Recent Advances in Flexible Resistive Random Access Memory

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Abstract: Flexible electronic devices have received great attention in the fields of foldable electronic devices, wearable electronic devices, displays, actuators, synaptic bionics and so on. Among them, high-performance flexible memory for information storage and processing is an important part. Due to its simple structure and non-volatile characteristics, flexible resistive random access memory (RRAM) is the most likely flexible memory to achieve full commercialization. At present, the minimum bending radius of flexible RRAM can reach 2 mm and the maximum ON/OFF ratio (storage window) can reach $10^8$. However, there are some defects in reliability and durability. In the bending process, the cracks are the main cause of device failure. The charge trap sites provided by appropriate doping or the use of amorphous nanostructures can make the conductive filaments of flexible RRAM steadier. Flexible electrodes with high conductivity and flexible dielectric with stable storage properties are the main development directions of flexible RRAM materials in the future.

Keywords: flexible RRAM; resistance transition mechanism; failure mechanism; flexible material system; fabrication process

1. Introduction

Flexible RRAM is a new non-volatile information storage technology [1] which has the characteristics of high speed, durability, stability, low power consumption, the non-volatile and a wide selection of materials, and it is regarded as one of the important candidates for future flexible memory devices. The integration of flexible RRAM is usually divided into the active array and passive array [2]. The active array eliminates cross-talk between storage cells but requires high-temperature preparation and cannot achieve three-dimensional (3D) integration [3,4]. In contrast, the passive array has a simple fabrication process and can realize 3D multilayer integration. Meanwhile, the minimum storage unit of the passive array is smaller than that of the active array. Therefore, the passive array with a cross structure is the lowest cost integration of memory devices at present, and it is also the focus of flexible RRAM research in the future [5–7]. The organic-inorganic composite medium has both stable storage performance and good flexibility. The metal electrode has good conductivity, the conductive oxide electrode has high transmittance, and the carbon/nitrogen electrode has excellent flexibility. It can be seen that the material system of flexible RRAM has great development potential. In this paper, the research status of flexible RRAM is summarized. The second part summarizes its basic working principle, the third part focuses on the current research status of the flexible RRAM material system, the fourth part briefly introduces the preparation process of flexible RRAM and
the fifth part summarizes the challenges and opportunities for the further development of flexible RRAM.

2. Basic Working Principle of Flexible RRAM

2.1. Structure of Flexible RRAM

Unlike transistor-based memory, RRAM does not need a complex cell structure such as a thin film transistor (TFT) to integrate with complementary metal oxide semiconductor (CMOS) technology. Although both TFT and RRAM are devices composed of several layers of films, a TFT is a device based on transistor structure, which is generally composed of a gate electrode, dielectric layer, active layer, source electrode, drain electrode and substrate. The structure is relatively complex, which makes it difficult to be flexible. RRAM is a simple “electrode/dielectric/electrode” sandwich structure, where the dielectric is the carrier for resistance transformation, so it is also called storage medium. Flexible RRAM uses a passive cross-array structure (a cross-array structure is considered to have the highest theoretical integration). Each cross node occupies only a 4F² area (F: minimum characteristic size) of the storage cell (Figure 1), thus easily achieving high-density integration to improve storage density [8]. The storage medium uses the high-resistance state (HRS) and low-resistance state (LRS) to represent the 0 and 1 of binary code, respectively [9]. The switch category can be divided into write-once-read-many-times (WORM) [10], the unipolar switch [11] and the bipolar switch [12,13].

Figure 1. (a) Schematic of a flexible crossbar resistive switching memory array bent with radius R; (b) schematic memory cell structure with measurement configuration.

2.2. Resistance Transition Mechanism of Flexible RRAM

In flexible RRAM, the main resistance transition mechanisms are the interface barrier mechanism [14–16] and conductive filaments mechanism [17,18].

The interface barrier mechanism means that the resistance change in devices is controlled by the height of the Schottky barrier or the width of the depletion layer at the interface, that is, the higher the Schottky barrier or the width of the depletion layer, the higher the resistance of devices, showing HRS. Conversely, the device exhibits LRS. The change in barrier height is usually caused by the accumulation of charge or migration of oxygen ions under the action of the electric field [16]. The mechanism is based on the quantum tunneling effect, and its LRS and HRS correspond to Poole–Frenkel carrier transport [14] and Schottky launches [15], respectively.

The conductive filament mechanism means that the device forms conductive filaments that run through the whole dielectric material and connect the two electrodes, depending on the migration of metal ions or oxygen vacancies. When the forward voltage is applied to the active metal electrode, the metal atoms will become metal ions (M → ze⁻ → M⁺) and then migrate to the inert electrode under the action of the electric field. When the metal ions pass through the dielectric and reach the inert electrode, they will go back to metal atoms (M⁺ + ze⁻ → M). Depending on the migration of metal ions, the device forms conductive metal filaments that connect the two electrodes, and the device is at LRS. If the voltage is
applied in the opposite direction, it will disconnect the conductive filaments and transform the device to HRS. Another conductive filament based on oxygen vacancy is mainly used to explain the resistance transition mechanism of oxide. Under the action of the electric field, oxygen ions migrate in the dielectric and form oxygen vacancies, resulting in conductive channels. The device of Ag/HfO$_2$/Pt was studied to research the conductive filament mechanism by Saadi [17]. It was found that the implanted electrons generate oxygen vacancy on the inert electrode (Pt) and the active electrode (Ag) through the formation of Ag$^+$ to form conductive filaments so that the device has bistable characteristics.

2.3. Failure Mechanism of Flexible RRAM

Flexible RRAM is bound to be bent repeatedly during use. In the process of bending, the device is prone to cracks, delamination, thermal effect accumulation and other problems, so the device loses the bistable characteristics, resulting in the failure phenomenon.

Shang [19] found that the generation of cracks in the electrode and dielectric layers affect carrier transport during the bending process, which is the main cause of device failure. The generation of cracks makes it difficult to form conductive filaments, and the formed conductive filaments will also break under the influence of cracks (Figure 2). With the increase in bending times, the density of cracks will increase, which will destroy the formation and stability of conductive filaments and then affect the stability of devices. On the other hand, according to the research of Zou [20], when the opening current is large, the conductive filaments are relatively thick, which will produce large Joule heat. At the same time, with the increase in bending times and cyclic scanning times, the Joule heat generated in the device will accumulate. The conductive filaments formed in the device will be partially dissolved due to the Joule heat effect, resulting in device failure. In addition, if the device is in the same state for a long time, it may cause the electrode material to deposit to the electrode at the other end, which will cause delamination and lead to device failure.

