In our lives, the acquisition, storage, search, sharing, and even visual presentation require huge amounts of data in storage and processing at any time. This propounds a strong demand for the stretchable and wearable memory devices with ultrafast switching, high density, and high energy efficiency. Herein, an overview of the advanced progresses made on developing stretchable resistive switching random-access memory (RRAM) is presented, concentrating on the fundamental components, materials selection principles, and configuration design strategies. The basic electrical and mechanical properties of stretchable components to meet the functionality of memory storage are summarized. Novel materials with the intrinsically stretchable features toward the realization of stretchable RRAM are discussed. Ingenious configuration design strategies to obtain stretchable RRAM are evaluated. Finally, the challenges and perspectives for the practical applications of stretchable RRAM are also addressed. Some helpful guidelines are provided in promoting the development on stretchable memory devices for making deployable and portable artificial intelligence and smart devices.

1. Introduction

In the 21st century, the information revolution and the development of digital information technology bring us to the era of big data.\(^\text{[1,2]}\) In our lives, the acquisition, storage, search, sharing, analysis, and even visual presentation that requires huge amount of data are in progress at any time.\(^\text{[3]}\) With the development and application of 5G technology and artificial intelligence technology, autonomous cars, aircraft, intelligent Internet of Things, and portable wearable devices will be of widespread use. These advanced electronics are required to have the ability to quickly store, read, and process a large amount of information at a high speed. This propounds a strong demand for memory devices with ultrafast switching, high density, and high energy efficiency.\(^\text{[4–18]}\) In general, two major categories of memory devices including the volatile and nonvolatile storages are required for the system operation.\(^\text{[11–13]}\) The volatile memory of complementary metal oxide semiconductor-based static/dynamic random-access memory (SRAM/DRAM) will lose stored information after power is cut off.\(^\text{[14–16]}\) The volatile memory has a high read/write speed but its scaling has encountered limitations. The nonvolatile memory such as ferroelectric, magnetoresistive, phase-change, and resistive switching random-access memory (RRAM) could store the data without power supply.\(^\text{[6,7,17–23]}\) Among these nonvolatile memories, the RRAM has attracted great interest for its simple sandwich structure and high operation speed, low power consumption, high-density data storage, and scalability.\(^\text{[24–35]}\)

Nowadays, wearable electronics including personal healthcare, sports performance monitoring, and even implantable medical equipments have been developed to meet consumers’ enthusiasm in pursuit of function, fashion, and avant-garde performances.\(^\text{[29–33]}\) To meet the conformable and compliant wearable applications, stretchable electronic devices with deformable and elastic mechanical properties are desirable for complex and dynamic mechanical environment applications. Correspondingly, as an essential data storage and data reading element of electronic equipment, the stretchable memory device is strongly expected to be explored and developed.\(^\text{[34–40]}\) Based on the features of the simple metal–insulator–metal structure and advantages of data storage as mentioned earlier, RRAM is considered to be an ideal choice for stretchable memory. Till date, a large number of studies has reported the patterning and crossbar-structured RRAM based on resistive switching mechanisms such as oxygen vacancies and conductive metal filaments mechanisms.\(^\text{[41–46]}\)

