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Advanced memristive architectures based on nanomaterials for biomedical applications: a mini review

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In recent years, the interest of science in big data sensing, storage and processing has been growing fast. Nano-materials have been widely used in resistive switching devices thanks to their distinguished properties. Furthermore, they provide nano-scale dimensions and compatibility with fabrication procedures and complementary metal oxide semiconductor (CMOS) technology. Nanomaterials can also enhance the performance of memristive structures. The operation of a memristor, which enables efficient resistive switching characterized by fast response, increased storage density, and low power requirements, depends largely on nano-materials and deposition techniques. Herein, a comprehensive brief review of nano-material RRAM arrays and their application in biomedical is discussed. First, we introduce planar and array resistive switching structures. Second, we report the different nanomaterial categories that can be used in resistive random-access memories (RRAMs). Then, we focus on the integration of 3D nano-material-based memristive crossbars for in-memory computing and biosensing arrays and discuss representative applications. The exploration of nano-materials enables the development of enhanced resistive switching architectures with increased signal integrity, great speed, and ultra-high sensitivity towards thermally and electrically stable memristive biomedical platforms.

KEYWORDS

nano-materials, memristor, resistive switching, crossbar, in-memory computing, biosensing arrays

1 Introduction

Since their breakthrough in 2008, HP memristors have been broadly used in various applications (Strukov et al., 2008), including analogue and digital circuits, biosensors, artificial neurons and synapses, as well as RRAMs for memory storage (Homsi et al., 2023; Li et al., 2018; Barraj et al., 2024). Resistive switching devices, often referred to as non-volatile memories, consist of sandwiched structures evolving metal-insulator-metal materials. They store data by alternating the insulator material's resistive level between high and low states. RRAMs are characterised by rapid switching speed and ON/OFF ratio, efficient endurance, and long retention time (Bouzouita et al., 2024). These memristive operating standards are often affected by different properties of nanomaterials (Zahoor et al., 2020). In other words,

memristive benchmarks interconnect the choice of nanomaterials with memristor performance (Wang Miao et al., 2018) through the diversity of intrinsic physical and chemical reactions of materials, as well as fabrication techniques and defect engineering that benefit the development and destruction of the conductive filament within the insulator layer. Various review papers on nanomaterials for memristive architectures exist in this context. In 2018, Ahn et al. explored sp² hybridised carbon nanostructures, including fullerene, carbon nanotubes and graphene for RRAMs, focusing on their roles in electrodes, interfacial layers, resistive switching media, and memory selectors (Ahn et al., 2018). In addition, by 2020, Rehman et al. focused in their review on 2D nanomaterial-based RRAMs and their nanocomposites by covering device structure, conduction mechanisms, resistive switching properties that exceed 10 years of retention time, fabrication technologies, challenges, and future prospects (Muqeet Rehman et al., 2020). In the same year Shen et al. reviewed RRAMs in means of thin film nanomaterials, resistive switching mechanisms, and artificial intelligence applications (Shen et al., 2020). Last but not least, in 2024, Singh et al. explored memristive devices in terms of 2D nanomaterials for various resistive switching scenarios and 2D neuromorphic systems reporting their main challenges (Singh and Gupta, 2024). The implementation of RRAM generates two main functions. By overpassing conventional Van Neumann architecture, RRAM has been widely integrated for in-memory-computing and multi-array sensing. In 2019, Bankman et al. elucidated a research paper on RRAM-based in-memory devices for deep neural networking deployment (Bankman et al., 2019). This study highlights the importance of implementing RRAM in-memory processing units (IPUs) for reducing energy consumption, memory capacities, and arithmetic operations without sacrificing the system's linearity. RRAM-based multi-sensing arrays generally pave the way towards compact human sensory perception for neural therapeutics and prosthetics. In 2019 Zhou et al. fabricated an organic RRAM (ORRAM) array for vision in-sensor application to achieve in-memory image sensing and neuromorphic pre-processing with high recognition accuracy, efficient edge computing and less complex circuitry (Zhou et al., 2019). Different from the existing review papers, in this work, we first report briefly the different memristive structures. Secondly, we classify the nanomaterials employed in the different parts of the RRAMs according to their dimensions. Then our interest focuses on reviewing the integration of RRAMs for in-memory computing and multi-sensing applications towards diversified memristive biomedical applications.