![Figure 2. Reason of flexible RRAM failure: the generation of cracks.](image)

3. Material System of Flexible Resistive Memory

3.1. Flexible Dielectric Materials

As the carrier of resistance transformation and information storage, the choice of storage media is very important to device performance. Flexible RRAM has a wide selection of media layers, which can be divided into three categories: inorganic materials, organic materials and organic-inorganic composite materials.
3.1.1. Flexible Inorganic Materials

In recent years, a large number of flexible inorganic materials with resistance transformation effects have been applied to the dielectric layer of flexible RRAM, including binary metal oxides (TiO$_2$, NiO$_x$, ZnO$_x$, AlO$_x$, WO$_x$, HfO$_x$, MoO$_x$, etc.), multimetal oxides (HfTiO$_4$, ZnSnO$_x$, etc.), solid electrolyte (Ag$_2$Se, Ge$_2$Sb$_2$Te$_5$, MoS$_2$, etc.) and other flexible inorganic materials (C$_3$N$_4$, BN, ZrN, Si$_3$N$_4$, SiO$_x$, amorphous silicon, graphene oxide, etc.).

In flexible inorganic medium materials, binary metal oxides with simple chemical composition and excellent resistance are the most common. Binary metal oxides are favored by researchers because of their simple structure, controllable composition, simple preparation, stable chemical properties, high ON/OFF ratio and compatibility with CMOS technology. The resistance transformation effect of most binary metal oxides is based on the valence state change caused by oxygen ion migration under an electric field or the conductive channel generated by oxygen vacancy accumulation. Few show the resistance transformation effect by forming metal conductive filaments, such as Ti-doped ZnO dielectric and Al-doped HfO$_x$ dielectric. At present, there is a flexible RRAM based on binary metal oxides with excellent performance. The characteristics of the binary metal oxides are shown in Table 1 below:

| Dielectric Materials | Polarity | ON/OFF Ratio | Durability | Retention Time | Number of Bending Cycles | Bending Radius |
|----------------------|----------|--------------|------------|----------------|--------------------------|---------------|
| TiO$_2$ [21]         | bipolar  | $10^3$       | $10^2$     | $2 \times 10^3$ s | 40                        | 9 mm          |
| NiO$_x$ [22,23]      | bipolar  | $10^6$       | $10^2$     | $10^4$ s       | 120                      | 10 mm         |
| ZnO$_x$ [24]         | bipolar  | $10^6$       | $10^2$     | $10^4$ s       | 98                       | 20 mm         |
| AlO$_x$ [25,26]      | bipolar  | $10^6$       | 30         | $3 \times 10^4$ s | 100                      | 10 mm         |
| WO$_x$ [28,29]       | bipolar  | $10^5$       | $2 \times 10^3$ | $10^5$ s       | 100                      | 15 mm         |
| HfO$_x$ [30,31]      | bipolar  | $1.2 \times 10^3$ | $5 \times 10^8$ | $10^4$ s       | $10^3$                   | 5 mm          |
| MoO$_x$ [32,33]      | unipolar | $2.38 \times 10^5$ | 50         | $2 \times 10^5$ s | 100                      | 10 mm         |

Durability is measured by the number of switching cycles the device undergoes under DC conditions; N/R: not reported.

As shown in Table 1 above, the resistive memory based on binary metal oxides has an excellent performance in retention time, flexibility and other aspects. The ON/OFF ratios of NiO$_x$, ZnO$_x$ and AlO$_x$ are high, owing to the high interface barrier with the electrode. LRS and HRS are easy to distinguish in the working process, resulting in small errors, but the flexibility of these is relatively low and is not conducive to the application of flexible devices. The material of HfO$_x$ has an outstanding advantage in flexibility, with the lowest bending radius and the highest number of bending cycles. Although the ON/OFF ratio is relatively low, it is within the acceptable range. In general, HfO$_x$ is the most widely used as flexible inorganic medium because of the best mechanical properties. Hf$^{2+}$ has large mobility and solid solution rate, which makes it form conductive filaments more easily, so it can withstand greater bending strain. However, the more detailed resistance transition mechanism of these binary metal materials is not clear, so it could still be a hot spot in this field to research the resistance transition mechanism for guiding the selection principle of flexible dielectric materials.

The multimetal oxides used in flexible RRAM are usually transitioning metal oxides, and these oxides have good performance. For example, a two-layer TiO$_2$:HfO$_2$ film for flexible storage media was reported by Ismail [34]. The storage medium exhibits superb $10^3$ direct-current (DC) switching cycles, outstanding $10^7$ pulse endurance and high-thermal stability ($10^5$ s at 125 °C), and its cross-array memory even has the potential to be used in biomimetic neuromorphic systems (Figure 3). Flexible RRAM based on multimetal oxides is mostly used in the field of synaptic bionics, which requires high durability. The
requirements for its flexibility are low, relatively. However, the composition content of RRAM based on multicomponent oxides is difficult to control in the fabrication process, the fabrication process is complex and most of them are incompatible with the CMOS process, so multicomponent metal oxides are rarely used as dielectric materials.

![Figure 3. W/TiO₂/HfO₂/TaN device: (a) schematic illustration displaying the cross-array structure; (b) DC endurance characteristics for 10⁶ cycles; (c) excellent pulse endurance for over 10⁷ switching cycles attained with 10 μs programming pulses; (d) retention tests made at LRS and HRS show no evident change for over 10⁴ s at 125 °C [34].](image)