These RRAMs usually consist of the inert/active metal electrodes including Au, Pt, Ti, Cu, Al, and Ag and metal oxide insulators such as hafnium oxide (HfO\(_2\)),\(^\text{[47–49]}\) aluminum oxide (Al\(_2\)O\(_3\)),\(^\text{[50–52]}\) titanium oxide (TiO\(_2\)),\(^\text{[53,54]}\) magnesium oxide (MgO),\(^\text{[55,56]}\) zirconium oxide (ZrO\(_2\)) et al.\(^\text{[57,58]}\) Other nonvolatile RRAM materials include 2D-layered insulting materials like hexagonal boron nitrides and molybdenum disulfide.\(^\text{[59–62]}\) But these materials encountered challenges in realizing the highly stretchable feature of RRAM due to its limitation of the physical processability and mechanical properties constraints.
As shown in Figure 1, progresses on stretchable RRAMs were achieved recently using the intrinsically stretchable materials including the donor–acceptor block copolymers, elastic Ag nanoparticle-doped thermoplastic polyurethanes, or through structural engineering such as wrinkled, mechanical hybrid (the rigid island in a soft matrix), and serpentine-structured configurations. In addition, the stretchable field-effect transistor memory was also fabricated with the similar materials selection principle and configuration design strategy. These significant progresses provide researchers the inspiration on promising routes to attain stretchable RRAM. There is a large diversity of material selection and the flexibility of structure configurations that can be used in the research of stretchable RRAM. Therefore, this Review on stretchable RRAM is aimed at summarizing the research progress to provide a timely overview and comprehensive understanding in this field.

This Review provides an overview of the advanced progresses made on the stretchable RRAM, concentrating on the fundamental components, materials selection principles, and configuration design strategies. Particularly, the basic electrical and mechanical properties of stretchable components including the conductor, insulator/intermedia, and substrate for RRAM were summarized and reviewed. Novel materials with intrinsically stretchable features toward the realization and performance improvement of stretchable RRAM are discussed. Ingenious configuration design strategies with conventional materials to obtain the stretchable RRAM are discussed and evaluated. Finally, the challenges and perspectives are discussed. This Review is expected to provide both the summary of progress in stretchable memory and some helpful guidelines in promoting the development of stretchable RRAM.

2. Stretchable Components for RRAM

The RRAM devices are composed of an insulating layer sandwiched between two electrodes. To realize stretchable RRAM devices, the main challenge is to obtain the stretchable components which can work under mechanical strains. In this section, we conduct a review of the stretchable components including the stretchable conductors, insulating layer, and substrate that can be used to fabricate the RRAM.

2.1. Conductors for Stretchable RRAMs

We categorized the conductors for stretchable RRAM into two types. One is used as the electrode on the individual memory cell. The other is used as the electrical interconnect between...
the memory cells. Based on their functionalities, different requirements may be imposed on the two conductors. For instance, RRAM based on metallic filament formation may require metallic materials such as Ag or Au for the electrodes to provide the metal ions under an electric field. On the contrary, the electrical conductivity and the critical strain at which the conductivity is lost will become the two major criteria for the stretchable electric interconnect. A few comprehensive reviews on elastic conductors can be found in recent publications.[73–78] Herein, we specifically focus on discussing the conducting materials for stretchable RRAMs. Table 1 shows the representative samples of these conducting materials. We begin this article by reviewing and summarizing the general strategies which have been developed to realize conductors for stretchable RRAMs. Similar approaches may apply to create stretchable structures for the other layers in the RRAM devices that are described in the later sections.

2.1.1. Electrode for Individual Memory Cell

The RRAM cell can be fabricated with relative small dimensions with the localized resistive switching phenomenon (the dimensions can be reduced to the nanometer range[84]), making it feasible to be isolated as a mechanically independent island in the stretchable devices. With a rational device structure design, the strain induced on the small island can be effectively minimized under mechanical deformations. For instance, by structuring the elastic substrate with raised islands, strain isolation and strain limitation behavior can be achieved in the electronic device arrays (Figure 2A).[85] In this case, the requirement on the mechanical conformability of the conductors is not stringent, which opens up a large range of materials selection for the electrodes. Conventional electrode materials, such as Au, silver, and aluminum thin films, can be successfully applied to fabricate the stretchable RRAMs.[97,63,67] Apart from the conventional rigid metallic films, other elastic conductors were also explored as the electrodes for the memory cells. Embedding conductive fillers in the elastic polymer matrix is a widely applied approach to realize the stretchability of the electrodes.[96–97] Considering that a smooth surface of the electrodes is required for the subsequent thin intermedia layer deposition in the stretchable RRAMs, an effective approach to prepare the stretchable electrodes is by preparing the conductive networks on the smooth surface of a sacrificial substrate, embedding the conductive networks with elastic polymer matrix and peeling off the substrate to complete the fabrication process,[99] as schematically shown in Figure 2B.[99]

