPy/ITO memristor devices based on a novel copolymer PFTBDD-LrTPy. The device demonstrated representative bipolar switching phenomena, superior electrical performance, and multilevel data storage. Combining cation regulation and charge transfer characteristics, this polymer memristor could achieve various functions, including multilevel data storage, biological synaptic simulation, and basic arithmetic operations.

Organic polymer memristors are promising storage devices with significant development prospects, characterized by scalability, flexibility, low cost, and ease of handling. However, despite significant progress in organic polymer memristors in recent years, challenges such as reliability, long-term stability, environmental operation, and large-area processing remain to be addressed

3. Conclusions

Optoelectronic memristors as fundamental units for neuromorphic computing, exhibit superior performance in low energy consumption, fast signal transmission, and complex computing functions. Therefore, researchers are committed to the development of novel optoelectronic memristors and exploring their applications in synaptic bionics. In the past few years, significant progress has been made in the research of memristors and the materials used in them. The study and development of various new resistive switching materials, as well as their performance in optoelectronic memristors, provide new possibilities for the future development of electronic devices. Research on resistive switching materials mainly focuses on oxide materials, ferroelectric materials, perovskite materials, two-dimensional materials, organic materials, etc. Further exploration of new resistive switching materials is needed to improve their stability, reliability, and controllability. Additionally, by combining these materials in a rational manner, polymorphic performance can be achieved, expanding the application scope of optoelectronic memristors.

Currently, although a large number of studies have designed and fabricated optoelectronic memristors capable of achieving brain-like bionic characteristics, the preparation and application of such novel optoelectronic memristors are still in the early stages and face many challenges. Further research is needed to overcome these challenges and explore their potential applications. Future research should focus on the following aspects:

Memristors prepared from different materials are complex and diverse. The stability, reliability, and yield of the devices are still insufficient due to factors such as experimental conditions and testing environments.

The operating current of optoelectronic memristors is relatively large, leading to high energy consumption for single synaptic devices. The power consumption of highly integrated memristive arrays will be even greater. Considering the use of low-dimensional resistive switching materials with high photosensitivity and small current amplitudes can reduce power consumption to some extent.

Currently, research on individual optoelectronic memristors is increasing, but there is still insufficient work on building complex artificial neural networks. The leakage problems that may arise during the large-scale integration of memristive arrays can be solved by forming hybrid memristive arrays through serial connection of multiple devices. However, issues such as the mutual connection between multiple

ISSN 2616-5880 Vol. 5, Issue 2: 64-69, DOI: 10.25236/AJMC.2024.050210

neurons, the coordination between multiple synapses, and the adjustment of synaptic weights need to be addressed comprehensively. The technology in this area is not yet mature, and most current research is still in the simulation stage. Achieving large-scale integration of hardware will greatly promote the progress of optoelectronic memristors in the direction of neuromorphic computing.

This article reviews the materials, application areas, and future development trends of optoelectronic memristors. Optoelectronic memristors are an exciting technology that combines the characteristics of optoelectronics and memristive effects, with broad application potential. They represent one of the future development directions in the fields of optoelectronics and neuromorphic computing, providing scientists and engineers with a new research field and platform for innovative applications. With continuous technological progress and application expansion, optoelectronic memristors are expected to achieve significant breakthroughs in multiple fields, bringing more convenience and progress to human society.

References

[1] Chua L. Memristor-the missing circuit element[J]. IEEE Transactions on circuit theory, 1971, 18(5): 507-519.
[2] Zhou Z, Pei Y, Zhao J, et al. Visible light responsive optoelectronic memristor device based on CeOx/ZnO structure for artificial vision system[J]. Applied Physics Letters, 2021, 118(19).
[3] Raeis-Hosseini N, Lee J S .Controlling the Resistive Switching Behavior in Starch-Based Flexible Biomemristors[J].ACS applied materials & interfaces, 2016(11):8.
[4] Silva C , Deuermeier J , Zhang W ,et al.Perspective: Zinc-Tin Oxide Based Memristors for Sustainable and Flexible In-Memory Computing Edge Devices[J].Advanced Electronic Materials, 2023(11):9.
[5] Wang M, Cai S, Pan C, et al. Robust memristors based on layered two-dimensional materials[J]. Nature Electronics, 2018, 1(2): 130-136.
[6] Zhou Y, Liu D, Wang J, et al. Black phosphorus based multicolor light-modulated transparent memristor with enhanced resistive switching performance [J]. ACS applied materials & interfaces, 2020, 12(22): 25108-25114.
[7] Hussein,Nili,Sumeet,et al.Memristors: Nanoscale Resistive Switching in Amorphous Perovskite Oxide (a-SrTiO3) Memristors (Adv. Funct. Mater. 43/2014)[J].Advanced Functional Materials, 2014, 24(43):6733-6733.DOI:10.1002/adfm.201470279.
[8] Xu Z, Ji T, Zhang S .The cationic interstitials induced resistive switching: a case study on Mn-doped SnO_2[J]. Materials Science and Technology: MST: A publication of the Institute of Metals, 2023.
[9] Ponzina F , Rios M , Ansaloni G ,et al.A Flexible In-Memory Computing Architecture for Heterogeneously Quantized CNNs[C]//IEEE Computer Society Annual Symposium on VLSI.IEEE, 2021.DOI:10.1109/ISVLSI51109.2021.00039.
[10] Sun H, Luo Z, Zhao L, et al. BiFeO3-based flexible ferroelectric memristors for neuromorphic pattern recognition[J]. ACS Applied Electronic Materials, 2020, 2(4): 1081-1089.
[11] Zhong W M, Tang X G, Liu Q X, et al. Artificial optoelectronic synaptic characteristics of Bi2FeMnO6 ferroelectric memristor for neuromorphic computing[J]. Materials & Design, 2022, 222: 111046.
[12] Subramanian A, Tiwale N, Kisslinger K, et al. Reduced Stochastic Resistive Switching in Organic-Inorganic Hybrid Memristors by Vapor-Phase Infiltration[J]. Advanced Electronic Materials, 2022, 8(7): 2200172.
[13] Jaafar A H, Gray R J, Verrelli E, et al. Reversible optical switching memristors with tunable STDP synaptic plasticity: a route to hierarchical control in artificial intelligent systems[J]. Nanoscale, 2017, 9(43): 17091-17098.
[14] Ren Y, Lin W, Ting L, et al. Iridium-based polymer for memristive devices with integrated logic and arithmetic applications [J]. J. Mater. Chem. C, 2020, 8(47):16845-16857.