Solid electrolytes are usually ionic conductors with high electron mobility. The resistance transformation mechanism of the material is based on the formation and disconnection of conductive metal filaments caused by the redox reaction of cations in electrolytes. Its outstanding characteristic is high flexibility, and it is also a kind of flexible RRAM with great potential. The chalcogenides most commonly used as solid electrolytes, including Ag₂Se [36], Ge₂Sb₂Te₅ [37], WSe₂ [38], etc., are all storage media with excellent performance. Their ON/OFF ratio is generally more than 10⁴ and their durability is generally more than 10⁶. In order to further improve the mechanical properties of flexible RRAM devices, a kind of inorganic sulfide compound and its composite materials with inorganic oxides have been developed. Using Al₂O₃ and CdS as the storage medium by Ju [49], the medium can withstand 100 times of repeated bending, and the minimum bending radius can be reduced to 5 mm (Figure 4). The Al₂O₃ layer between the Ag top electrode and the CdS layer act as the barrier that prevents Ag from significantly migrating into the CdS layer. The Al₂O₃ layer with appropriate thickness prevents the unnecessary migration of a large amount of Ag into the CdS layer to control the thickness of filaments, which prevents delamination and reduces thermal effects. Although solid electrolytes have excellent performance, chalcogenides are not easy to manufacture. They are complex and costly, and their performance is similar to that of binary metal oxides. Therefore, solid electrolytes are not widely used relatively.
As for other flexible dielectric materials, SiO$_x$-based flexible RRAM was reported by Li [42], with the ON/OFF ratio of $10^5$ after 2000 bending cycles at a 2.5 mm bending radius. The device uses vacuum ultraviolet irradiation to prepare SiO$_x$ films from perhydropolysilazane. The report inferred the flexibility is attributed to the high uniformity and compactness of the SiO$_x$ layer from vacuum ultraviolet irradiation. The SiO$_x$ layer prepared by this approach not only avoids interface mismatch but also guarantees low internal stress inherently due to its mass increment process. In addition, another material that is ideal for use as flexible RRAM is graphene oxide (GO). In the latest study, high-performance flexible RRAM with GO was reported by Tariq [46]. This graphene together with its distinctive inter-layer oxygen diffusion path enables excellent oxygen ion/vacancy diffusion. Without an interfacial redox reaction, oxygen ions can diffuse to form conductive filaments with two inert metal electrodes by applying bias voltage. Meanwhile, the hydroxyl increases the bending modulus of GO, making it more flexible than graphene [50]. The device has shown superior performance, including a high ON/OFF ratio of $10^5$, a long-term retention of $10^6$ s, reproducibility over $10^4$ cycles and long-term flexibility at a bending radius of 4 mm, indicating that the material has great potential for flexible RRAM.

Figure 4. Al$_2$O$_3$/CdS film: (a) $R = 5$ mm; (b) the I–V characteristics before and after 40 or 100 bending cycles; (c) endurance characteristics up to 100 bending cycles [51].

Although inorganic storage media have standard semiconductor compatibility, stable chemical properties and high ON/OFF ratios, their inherent fragility makes devices work only under relatively low bending conditions, which cannot meet the bending radius requirements of flexible memory [51,52]. To improve the flexibility of inorganic storage media, researchers have introduced nanostructures. Ji [52] used an electrochemical anodizing process to convert the metal film into metal oxide, which makes WO$_3$ resulting in nano porous (NP) structures. NP structures have the potential to be used in flexible RRAM as mobile ion or charge trap sites and vacancy-assisted switching materials. The device using this structure shows an ON/OFF ratio of $10^3$ and a stable retention time over $5 \times 10^3$ s, with an endurance over $10^3$ cycles. More importantly, the device has a bending radius of 5.53 mm and shows robust switching behavior after $10^3$ bending cycles (Figure 5). A novel HfO$_x$ medium with amorphous nanostructures was developed by Shang [19]. The device can continuously erase more than $10^7$ at a bend radius of 6 mm, and the randomness of resistance is less than 4.3%, but at the bending radius of 4.5 mm, the reset voltage fluctuates and the device becomes unstable. In addition, according to the report’s extrapolation results, the device can stably store data at a bend radius of 6 mm for more than 10 years (about $3 \times 10^8$ s). In these nanostructures, amorphous regions often appear between the nanograin boundaries, which accumulate defects such as oxygen vacancies. They help to release the strain accumulated during the bending operation and
induce the stable evolution of the nano conductive filaments, thus improving the bending performance of the device.

3.1.2. Flexible Organic Materials

In order to make flexible RRAM more flexible, organic materials with excellent mechanical properties are increasingly being used as resistance dielectric layers. Flexible organic materials have excellent flexibility due to the strong cross-linking between their molecules. Moreover, they have become important candidates for flexible electronic devices due to their characteristics of high flexibility, large-area film forming and light weight. This material relies on functional groups to produce ions to form conductive filaments. To date, many materials have been used in flexible RRAM, including polymer materials (chitosan [53], lignin [54], poly(paraxylene [55], polyimide (PI) [56], polyethylene glycol dimethacrylate (pEGDMA) [57], poly (1,3,5-trimethyl-1,3,5-trivinyl cyclosiloxane) (pV3D3) [58], starch [59], nitrocellulose (NC) [60], conjugated polystyrene (CPR1) [61], etc.) and organic composite materials (P3HT:OD [62], PMMA:P3HT [63], PMMA:PCBM [64,65], PI:PCBM [66], PS:PCBM [67], PAA:PEI [68], Rhodamine B:Rhodamine 6G (Rh B:R 6G) [69], etc.). The performance comparison is shown in Table 2:

Table 2. Comparison of properties of organic dielectric materials.

| Organic Materials        | ON/OFF Ratio | Durability/Times | Retention Time | Number of Bending Cycles | Bending Radius |
|--------------------------|--------------|------------------|----------------|--------------------------|----------------|
| chitosan [53]            | $10^2$       | $10^2$           | $10^4$ s       | $10^3$                   | 5 mm           |
| lignin [54]              | $10^2$       | $10^2$           | $10^3$ s       | $10^2$                   | 15 mm          |
| poly(paraxylene [55]     | $10^4$       | $5 \times 10^2$  | $2 \times 10^3$ s | $5 \times 10^2$         | 10 mm          |
| PI [56]                  | $10^6$       | $10^3$           | $4 \times 10^3$ s | $10^3$                   | 5 mm           |
| pEGDMA [57]              | $10^2$       | $5 \times 10^2$  | $10^6$ s       | $10^3$                   | 4 mm           |
| pV3D3 [58]               | $10^5$       | $10^5$           | $10^5$ s       | $10^3$                   | 3.8 mm         |
| starch [59]              | $10^3$       | $10^2$           | $10^4$ s       | $10^3$                   | 5 mm           |
| NC [60]                  | $10^3$       | $10^2$           | $10^4$ s       | $5 \times 10^2$         | 10 mm          |
| CPR1 [61]                | $10^6$       | $10^3$           | $3 \times 10^7$ s | $10^3$                   | 3 mm           |
| P3HT:OD [62]             | $5 \times 10^2$ | $10^2$       | $10^4$ s       | N/R                      | 25 mm          |
| PMMA:P3HT [63]           | $10^2$       | 80               | $8 \times 10^3$ s | N/R                      | 10 mm          |
| PMMA:PCBM [64,65]        | $10^3$       | $10^5$           | $1.2 \times 10^4$ s | $10^4$                   | 10 mm          |
| PI:PCBM [66]             | $10^4$       | $2 \times 10^2$  | $10^4$ s       | $10^2$                   | 10 mm          |
| PS:PCBM [67]             | $10^4$       | 43               | $10^3$ s       | N/R                      | 4 mm           |
| PAA:PEI [68]             | $10^3$       | $2 \times 10^4$  | $10^4$ s       | $10^4$                   | 2 mm           |
| Rh B:R 6G [69]           | $10^3$       | WORM             | $5 \times 10^3$ s | $10^3$                   | 8.8 mm         |