2 Introduction to RRAM

2.1 RRAM structure

Various RRAM structures have recently been investigated to enhance performance and address emerging computational and storage challenges. This paper discusses three primary RRAM architectures: (a) metal-insulator-metal (MIM) or planar, (b) Crossbar arrays, and (c) three-dimensional (3D) RRAM.

2.1.1 Planar structure

In 1967, Simmons et al. introduced the first proposed MIM structure (Figure 1a) in which resistive switching is attributed to the

formation and rupture of conductive filaments in the insulating layer (Simmons and Verderber, 1967; Sawa, 2008). The MIM structure has shown high endurance $>10^{12}$ cycles, extended data retention (>10 years), and fast switching speeds <10ns (Ahn et al., 2015; Lee et al., 2011; Kim et al., 2023; Fang Lu et al., 2021). Nevertheless, MIM architectures are limited in scalability due to lithographic limits and increased variability at small feature sizes (Wang et al., 2007; Stelling and Retsch, 2018).

2.1.2 Crossbar array structure

In crossbar array structures (Figure 1b), RRAM cells are placed at the intersections of perpendicular electrode lines, allowing a higher integration density than the planar configuration. Crossbar arrays have the potential for cell sizes as small as $4F_2$, where F is the minimum feature size, resulting in storage densities exceeding 100 Gb/cm² (Ali et al., 2016). However, crossbar arrays are plagued by sneak path currents that corrupt stored data and can lead to erroneous readouts (Huang et al., 2011; Son and Min, 2014). To overcome this problem, selector devices, including diodes or transistors, are added to RRAM cells to control sneak currents and enhance the reliability of the array (Linn et al., 2010; Yin Chee et al., 2022).

2.1.3 3D RRAM structure

3D RRAM structures (Figure 1c) utilize vertical space in the three dimensions to construct multilayer memory arrays and increase storage capacity with a reduced footprint (Deng et al., 2013). Its implementations can be broadly categorized into (i) Horizontal Stacked 3D RRAM (HRRAM) and (ii) vertical (VRRAM) schemes, which are distinguished to scalability and fabrication complexity (Kim and Li, 2020).

HRRAM is a memory architecture composed of multiple planar 2D RRAM layers superimposed as shown in Figure 1cII. (Chen et al., 2021; Wang et al., 2015). Wang et al. fabricated a 3D RRAM cube by strategically stacking a 2D MoS₂ layer, which proved a promising pathway for high-density neuromorphic computing systems (Yoon et al., 2023). Despite the innovative design of HRRAM, some critical technological challenges have been reported (Wang Chenyu et al., 2023). One of the significant issues is the fabrication complexity (Kim and Li, 2020; Wang Chenyu et al., 2023). Then, the manufacturing overhead indicates increasing stack quantities, leading to linear cost increase (Kim and Li, 2020; Wang Chenyu et al., 2023; Park et al., 2022). Finally, scalability constraints have been mentioned due to the 2D layer nature compared to VRRAM (Wang Chenyu et al., 2023; Park et al., 2023).

The conventional horizontal crossbar array structure is vertically extended for 3D VRRAM (Figure 1cIII; Chen et al., 2022). It is organized in two configurations: The plane word line (WL) structure and the even/odd WL structure (Kim and Li, 2020). It shows increased storage capacity with a small footprint (Kim and Li, 2020; Yue et al., 2014). In particular, compared to 2D crossbar array, 3D VRRAM with an even/odd WL structure occupies only $256F^2$, 6 times less than the total size of an 8×8 image when processed (Yue et al., 2015). This shows higher bit-cost scalability than other 3D RRAM structures, and is most promising for deploying neural networks (Yue et al., 2014; Yue et al., 2015; Yu et al., 2016).

Structural innovations introduced in these devices give rise to next-generation memory solutions in diverse computing applications (Ielmini and Wong, 2018).