2.1.2. Electrical Interconnect Between the Memory Cells

While many studies only report stretchable RRAM with electrically isolated memory cells, it is inevitable to form an electrical interconnect between the memory cells for practical applications, such as the wordline or bitline. The choice and scaling of wordline/bitline certainly affect the write/read performance, energy dissipation, and reliability of the cross-point memory arrays. Compared with the electrode materials, the conductors for electrical interconnect in the stretchable RRAMs are required to possess high conductivity, stretchability, mechanical stability, and even effective wire energy dissipation to reduce latency. The structural approach and material approach are two major strategies used to realize highly stretchable conductors.[73,102]

With the structural approach, conventional metallic materials with a high conductivity can be directly used. As shown in Figure 2C, the serpentine structure can alter to relax the stretching strains under mechanical deformations so that the thin-film metallic films can accommodate the minimal mechanical strains.[80] The photolithograph process is generally required to pattern the serpentine structures in the electrical interconnects, which will involve multiple fabrication steps of patterning, transferring, and bonding processes. Alternatively, the buckling or wrinkling structures can also be applied to achieve the stretchable structures (Figure 2D).[100] The buckling or wrinkling structures can be created by adhering the materials with a thin layer onto a prestretched substrate. After relaxation, the thin conducting layer will buckle to accommodate the compressing strain, and the layer can be reversibly stretched to a maximum strain not larger than the prestretched value. To use the approach, it is important that the conducting layer is very thin to adhere well to the underneath elastomeric substrate. In addition, the thin profile of the conducting layer can effectively reduce the strain in the buckling films as the bending strains decrease linearly with thickness.[103] This approach has been successfully applied to the full device structures, such as stretchable light-emitting devices,[104] organic solar cells,[100] and field-effect transistors.[105]

And it will also be an optional approach that can be applied to realize stretchable RRAMs. Nevertheless, there may be increasing concerns on thin profile wires carrying high current densities that may eventually lead to reliability issues especially with the deep scaling of memory feature size. This is exacerbated by the bending and curving of the buckling structures which inevitably lead to current crowding or localized joule heating that impacts electromigration or stress-voids formation.

As a different strategy, the material engineering method focuses on creating intrinsically stretchable conductors with the promising merits of large-area, cost-effective, and simple fabrication processes. Printable conducting composites with conductive fillers for electron transport and elastic polymer matrix for mechanical conformability have been intensively studied.[30,87,88,90,92,101,106,107] For instance, Someya’s group

| Materials                          | Conductivity at 0% strain | Maximum strains |
|-----------------------------------|---------------------------|-----------------|
| Au thin film on prestretch PDMS[79] | $5 \times 10^5 \text{ S m}^{-1}$ | 10%            |
| Serpentine Cu or Au thin films on silicone substrate[80] | – | 300%          |
| Silver flakes and EGaIn in elastic polymer[81] | $8.31 \times 10^5 \text{ S m}^{-1}$ | 1000%        |
| EGaIn filled in PDMS channels[82] | $\approx 2.4 \times 10^5 \text{ S m}^{-1}$ | $\approx 220\%$ |
| PEDOT:PSS modified with bis(trifluoromethane) sulfonamide lithium salt[83] | $3.10 \times 10^5 \text{ S m}^{-1}$ | 800%          |