Durability is measured by the number of switching cycles the device undergoes under DC conditions; WORM refers to a device of write-once-read-many-times; P3HT: poly real (3-hexylthiophene-2,5-diyl); OD: orange dye; PMMA: poly (methyl methacrylate); PCBM: 6,6-phenyl-C65-butyric acid methyl ester; PS: polystyrene; PAA: poly (acrylic acid); PEI: polyethyleneimine; N/R: not reported.

Figure 5. Nano porous (NP) structure of WO$_3$: (a) the current-voltage characteristics; (b) change of ON/OFF ratio after multiple bending cycles [54].
Compared with flexible inorganic materials, flexible organic materials show a little weakness in electrical properties, but better flexibility. An Al/CPR1/ITO device prepared by Zhou [61] has higher flexibility due to the hydrogen bond of CPR1. The favorable solubility and intrinsic flexibility of CPR1 allow for the large-scale fabrication of flexible CPR1 RRAM device arrays by full-printing technology with an endurance of $10^3$ bending cycles at a minimum bending radius of 3 mm and a high ON/OFF ratio of $10^8$. The Cu/pEGDMA/ITO/PET device prepared by Lee [57] has a typical I-V characteristic curve (Figure 6a—1). Its retention time exceeds $10^6$ s and its ON/OFF ratio reaches $10^2$ (Figure 6a—2). It also maintains good storage performance at the bending radius of 4 mm (Figure 6a—3). On the other hand, pV3D4 was selected as the storage medium to prepare Cu/pV3D4/Al/PES flexible RRAM by Jang [58], which also has a typical I-V characteristic curve (Figure 6b—1), and its steady-state hold time exceeds $10^5$ s, the ON/OFF ratio is $10^7$ (Figure 6b—2) and the minimum bending radius is close to 3 mm (Figure 6b—3). These two storage media materials show excellent performance in terms of durability, retention time and flexibility. According to the latest report, an ITO/PAA:PEI/ITO device with high flexibility and excellent storage performance was prepared by Ren [68] (Figure 7). An ON/OFF ratio of 50 can be maintained without degradation for up to $2 \times 10^4$ cycles (flat state) and over $4 \times 10^3$ cycles (bending to 2 mm radius $10^4$ times) in the direct-current sweep mode. Although organic materials have their unique advantages of flexibility, these organic media have some disadvantages, such as poor thermal stability, relatively low cycle times, poor fatigue resistance and incompatibility with CMOS, which limits the application and development of organic storage media.

**Figure 6.** Flexible RRAM based on pEGDMA (a) and pV3D3 (b): dielectric materials: 1—typical I-V curve (the inset is a real cross-section TEM image of the device); 2—cycle test and retention time of HRS and LRS; 3—relationship between resistance status (HRS/LRS) and bending radius [57,58].
3.1.3. Flexible Organic-Inorganic Composite Materials

Considering the excellent flexibility of organic materials and the electrical reliability of inorganic materials, using organic-inorganic composite materials seems to be a good choice. The resistance transition of inorganic dielectric materials is usually stable, but the flexibility is poor. Organic materials have good flexibility but poor stability of resistance transition. Therefore, organic-inorganic composite materials can be selected as storage media, in order to combine the stability of the storage performance of inorganic materials with the flexibility of organic materials. HKUST-1 (Cu$_3$(BTC)$_2$, BTC = benzo-1,3,5-tricarboxy acid), a composite material based on a metal-organic framework, was used by Pan [70]. This kind of material has the characteristics of high porosity, three-dimensional highly ordered structure, stable physical properties, easy design, easy film and so on. It is expected to realize flexible and stable resistance devices. In their study, they found that Au/HKUST-1/Au/PET devices can maintain uniform resistance over a temperature range of ±70 °C. Moreover, it has a minimum bending radius of 3.2 mm and an excellent durability of more than $10^7$ cycles (Figure 8). Metal-organic frameworks, which are formed by the association of metal cations or clusters of cations (“nodes”) with soft organic bridging ligands (“linkers”), are a fascinating class of flexible crystalline composite materials offering a potential strategy for the construction of flexible RRAM.

Moreover, poly (4-vinylphenol): graphene oxide (PVP:GO) and ultrathin HfO$_x$ were composited by Varun [71]. Under the scanning of AC and DC, the endurance can be over 1400 and 800 cycles, respectively. The same device configuration realized over a flexible PET substrate exhibited an ON/OFF ratio of >$10^3$ even after undergoing large mechanical strain (corresponding to a 5 mm bending radius). According to this report, the incorporation of GO into the PVP solution resulted in achieving better control over conductive filament growth and, therefore, improved repeatability and reliability.
The reported bending radii and bending cycles with different material systems: inorganic and organic. 

3.1.4. Further Discussion of the Material Systems

At present, there are other organic-inorganic composite materials (black phosphorous quantum dots: PMMA [72], graphene quantum dots: PVP [73], boron nitride: polyvinyl alcohol [74], copper phthalocyanine: PMMA [75], (CH$_3$NH$_3$)$_2$PbI$_2$(SCN)$_2$ [76], MnO$_2$/activated carbon fibers [77], MoS$_2$/zeolitic imidazolate frameworks [78], Al/CH$_3$NH$_3$PbI$_3$ [79], Fe/reduced graphene oxide [80], hybrid organic-inorganic perovskite [81], etc.) have excellent flexibility in organic materials and overcome the shortcomings of poor fatigue resistance in organic materials but also have good storage performance and stability in inorganic materials. However, their fabrication process is complex, and the resistance transition mechanism is not very clear.