2.2 Nano materials in RRAM structures

Nanomaterials thanks to their properties at the nanoscale, have been revolutionizing modern science and technology. Unlike their rival bulks, Nano-materials's size and shape determine their unique optical, chemical, thermal, electrical, mechanical, and catalytic properties (Mekuye and Abera, 2023). Nanomaterials are generally known for their dimensional configuration



configuration with memory cells at word-line bit-line intersections. (c.II): Horizontal 3D stacking (HRRAM). (c.III): Vertical 3D architecture (VRRAM). (d) 0D nano-materials used in RRAMs. (e) 1D nano-materials used in RRAMs. (f) 2D nano-materials used in RRAMs. (g) Proposed M3D-LIME Chip Architecture with multiple layers. (h) RRAM-based Processing-in-Memory (PIM) Architecture. (i) RRAM array for multisensory perception. (j) Device-level calibration of the PSA sensing array baseline. (k) Sneak path effect that corrupts the stored data and lead to erroneous readouts. (l) examples of RRAM challenges. (m) Proposed solution: synthesis methods and applications of metal-organic frameworks (MOFs). (a) Reprinted from Sawa (2008). Copyright (2008), with permission from Elsevier. (b, c, g, h, j, k, l, m) reprinted with modification and permission. "Creative Commons Attribution" from (Chen et al., 2024; Wang Chenyu et al., 2023; Wang Hongzhe et al., 2023; Li Yijun et al., 2023; Tzouvadaki et al., 2020; Shi et al., 2020; Adam et al., 2018; Li et al., 2024). (i) Reprinted from Zhu et al. (2022). Copyright (2022), with permission from John Wiley and Sons.

diversification ranging between 1 and 100 nm (Baig et al., 2021). These invaluable features pave the way for emergent fields of applications citing, energy storage and conversion (Yu et al., 2024), environment pollutants degradation (Saroha et al., 2023), disease diagnosis and drug delivery (Karnwal et al., 2024; Singh et al., 2022; Singh et al., 2023), as well as memory architectures like

RRAMs (Xu and Liu, 2024). In general, nanomaterials are important for RRAMs' operational characteristics (Xiao et al., 2023). Indicatively, various memristive metrics such as resistive switching behaviour, ON/OFF ratio, set/reset voltages, retention time, endurance and power consumption (Zayer Fakhreddine et al., 2019) are directly linked to the RRAM nanomaterial properties, namely, the electrical, physical, thermal and mechanical properties (Dongale et al., 2016; Shen et al., 2020; Das et al., 2023). Hence, it is essential to study the different memristive materials dimensional configuration categories (0D, 1D, 2D) (Panisilvam et al., 2024). Numerous nano-materials can be employed for the fabrication of RRAMs, the most common ones are metal oxides, perovskites, and organic materials. Furthermore, other nanomaterials contribute to RRAM modelling, for instance, transition metal oxides (TMOs), and transition metal dichalcogenides (TMDs), etc. Table A1 of the appendix and Figures 1d–f outline the categories of nanomaterials used in RRAMs according to their dimensions.

2.2.1 0D nano-materials in RRAMs

0D carbon nano-materials like carbon nano-dots, graphene quantum dots, and fullerenes assist in memristive functional layer growth through the generation of directional conductive filament formation (Yang Fan et al., 2024). Furthermore, metallic nanoparticles (MNPs) like copper (*Cu*) and sliver (*Ag*), are proven to exhibit an important influence on the memristive switching with alternating voltage under 4 mV showing extremely low variability (Liu et al., 2022; Jean Yoon et al., 2019). Within the framework of their study, Wang et al. demonstrated that $NiFe_2O_4$ MNP-memristors have shown low Set/Reset voltages less than 2.15 V/ 0.83V, an elevated retention time of 10³ s, and a 10³ of endurance cycles (Wang et al., 2022).

2.2.2 1D nano-materials in RRAMs

1D nano-materials, thanks to their nanoscale size, unique transport and high electric conductivity at low voltage, are widely used in RRAMs (Zhang et al., 2023). For instance, 1D carbon nano-materials are proven to be good candidates for elaborating highly performing RRAMs because of device stability and weight tunability (Patil et al., 2024). Moreover, metal oxide nanowires and nanorods, such as zinc oxide (ZnO), titanium dioxide (TiO₂), nickel oxide (NiO), and tungsten trioxide (WO₃) are employed in memristors to improve the volume to surface ratio (Lu and Lieber, 2007) and scalability, as their bottom-up selfassembly capability guarantees high-precision control of the dimension (Milano et al., 2019). In 2018, Ting-Kai Huang et al. reported Ni/NiO/HfO2-based memristive 1D-RRAM with endurance of 200 cycles, retention up to 107s, ON/OFF ratio close to 104, and set/rest maximum voltages equal to 3V and 2 V respectively (Huang et al., 2018).