Table 1. Conductivity and maximum strains of conductors for stretchable RRAMs.
Figure 2. A) Scanning electron microscopy images of the elastic substrate with structural design for strain isolation on the device islands. Reproduced with permission.[85] Copyright 2012, Wiley-VCH. B) A schematic showing the fabrication process to embed the conductive networks into elastic polymeric matrix. Reproduced with permission.[99] Copyright 2014, The Royal Society. C) Optical image of the stretchable electrode with the serpentine structure and the corresponding finite element analysis (FEA) under strain. Reproduced with permission.[80] Copyright 2013, Springer Nature. D) A schematic image showing the structure of the ultrathin organic solar cell device and the photographs of the buckling device structure after attaching onto the elastic substrate. Reproduced with permission.[100] Copyright 2012, Springer Nature. E) A schematic showing the working mechanism of the elastic conductor with liquid metal particles and the performance of the elastic conductor under extreme strains. Reproduced with permission.[101] Copyright 2018, Wiley-VCH. F) A stretchable freestanding PEDOT:PSS thin film and its electrical performance under strain. The schematic diagram below shows the morphology of the stretchable PEDOT:PSS. Reproduced with permission.[83] Copyright 2017, AAAS.
and our group have demonstrated printable elastic conductors by embedding different conductive fillers in elastic polymers to enable high stretchable conductors with good conductivity and high stretchability. Silver flakes, AgNWs, SWCNTs, graphene, metal meshes, etc. have been reported as promising conductive fillers for elastic conductors. The trade-off between conductivity and stretchability is a grand challenge for printable elastic conductors because while more conductive fillers will improve the conductivity in the composites, the rigid conductive fillers will lead to degraded stretchability in the elastic polymer matrix. Our group recently developed an effective approach to significantly increase the stretchability and mechanical stability of the printable elastic conductors using liquid metal particles to bridge the electrical connections between the conductive fillers under mechanical deformations. The elastic conductor has a high conductivity of $\approx 8000 \, \text{S cm}^{-1}$ and extreme stretchability up to 1000%, as shown in Figure 2E. 

Intrinsically stretchable materials such as liquid metals and conductive polymers are also interesting candidates to realize the stretchable electrical interconnect for RRAMs. Eutectic gallium indium alloy (EGaIn) is attracting increasing interest for soft electronic research with its high conductivity, low toxicity, and low melting point. They can be encapsulated into an elastic channel or embedded into polymer matrix to create highly conductive and stretchable pathways. Detailed summary on the application of liquid metal for electronic devices can be found in the recent review articles. Poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) is a widely exploited conductive polymer for organic electronics. Though PEDOT:PSS offers a small stretchability (≈5% strain) which makes it a favorable choice for the electrode materials in flexible electronics, however, it fails for stretchable devices which required mechanical strains above 10%. Efforts to include different additions into the PEDOT:PSS have been reported, such as Zonyl and Triton X-100. Bao’s group recently reported an inspiring approach to use ionic liquid to simultaneously increase the conductivity and stretchability of the PEDOT:PSS film, as shown in Figure 2F. The elastic PEDOT:PSS provided the conductivity of 4100 $\, \text{S cm}^{-1}$ and the maximum strain of 800%. The modified PEDOT:PSS was prepared in aqueous solution and could be patterned with an inkjet printing approach to create designed structures, making them a promising candidate for electrical interconnects in the electronic devices.

Conductive fibers have been attracting great interest for wearable electronic textiles and stretchable conductors. They can form interconnected percolating networks to provide excellent mechanical conformability for stretchable conductors or be woven into varied types of fabrics for wearable electronics. To realize conductive fibers, a highly effective approach is to coat conductive films onto the based fibers with cotton, silk, or polymers. For instance, by coating metallic films including Cu, Ag, Al, and Au with a standard thermal evaporation process onto the electrospinning nanofibers, stretchable and transparent conductors could be achieved on the elastic substrates. With a similar approach, Miyamoto et al. also demonstrated highly stretchable conducting fibers based on Au-coated nanofibers for e-textile applications. The thin percolating conductive fibers possess the additional advantage of good gas permeability, they can be directly applied onto the human skin for wearable electronics with minimal invasiveness. Alternatively, embedding conductive fillers into the polymer matrix during the fibers fabrication process was also proven to be successful. As an example, highly stretchable conductive fibers can be fabricated using a facile wet spinning method with AgNWs-dispersed poly(styrene-block-butadiene-block-styrene) (SBS) solution. The conductive fibers can maintain their conductivity up to the strain of 220%. With the inherent stretchable structures and cost-effective fabrication approach, conductive fibers will be emerging materials to be explored for stretchable and wearable RRAMs.