3.2. Flexible Electrode Materials

As shown in Figure 9, through the comparison of several typical inorganic and organic materials, it can be found that the inherent brittleness of inorganic storage media makes the device only work under relatively low bending conditions, usually the bending radius is not less than 5 mm and the number of bending cycles is generally less than 1000 times. Compared with flexible inorganic materials, flexible organic materials are slightly inferior in electrical properties but exhibit better mechanical flexibility: higher bending cycles (1000 times) and smaller bending radii (<4 mm).
Therefore, researchers began to look for a suitable material system. Organic-inorganic composite materials have the excellent flexibility of organic materials and at the same time have the good storage performance and stability of inorganic materials, and thus they have been developed.

3.2. Flexible Electrode Materials

In the flexible RRAM device, flexible electrodes not only play the role of conducting currents but also play an important role in resistance transformation, which can directly participate in resistance transformation. Meanwhile, the electrode materials have crucial influence on the mechanical properties of the devices. Depending on their chemical composition, flexible electrode materials can be divided into metal electrodes, conductive oxide electrodes, carbon/nitrogen electrodes and other electrodes. So far, a large number of flexible electrode materials have been used to fabricate flexible RRAM, among which flexible metal electrodes are widely used at present because of their good electrical conductivity and great influence on resistance transformation.

3.2.1. Flexible Metal Electrode

Flexible metal electrodes include pure electrode (Pt [13,17,28,36,47,82–85], Au [32,36,38,54, 70,84–88], W [17,34,36,89,90], Ag [8,17,21,22,24,43,62,73,74,87,88,91,92], Al [18,24,27,44,45,55,58,79], Ni [23,63,93], Ti [47,94], Mg [53,90], Cu [41,58,94], etc.) and alloy electrode (Au/Ni [95], Pt/Ti [19], Ni/Ti [96], CdTeSb [25], GaInSn [97], etc.), with excellent electrical conductivity and mechanical properties, being easy to manufacture and easy to connect with the advantages of power. Using numerical simulations based on finite element analysis, Li [98] demonstrated that the defects, the cracks and the delamination of the electrode/active layer (storage medium) interface influences the formation of the conduction path (Figure 10). According to this report, the interface of metal/oxide/metal device has a high Schottky barrier to ensure the electrical stability generally. Zhang [99] has proved that the barrier height between the metal electrode and storage medium is an important factor affecting the resistance transition, and the higher the Schottky barrier is, the more stable the bipolar device is observed. On the other hand, the surface of the metal electrode provides ions to form conductive filaments by undergoing a redox reaction, which ensures the progress of resistance transition.

![Figure 10. Finite element analysis of poly(N-vinylcarbazole)-TiO2/Cu/PET sample under tension stress [98].](image)

3.2.2. Conductive Oxide Electrode and Carbon/Nitrogen Electrode

The metal electrode is widely used because of its good electrical conductivity, but its transmittance is relatively poor. Therefore, with the development of flexible transparent electronic technology, conductive oxide electrodes (ZnO [100], indium zinc oxide (IZO) [26,101], ITO [10,18,21,22,27,44,54,68,69,74,76,92,102,103], GO [30,39,103–105], etc.) are gradually increasing because of their high transmittance. Using ITO as the top and
bottom, the flexible RRAM device reported by Ren [68], shows an excellent transmittance of more than 85% in the whole visible region (Figure 7c). The carbon/nitrogen electrodes are characterized by high flexibility, including TaN [34], TiN [17,40,102], carbon powder [106], carbon nanotubes (CNTs) [107,108] and so on. The carbide electrode and nitride electrode have similar characteristics (high conductivity, good stability, excellent plasticity, good mechanical properties and hydrophilicity). The resistance transition mechanism of these electrodes is based on the formation and fracture of conductive filaments in the dielectric layer due to carbon or nitrogen vacancies.

Compared with metal electrodes, the conductivity of conductive oxide electrodes and carbon/nitrogen electrodes is still not ideal. Under long hours of work, the calorific value is much bigger than the metal electrode, causing serious device heating and affecting the stability of the device working. Thus, it is hard to meet the development trend of low power consumption, and they are not widely used.

3.2.3. Other Flexible Electrode Materials

Other flexible electrode materials include silver nanowire (AgNW) [91,105,109], ZnS [110, 111], poly(3,4-ethylenedioxythiophene):polystyrene sulfonate (PEDOT:PSS) [15,109,112], Si [13,43,113] and so on. According to the report of Jang [105], the AgNW electrode exhibits a light transmittance of more than 85% at a 550 nm light source, as well as excellent flexibility and stability (after bending $10^4$ times at a 4 mm radius, the resistivity changes only 0.2%). ITO is widely used due to its low resistivity and high light transmittance. However, ITO has many problems such as high raw material price, high manufacturing cost and high technological requirements (it easily deteriorates when exposed to plasma in the process of treatment, resulting in characteristic changes). Therefore, AgNW becomes an excellent alternative material for ITO. The AgNW’s grid layer exhibits high optical transparency, excellent flexibility and conductivity, and has the advantages of mass production by the solution process, which is suitable for transparent electrode.

Using ZnS as a flexible electrode, it was proved that multishell ZnS can provide quantum dot sites according to the report by Kim [111]. When the device is in HRS, all traps in the quantum dots are fully filled, enhancing the charge storage capability of the device and making the formation of conductive filaments in the device more rapid and stable. Meanwhile, the ZnS electrode also has the characteristics of transparency and flexibility, and its transmittance in the visible light band is more than 70%. After bending at a 10 mm radius 100 times, there was no significant change in conductivity.