2.2.3 2D nano-materials in RRAMs

2D nano-materials are known for ultra-thin, flexible, and layered structures (Muqeet Rehman et al., 2020). 2D TMDs semiconductors such as tungsten disulfide (WS_2) and molybdenum diselenide ($MoSe_2$) exhibit high scalability and integrity. Thus, they are used for high-density and low-powerconsumption RRAMs (Xiang et al., 2018). 2D graphene oxide (GO), reduced graphene oxide (rGO), and hexagonal boron nitride (h-BN), also called white graphene, show potential utility for performant RRAM cells. The origin of memristive behaviour in these materials is the vacancies-initiated conductive filament alternative construction (Qian et al., 2016; Kim et al., 2015). In 2023, Yan et al. modelled RRAM based on strontium titanate ($SrTiO_3$) perovskite oxide thin film highlighting the set voltage and endurance of 2V and 100 cycles, respectively (Yan et al., 2023a). Vertically stacked 2D nanomaterials can enhance the performance of RRAMs as they guarantee distinguished uniformity and resistive switching (Huh et al., 2020). In 2019, Zhang et al. (2019) performed a study on vertical TMDs, molybdenum ditelluride ($MoTe_{2-}$) RRAM with retention time, set voltage and, power consumption of 10³ s, 2.3 V, and 2.3 mW respectively.

3 3D RRAM for in-memory computing for biomedical applications

3D RRAM-based systems provide better performance and energy efficiency than traditional computing architectures by computing directly within the memory array (Yan et al., 2019). Several 3D RRAM-based in-memory computing architectures have been proposed and demonstrated in recent years. Wang et al. propose a 3D RRAM-based processing-in-memory (PIM) architecture (Figure 1g) that integrates 3D RRAM with CMOS logic, enabling in situ computation with reduced data movement and energy (Wang Hongzhe et al., 2023). For heavy-load convolutional neural network (CNN) algorithms, the RRAM PIM generally outperforms other architectures in terms of identification rate, speed, and image size (Burr et al., 2017). Building on this, Yao et al. developed a 3D RRAM-based neuromorphic computing system for more efficient processing and analyzing of large-scale datasets, such as images and time series. This system achieved a classification accuracy of 97.8% on the Modified National Institute of Standards and Technology (MNIST) dataset (Peng et al., 2020). Hao et al. presented a study of computing-in-memory macro based on 3D RRAM in 2022. Their work was based on a 3D VRRAM in which they introduced a two-kilobit non-volatile computing-in-memory (nvCIM) macro. They demonstrated an energy efficiency of 8.32 teraoperations per second per watt on 3D vector-matrix multiplication operations. This advancement can facilitate more efficient management of medical imaging data, improving diagnostic accuracy (Wang Tian-Yu et al., 2021). It was found that the brain Magnetic Resonance Imaging (MRI) edge detection performance and the inference accuracy on the Canadian Institute for Advanced Research (CIFAR-10) dataset were improved compared to conventional approaches (Wang Tian-Yu et al., 2021). In 2023, Li et al. recently introduced the monolithic three-dimensional integration of hybrid memory architecture based on RRAM, named M3D-LIME. This M3D-LIME chip (Figure 1h) demonstrated approximately 96% accuracy on the Omniglot dataset with $18.3 \times higher energy$ efficiency compared to graphics processing unit (GPUs), showcasing its potential for efficient processing of complex healthcare datasets and potential use of this technology for medical image processing and efficient analysis of MRI or computed tomography (CT) scans (Li Yijun et al., 2023). In the same year Ge Shi et al. proposed a two-kilobit CIM macro based on an 8-layer 3D vertical RRAM to implement complex neural networks for drug discovery processes and potentially helping in identifying the new therapeutic compounds (Wang Chenyu et al., 2023).