### 2.2. Insulator/Intermedia Layers for Stretchable RRAMs

Following the realization of conductors for stretchable RRAMs, similar strategies can be applied for the memory insulator/intermedia active layers. The insulator/intermedia layers can be classified as layers with stretchable structures and layers with stretchable materials.

#### 2.2.1. Insulator/Intermedia Layers with Stretchable Structures

Using structural designs to isolate memory cells from strain under mechanical deformations, rigid insulating layers, such as TiO$_2$ nanomembrane and IZO nanomembrane, can be directly exploited. Together with the rigid thin metallic electrode, the individual memory cell was fabricated for the stretchable RRAM arrays. In the stretchable RRAMs, the localized memory cell encounters a very small strain under stretching. As a result, choices for the insulating layers can be extended to many conventional dielectric materials such as Al$_2$O$_3$, HfO$_2$, WO$_x$, SiO$_2$, FeO$_x$, etc.

#### 2.2.2. Insulator/Intermedia Layers with Stretchable Materials

Composite materials with embedded functional materials in the polymer matrix (Figure 3A) are also exploited to fabricate the intermedia layer for stretchable RRAMs. Different from the composite materials for stretchable conductors, which typically focus on establishing stable percolation between the conductive fillers for charge transport, the composites for the stretchable intermedia layers consist of separated functional materials in the elastic polymers. The operation of stretchable RRAMs requires the intermedia layer to possess the capability to alternate its resistance under an electric field. Different resistance switching mechanisms have been utilized in the insulating layers, including phase change, reduction and oxidation of metal ions, and the formation of the conductive filament by high density defects. Based on the metal ions reduction and oxidation mechanisms, different intermedia composites for the stretchable RRAMs can be developed. For instance, stretchable insulating layers with embedded Ag nanoparticles in elastic polyurethanes (TPU) can form conductive filaments in the polymer matrix under an electric field to switch the conductivity of the insulating layer. Au nanoparticles embedded in the TiO$_2$ nanomembrane can significantly reduce the set and reset currents of the insulating layer in the stretchable RRAMs. Similarly, introducing different
nanofillers such as Ag nanoparticles,[127] Au nanoparticles,[128] carbon nanotubes,[129] and CdSe nanoparticles[130] into the elastic polymer hosts such as polydimethylsiloxane (PDMS), TPU, styrene-(ethylene-butylene)-styrene (SEBS), ethylene-vinyl acetate (EVA) copolymer, etc. can be explored to realize intrinsically stretchable intermedia layers for conformable RRAMs.

While stretchable conductive polymers could be readily synthesized for stretchable conductors, the development of stretchable polymers for the intermedia active layer in which the resistance can be switched under electric filed has also been demonstrated.[63,98] Hung et al. has reported the synthesis of MH-b-PI copolymer as the stretchable intermedia layer for stretchable RRAMs (Figure 3B).[98] The PI copolymer serves as the neutral rubber to provide stretchability, and the MH blocks provide the charge-trapping capability to switch the memory between a high-resistance state (HRS) and low-resistance state (LRS). The modification of the organic intermediate layers, such as P3HT[133] and PEDOT:PSS,[134] which can switch their resistance under an electric field based on the filament conduction mechanism associated with carbon-rich filaments by local degradation of organic films or metallic bridges from the migration of the electrode, is also promising to realize the stretchable intermedia layers for stretchable RRAMs.