PEDOT:PSS is one of the common polymers with excellent stretchability [109]. This polymer has the characteristics of a simple fabrication process at room temperature, low cost, good flexibility and strong compatibility with flexible substrates, but it is not resistant to high temperature, which is a common shortcoming of organic materials. It is often used together with AgNW as an electrode of the device. As shown in Figure 11a, AgNWs were placed randomly with each other. Due to having overlapping parts of AgNWs, the contact resistance between AgNW layers was very high, resulting in an increase in the resistivity of AgNW layers. The stacking of PEDOT:PSS layers on the AgNW layer can improve the contact between them (Figure 11b). The PEDOT:PSS layer was deposited on AgNW film by spin coating, which not only helped to improve the conductivity of AgNW but also improved the surface uniformity of AgNW electrode and reduced its roughness (Figure 11c,d). Meanwhile, the maximum transmittance of the PEDOT:PSS/AgNW composite layer is 89% (the highest transmittance for electrode). As for Si electrodes, p-doped and n-doped monocrystalline silicon are generally used as the top and bottom electrodes, respectively. Such electrodes can be used to prepare flexible memory with self-rectification resistance transformation characteristics. The self-rectifying effect is due to the different doping types in the silicon disk that forms a self-sufficiency diode within each junction.
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In addition, the effect of the flexible electrodes on the mechanical properties of the device was studied by Shang [19]. The devices of ITO/HfO$_x$/ITO and ITO/HfO$_x$/Pt were prepared separately. The result shows that the minimum bending radius of the device is 3 mm when the top and bottom electrodes are ITO electrodes, while the minimum bending radius of the device can be reduced to 2 mm when the top electrode is ITO and the bottom electrode is Pt. The mechanical properties of the device can be improved by using Pt as the electrode, mainly because there is a great difference between the crack formed by Pt fracture and the crack formed by ITO fracture when the curvature exceeds the critical value. The ITO electrode will form a crack through the whole film, which makes it lose the ability to conduct current. Pt electrodes, on the other hand, form a maze of cracks, which affects the conductivity, but still retain the ability to conduct currents. It can be seen that the mechanical properties of electrode materials are one of the keys to affecting the flexibility of the device.

3.3. Flexible Substrate Materials

In the practical use of flexible RRAM, devices will inevitably undergo frequent deformations such as bending, twisting and folding, which makes them vulnerable to mechanical damage. Therefore, the choice of flexible substrate material is very important.

Flexible substrates shall have the following characteristics: (1) Mechanical properties: Strong mechanical flexibility is the most essential requirement of flexible substrates to comply with deformable transformation under severe stress and strain, in addition to the efficient stress-release property while maintaining the original functionality. Moreover, the substrate is the carrier of flexible RRAM memory cells and plays the role of mechanical support. Therefore, the substrate is required to have good dimensional stability, that is, the ability of the material to maintain its original size under different processing conditions and uses. (2) Chemical stability: In the process of device preparation and use, the substrate will
be exposed to a variety of chemical atmospheres, so the substrate must have high chemical inertness to ensure the normal production and normal operation of the device. (3) Barrier characteristics: Although the electrode materials and dielectric materials composed of the device have good electrical properties, they are often susceptible to the corrosion of oxygen, moisture and other atmospheres, resulting in the degradation of the functional layer and device performance. Therefore, the substrate material is needed as a protective layer to prevent or delay the corrosion of the functional layer. (4) Heat resistance: Including thermal transition temperature and coefficient of thermal expansion. The maximum temperature should not exceed the temperature of glass transition (T_g) during work and processing. The difference in the coefficient of thermal expansion between adjacent layers in a multilayer device will exert additional stress on the lattice, resulting in the strain and cracking of the device during heating or cooling. (5) Surface uniformity: A good substrate should have a smooth surface. The electrode is directly deposited or grown on the substrate surface, so the excessive roughness of the substrate surface will lead to uneven electrode thickness, which may lead to short circuits and other problems. In addition, issues such as machinability, cost and weight should also be considered.

Flexible substrate materials can be roughly divided into polymer materials, metal materials and other materials based on their physical properties. In the development of flexible electronic devices, polymer substrates have always been the primary material of choice because they have excellent mechanical properties, stable chemical properties and good processability.

### 3.3.1. Flexible Polymer Substrate

Polymer materials are currently the most commonly and widely used flexible substrates, including polyimide (PI) [23,24,37,42,49,56,66,82,91,114], polyethylene naphthalenediace (PEN) [15,30,36,102,111,114,115], polyethylene terephthalate (PET) [8,10,18,19,22,25–27,40,45,51–54,59,68,70,71,73–76,86–88,90,93,95,101,104,105,109,110,112,114], polycarbonate (PC) [116], polyethersulfone (PES) [58,117] and so on. The comparison of their mechanical properties is shown in Table 3:

| Features                              | PET [118–120] | PEN [121–124] | PI [120,125] | PC [126–128] | PES [129,130] |
|---------------------------------------|---------------|---------------|--------------|--------------|---------------|
| tensile strength (flexibility, MPa)   | 742           | 708           | 144          | 60           | 44            |
| Young’s modulus (elasticity, GPa)     | 2.8           | 9.0           | 2.5          | 2.5          | 0.64          |
| temperature of glass transition (°C)  | 84            | 129           | 320          | 150          | 220           |
| melting temperature (°C)              | 255–265       | 260           | 590          | 220–230      | 330           |
| thermal conductivity (W/m-K)          | 0.3           | 0.164         | 0.8          | 0.2          | 0.17          |
| coefficient of thermal expansion (ppm/°C) | 60           | 18.45         | 15           | 50           | 65            |
| transmittance                         | 90.4%         | 88.18%        | 60%          | 90%          | 89%           |
| corrosion resistance                  | good          | good          | good         | poor         | good          |
| dimensional stability                 | good          | good          | fair         | fair         | good          |

These polymer materials have good mechanical flexibility, which can withstand repeated bending and twisting, and they also have stable physical and chemical properties. PEN (Figure 12a) and PET (Figure 12b) are currently the most popular polymer substrates in the field of flexible electronic devices, which possess great flexibility and chemical stability. However, their relatively low temperature of glass transition and melting temperatures limit them from producing and operating. In addition, they have high water vapor transmittance (1.1 g/m² per day at 45 °C for 100 μm thickness [131]), resulting in the role of protecting the functional layer being small, which may lead to the unstable performance of the functional layer. Therefore, if the device requires long-term stability, it is necessary to carry out appropriate packaging, which involves the packaging process and cost. PI (Figure 12c) has a relatively high temperature of glass transition and melting temperature, but its low transmittance and high cost make it difficult to meet the application require-
ments of low cost and high transparency in industrialization. PC has a high transmittance but also has flame retardant and oxidation resistance, which can play a very protective role on the functional layer. The main defect of PC is low hydrolysis resistance, easy to be corroded by organic chemical solvents or atmosphere, resulting in performance decline or material degeneration. PES has high transparency, but it has problems of poor flexibility and low operating temperature. Moreover, PES has a high coefficient of thermal expansion, which may cause material strain due to mismatching with the thermal properties of electrode materials during cooling or heating up. In a word, the unique advantage of polymer materials is excellent flexibility, but the disadvantage is low operating temperature.