4 3D RRAM for multi-sensing arrays for biomedical applications

RRAM architectures can be integrated as chemical signal sensors for disease biomarker detection (Bouzouita et al., 2024) and drug delivery and electrical signal sensors for human multisensory perception (Tzouvadaki et al., 2023). Indeed, thanks to their efficient ON/OFF ratio, fast response, low power requirements, low cost, etc. RRAMs can be implemented as highly performing bio/chemical sensing arrays (Tzouvadaki et al., 2020b). In this case, each RRAM layer or section is transformed into a single sensing device, leveraging advancements in memristive nano-materials and biomedical technologies, like lab-on-chip perspectives to simultaneously harvesting multiple responses. In 2018, Adeyemo et al. simulated the detection of three different gases with an 8 \times 8 memristive crossbar array. The simulation results highlighted a ten-times increase in accuracy compared to single biosensing devices (Adeyemo et al., 2018). In 2020, Ioulia Tzouvadaki et al. demonstrated a chemical biosensor array (Figure 1i) directly transducing Prostate Specific Antigen (PSA) cancer biomarker concentration levels to discrete memory states, expressing a device-level calibration of the sensing array baseline (Tzouvadaki et al., 2020a). Moreover, in 2023, Doowon Lee et al. developed a highly accurate zirconium nitride (Zr_3N_4) gas sensor array with efficient power nitric oxide (NO) gas detection and increased accuracy by 2.5% (Lee et al., 2023). This study is dedicated to artificial olfactory sensory perception (Milozzi et al., 2024), (Wang Tong et al., 2021). In fact, parallel to multi-chemical sensing applications, RRAMs are considered a promising technology for artificial multi-sensory perception (Kwon et al., 2024) as they can mimic biological neural systems because of their characteristics such as synaptic plasticity, analogue data processing, complex parallel computing (Moon et al., 2019), etc. In this context, the feasibility of multi-sensory chips (Figure 1j) (Li Zhiyuan et al., 2023) is proposed to overcome some neural impairments by advancing promising neural sensory prosthetics. In 2023, Xiaobing Yan et al. implemented a ferroelectric memristor crossbar for visual-tactile multimodal sensory. This study shows high stability with an endurance value of 1010 and low field voltage between -1.3 V and -2.1 V (Yan et al., 2023b).

5 Challenges and future directions

5.1 Challenges of 3D RRAM technologies

While memristive crossbar arrays offer considerable potential in the biomedical field, they face numerous obstacles that could impact their development and implementation. While advancing RRAMs, certain critical barriers have emerged in both vertical and horizontal directions. In their work, Gina C. Adam et al. reported potential challenges of memristive matrix integration (Figures 1k, l) citing variability, density, latency, and technology feasibility (Adam et al., 2018). For instance, as RRAM arrays simulate processing operations, errors may appear due to large device variability. Indeed, memristive crossbar arrays often exhibit vast variability due to stoichiometric inconsistency of inharmonious temporal memristive dimensions. Specifically, the variability of selected cross-points within the RRAM causes signal decay (Chen and Lin, 2011). A.P. James et al. emphasized that the variability problem originates either from device-to-device variations, or the nonlinearity of latency and programming. In memristive crossbars, the current flowing through the metal wires (Figure 1k) causes the reduction of voltage drop through the structure. This problem induces the damage of metal wires. The elevation of resistance is a further challenge causing thermal crosstalk and energy loss. All these circumstances initiate signal integrity obstacles (Li et al., 2021; Xu et al., 2015). In general, RRAM challenges reflect on the correct functioning of memristive biosensing devices, therapeutic systems and neural prosthetics since these applications necessitate specific performance metrics citing the response time, sensitivity, stability, reliability, etc., which are linked originally to used memristive device and nanomaterials (Yang Yulong et al., 2024; Zhu et al., 2024).

Various works have been addressing RRAM challenges and proposing solutions. Several nanomaterials have shown a potential to solve some remaining RRAM bottlenecks. For example, it is proved that titanium nitride, while deposited as an atomic layer (ALD-TiN), reduces the penetration of titanium (Ti) atoms to hafnium oxide HfO_x . Thus, the layer enhances the oxygen vacancy generation and conductive filament forming (Fang et al., 2018). In addition, innovative bias scheming and interconnect nanomaterials like CNT can tackle voltage dropping and crosstalk challenges ensuring uniform voltage and thermal distribution (Zayer F. et al., 2019). Equally important, structural and training compensation techniques are adopted to enhance the overall RRAM performance (Pappachen James and Chua, 2022). For instance, the integration of modular arrays, or tiled crossbars to decrease the sneaky-path current and employing offline training to improve RRAM variability and sensitivity approaches.

5.2 Future directions

There are several significant research opportunities in the future for the 3D RRAM nanomaterial memristive architectures for biomedical applications. Among these, the interconnecting and evolving of new nanomaterials and 2D materials with CIM sensing, and computing properties are paramount. Materials like TMDs, graphene and metal-organic frameworks (MOFs) (Figure 1m) bear the pre-scientific potential to improve the device performance high-density, in multifunctional biomedical implants, diagnostics and real-time health monitoring devices (Zahoor et al., 2023). Specifically, MOFs, with their high surface area and tunable porosity, improve the switching behaviour of RRAM devices through increased ion mobility and low energy consumption. This makes them suitable for neuromorphic systems in implantation devices thus making them potential implant sources (Zhang et al., 2022; Li et al., 2024). Improving these materials to get the best switching characteristics is very important for protracted implantable devices (Shen et al., 2020). However, reducing device variability is an issue that has not yet been solved successfully.

More investigations should be dedicated to the reliable suppression of device-to-device and cycle-to-cycle fluctuations to promote the stability of neuromorphic designs for neural computing: sensory signal processing in real-time, control of prosthetic devices, etc., (Shen et al., 2020; Donati and Valle, 2024; Buccelli et al., 2019). The bio-electronic interface is another area with considerable potential for advancement. Therefore, it is desirable that future studies must further extend efforts in stable and long-lasting operation of these interfaces in physiological conditions for the further effectiveness of implantable devices (Zahoor et al., 2023). Recent research on combining RRAM with CMOS chips has shown that on-chip processing is possible at an energy density close to 10 fJ/operation - a critical feature for implantable biomedical applications (Shen et al., 2020). Prospective work should focus on how to continue driving energy consumption down while, at the same time, enhancing the computational abilities of the end-devices, so that in the future it can be possible to perform local analysis and decision making of the data (Li Yijun et al., 2023; D'Agostino et al., 2024; Huo et al., 2022; Aziza et al., 2021). Furthermore, identifying ways in which 3D RRAM devices may be applied for closed-loop therapeutic systems, personalized healthcare, and AI-based diagnosis is another interesting research direction. Additionally, when combined with other new technologies like flexible electronics and biodegradable materials the 3D RRAM may revolutionize the wireless implantable and wearable medicine (Zahoor et al., 2023). Overall, these aspects can open new paths for the use of 3D RRAM nanomaterial memristive architectures in biomedical applications, significantly benefiting healthcare.

Author contributions

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Appendix

TABLE A1 Nano-materials for RRAM applications.

Material	Device	ON/OFF ratio	V reset (V)	V set (V)	E (cycle)	RT(s)
0D Nano-materials	Ag/Ag: Ag ₂ O/Ag (Jean Yoon et al., 2019)	10 ⁹	< 0.3	> - 0.6	10 ²	~ 10 ⁵
	ITO/CdS QDs PV P/Al (Betal et al., 2023)	> 10 ⁴	-0.72	1.08	> 10 ²	> 10 ⁴
	ITO/HfO _x /MoS ₂ Pd NPs/ITO (Wang et al., 2018b)	> 10 ³	< -0.8	-0.1 to 0.3	200	10^{4}
1D Nano-materials	Ag/ZnO doped Ga_2O_3/Au (Li et al., 2022)	_	> -1.49	> 0.51	60	10^{4}
	ITO/GZO/ZnO/ITO (Huang et al., 2014)	10 ²	-7.2	5.7	7x10 ³	105
	Cu/TiW/Nds/ZnO/ITO/Glass (Singh et al., 2018)	30	-2	1.5	10 ²	10 ³
2D Nano-materials	ITO/PVA GO/ITO (Wen et al., 2020)	> 10	0.2	-0.2	5x10 ²	104
	Ag/InSe/Ag (Tang et al., 2021)	$4.5 x 10^4$	-0.7	0.3	3x10 ²	3.5x10 ³
	<i>Pt/h – BN/Ag</i> (Li et al., 2016)	10 ⁸	-0.1	0.3	107	-

V Reset: reset voltage, V Set: set voltage, E: endurance, RT: retention time.