| Properties                  | PET [118,5,91] | Glass [65,90] | SiO₂ [65,90] | Stainless Steel [132,133] | Tungsten Foil [89] | Ni-Cr Foil [83] | Copper Foil [115] | Polymer [69,92] |
|-----------------------------|----------------|---------------|-------------|---------------------------|--------------------|-----------------|------------------|----------------|
| Tensile Strength (MPa)      | 742            | 688           | 129         | 150                       | 220                | 18.45           | 75               | 60             |
| Coefficient of Thermal Expansion | 0.3           | 0.164         | 0.8         | 0.2                       | 0.17               | 50              | 65               | 50             |
| Melting Temperature (°C)    | 255~265        | 590           | 330         | 220~230                   | 600                | 129             | 320              | 220            |
| Corrosion Resistance        | Good           | Good          | Good        | Poor                      | Good               | Good            | Good             | Good           |
| Ozone Degradation (ppm/°C)  |                |               |             |                           |                    |                 |                  |                |

Table 3: Mechanical properties of flexible RRAM polymer substrates.

![Figure 12](image.png)

Figure 12. Structural formula of common polymer substrates: (a) PEN; (b) PET; (c) PI.

3.3.2. Flexible Metal Substrates and Other Flexible Substrates

Metal materials used as flexible substrates include titanium foil [115], copper foil [94], tungsten foils [89], Ni-Cr foil [83], stainless steel foil [132,133] and so on. Polymer materials generally do not withstand high temperatures, so metal foil is a good substitute for polymer substrates because of their high-temperature resistance and chemical stability. However, metal materials are less flexible than polymer materials and almost opaque. Meanwhile, they will affect the work of the functional layer of the device because of their conductivity. Moreover, the density of metal materials is generally high, so the weight is heavy, which is not conducive to the development of lightweight devices.

Other flexible substrates include flexible mica [84,85,100,134], flexible glass [135], SiO₂ [65,90] and so on. Flexible mica is a natural transparent crystalline material with high flexibility due to the layered framework structure of aluminosilicate. It also has a low cost, light weight, high transmittance (>90%), high-temperature resistance (~600 °C) and low coefficient of thermal expansion (10⁻⁶ per centigrade). According to the report by Ke [134], the performance of ITO electrode depositing on flexible mica has no obvious change after being bent 10⁵ times. However, the compatibility between flexible mica and metal electrodes is poor due to its unique structure. For flexible RRAM, flexible metal materials are the most commonly used electrode materials, so flexible mica is not widely used as substrate material. Flexible glass and SiO₂ were used only as substrate material under special circumstances. In the vast majority of cases, the substrate material obviously has better choices. All in all, flexible substrates, as the carriers of the flexible RRAM memory, need to be considered comprehensively and chosen carefully.

3.4. Summary of the Material System

In flexible dielectric materials, the inorganic dielectric has good electrical properties and stable storage properties. The organic dielectric has good flexibility and can basically withstand more than 10⁵ bending cycles under the minimum bending radius. The organic-inorganic composite material not only has excellent electrical properties and stable storage properties but also has good flexibility. In terms of material structures, there are nano porous structures, amorphous nanostructures and the metal-organic framework to be used. As mentioned above, the electrical properties and flexibility of dielectric materials can be significantly improved by using these structures.

In the flexible electrode materials, metal electrodes are the most used flexible electrode materials. Metal electrodes have excellent conductivity and good flexibility, conductive oxide electrodes have high transmittance and carbon/nitrogen electrodes have excellent flexibility. In addition, the grid layer structure of metal nanowires can only improve
the light transmittance but also improve the electrical conductivity and flexibility of the metal electrode. On the other hand, the shape of the crack formed by the electrode material under multiple bending cycles will also affect the function of the electrode material. For example, the labyrinth crack formed by the Pt electrode can still conduct currents although its conductivity will be reduced.

In the flexible substrate materials, polymer substrates have been the first choice among flexible substrates, with PET and PEN being the most popular. These polymer materials have good mechanical flexibility, which can withstand repeated bending and twisting, and they also have stable physical and chemical properties. Although metal substrates and other types of substrates are used, polymer substrates are clearly the best choice in non-special cases.

3.5. Flexibility of RRAM

In flexible thin film transistors (TFT), the neutral plane concept or island structure concept are well developed in order to realize a flexible device.

Likewise, the flexibility and elasticity of RRAM has attracted increasing attention from the scientific community. An in-depth understanding of the mechanical and electrical failure mechanisms of RRAM is of great significance for realizing its flexible integration. Organic elastic polymers are used as resistive switching media, which have poor resistance to the rotation of molecular chains and environmental water and oxygen under stretching, and the electrical stability of the prepared resistive memory under strain is generally poor. Li et al. [61] further proposed to use organic-inorganic hybrid metal-organic framework (MOF) materials as resistive switching media to construct RRAM devices to simultaneously improve the storage performance, elastic mechanical properties and thermal stability of flexible RRAM devices. MOF materials are organic-inorganic hybrid crystal framework materials constructed by organic ligands and metal ions or clusters through coordination bonds. Through the rational selection of organic and inorganic constituent units, MOF materials can achieve material deformation (breathing effect) through the change in the coordination bond angle and the regular change in the framework structure. This characteristic provides more space for the flexible elasticization of a resistive medium. Zhou et al. [97] have made new progress in the field of high-performance flexible resistive memory. The CPR1-based RRAM device has a simple sandwich structure and has excellent non-volatile memory performance: it has a very high switching ratio ($10^8$), a super fast 29 ns fast response, good reliability and reproducibility and long-term stability. Finally, the flexible RRAM device array fabricated by the all-printing technique exhibits high RRAM performance and robustness.

Compared with flexible TFT, the simple sandwich structure of RRAM is usually not the main factor affecting its flexibility. The currently reported work on flexible RRAM mainly considers the material structure and process to achieve good device flexibility.

4. Fabrication Process of Flexible RRAM

The typical steps in making flexible RRAM are shown in Figure 13:

As a typical film electronic device, the fabrication of flexible RRAM mainly relies on various film deposition methods. Generally speaking, the fabrication process of flexible RRAM is divided into dry methods (DMs) and wet methods (WMs) [136]. DMs are mainly vacuum evaporation, including chemical vapor deposition (CVD) [137,138], physical vapor deposition (PVD) [22,139] and molecular beam epitaxy (MBE) [140,141]. These methods are less affected by impurities because they are operated in a vacuum environment. In the process of industrialization, the flexible film can be improved by changing the gas flow rate, gas pressure, ambient temperature and operating voltage. However, these methods have high costs, long synthesis times and difficulty in mass production; hence the emergence of WMs. WMs include coatings (spin coatings [142], spraying [143] and casting [144]), printing (screen printing [145], inkjet printing [15,146] and nano imprinting [147]) and immersion (dipping method [142], Langmuir–Blodgett (LB) method [148] and self-assembly [88]).
WMs are inexpensive and easy to manufacture in large areas, but these methods often introduce impurities, resulting in less uniform films and less flexibility. On the other hand, they are difficult to graph, making them difficult for operations that require high precision and resolution. Table 4 below summarizes the advantages and disadvantages of each method:

| Methods          | Advantages                                                                 | Disadvantages                                                                 |
|------------------|-----------------------------------------------------------------------------|-------------------------------------------------------------------------------|
| CVD [137,138]    | Pure film and good density and uniformity                                   | High temperature of reaction, difficulty in mass production and toxic byproducts |
| PVD [22,139]     | Low temperature of reaction and high degree of process automation          | Poor uniformity and difficulty in mass production                             |
| MBE [140,141]    | Pure film, grows at low temperatures and has atomic interfaces             | High cost and difficulty in mass production                                   |
| spin coatings [142] | Uniform film thickness                                                     | Mass solution loss                                                             |
| spraying [143]   | No solution loss and quick film forming                                     | Difficulties in process optimization of solvent mixing and solution adjustment |
| casting [144]    | High degree of crystallinity                                               | Poor uniformity and long molding cycle                                         |
| screen printing [145] | Simple manufacturing process and wide range of application                  | The large thickness is not conducive to miniaturization                        |
| inkjet printing [15,146] | Automatic control, small material loss, rapid production of multilayer structure and no selectivity for substrates | High requirements for ink conditions and droplets coalesce easily              |
| nano imprinting [147] | Composition of tens of nanometers can be achieved under normal temperature and pressure | Immature technology and expensive equipment                                    |
| dipping [142]    | Large area of film forming                                                 | Large thickness relatively and inability to perform fine manipulation         |
| LB [148]         | Single-layer controlled production and nanoscale accuracy                  | Corresponding materials are limited and poor mechanical properties           |
| self-assembly [88] | Highly oriented monolayer and quick film forming                          | High requirements of process and strict conditions of preparation             |

Figure 13. Typical steps in making flexible RRAM.

Table 4. Advantages and disadvantages of various film deposition methods.
In recent years, some new fabrication methods have been developed, such as atomic layer deposition (ALD) [9,17,149,150], pulsed laser deposition (PLD) [151,152], layer-by-layer (LBL) [108,153], the sol-gel process (SGP) [21,85,92] and drop-on-demand (DOD) [154]. ALD can be used for the deposition of metal, dielectric, polymers, etc. The uniformity and density of the film are very good with this method, and the films have also excellent flexibility, but it needs high temperature to deposit film (up to 500 °C), so using this method needs to use high-temperature-resistant materials as substrate. In addition, the deposition directivity is very poor and the deposition rate is slow. PLD has a higher deposition rate and lower temperature for substrate, and it can accurately control stoichiometry. Its disadvantage lies in that, in the process of deposition, there is micrometer-scale particle pollution such as molten small particles or target fragments, which leads to a reduction in uniformity and flexibility. At the same time, this method is very difficult for the fabrication of large-area films. LBL assembly is one of the most widely studied WMs in recent years. Due to its simple process and high controllability, the process can be used to produce uniform and multifunctional films. Moon [108] prepared the flexible single-wall carbon nanotubes electrode by this method, and the relationship between the deposition layer and the electrode morphology, thickness, transparency and resistivity was investigated. SGP is a simple method for the deposition of high-quality pure metal oxide layers. By introducing two or three precursors, the structure of metal oxide products can be easily adjusted to improve their electrical properties and flexibility. However, this method can only be carried out at low temperature and the period of fabrication is very long. DOD also has considerable potential in the future of film manufacturing as a printing technology for contactless direct prototyping and rapid manufacturing.

In actual production, the different fabrication processes can be combined to obtain the best performance. On the other hand, the prepared films are usually subjected to subsequent optimization processes such as annealing [92] or pattern [155] to eliminate the defects of the film or meet special requirements, improving the performance of the device.

5. Conclusions and Challenges

To sum up, as the next generation of flexible RRAM with the most development potential, the research of flexible RRAM materials has made great progress. However, in order to meet the higher requirements of high flexibility, small size, high durability and low cost, flexible RRAM still faces some important challenges: Firstly, it is urgent to explore the resistance transition mechanism and failure mechanism of flexible RRAM and the three-dimensional configuration of devices (such as vertical cross array and stacked cross array), so as to further guide device performance optimization. Secondly, material cracks are the main cause of device failure under severe bending conditions, so it is urgent to explore materials with better flexibility (liquid materials are also a development direction). Finally, since the practical application environment of flexible devices is usually more complex than the bending conditions in the laboratory, the performance of flexible RRAM under torsion and tensile conditions needs to be emphasized and strengthened. In the research direction, the research on dielectric materials with both stable storage performance and good flexibility, the research on electrode materials with good conductivity, mechanical stability and flexibility and the research on the regulation of electrode/dielectric interface has achieved initial results, but there is still a lack of more in-depth research. Another question is whether the test of flexibility is too ordinary. The test in the laboratory has a very great deal of difference with the actual use, so a more effective testing method needs to be explored. If the above challenges can be well solved, it can be expected that flexible RRAM, as an important component of flexible electronic products, will bring great convenience to people’s lives and promote the further development of society.
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