In contrast, ionic conducting polymers in which charges are transported by ions have gained increasing interest for soft and stretchable electronics.[135–144] Ionic conducting polymers can be easily fabricated into a stretchable structure with exceptional transparency and stretchability using polymeric materials as the ion transport layers. By the migration of the dopant ions in the polymeric matrix, rewriteable switching effects can be achieved in the ionic memory devices (Figure 3C).[132,145] It is believed that the ionic conducting polymer will also become a strong candidate for developing intrinsically stretchable intermedia layers for elastic RRAMs.

### 2.3. Stretchable Substrate

Stretchable polymeric elastomers function as supporting substrates for various stretchable electronics. It provides mechanical integrity for devices that are built on the surface. It should have a moderate Young’s modulus, high elasticity, and high elongation before fracture. Such material properties are typical for various stretchable elastomers like PDMS, polyurethane (PU), ecoflex, and acrylic elastomer which are commonly used as substrates for stretchable electronics. These materials are also known to be able to undergo repetitive cycles of stretch/release. Among these substrates, ecoflex, polyurethane, and acrylic elastomers have significantly higher maximum strains, significantly higher than 100% (Table 2). Despite having excellent stretchability, low service temperatures (<90 °C) of polyurethane[146] and acrylic elastomer[147] limit the use of these materials as substrates for stretchable RRAM, where subsequent device fabrication/material growth takes places at higher temperatures. PDMS is the most extensively used stretchable substrate as it is low cost, transparent, biocompatible, and easy to process. Although its surface is known to be hydrophobic, O₂ plasma treatment[148] can be applied to increase its hydrophilicity for better wetting or interfacial adhesion with the subsequent layer in RRAM fabrication.

As introduced earlier, the stretchable resistive memory device typically requires a three-layer stack of top and bottom electrically

### Table 2. Elastomers for stretchable substrates and their elastic properties.

| Material                            | Tensile strength [MPa] | Maximum strain [%] |
|-------------------------------------|------------------------|--------------------|
| PDMS[149]                           | 6.25                   | 160                |
| Ecoflex 00-30[147]                  | 1.38                   | 900                |
| Polyurethane[150]                   | 7.32                   | 760                |
| Acrylic elastomer (VHB 4910)[151]   | 0.69                   | 500                |
conducting electrodes and an insulating switching medium to form complete devices. Ensuring mechanical integrity and retaining memory switching while being stretched still remain a challenge before these devices are adopted for widespread applications. Several strategies have been in place to solve these shortcomings. Graphene has been a good choice as electrode material due to its high electrical conductivity and mechanical strength. Graphene grown by chemical vapor deposition (CVD) on copper foil has been used as a conducting electrode for stretchable memory device, where the transfer method can be readily used to deposit graphene onto PDMS (Figure 4A). To prevent the mechanical rupture of graphene and maintain its electrical conductivity, the wrinkled structure has been used. This structure can be realized using a prestrain on the PDMS substrate before the transfer of graphene. After slowly releasing the substrate from the determined prestrain, the graphene electrode (alongside the memory device fabricated on top of it) will inevitably wrinkle up to form waves. This final wrinkled structure will enable the electrode stretchability by converting the vertical displacement and lateral strain into elongation. Scanning electron microscope (SEM) images of the top view and tilted view of the wrinkled graphene on PDMS are shown in Figure 4B,C, respectively. The wrinkles provide a “spring-like” compressive strain, which can be released to the neutral state when stretched. The finite element simulation of the wrinkled structure (Figure 4D) shows the strain distribution in the wrinkled structure when the prestrain has been released, which results in wave-like shapes with a radius of curvature of 2 μm. Atomic force microscope (AFM) imaging (Figure 4E) shows nanosized wrinkles formed when the prestrain on the PDMS is released. When being stretched during the electrical characterization of the memory device, the interconnecting structure of graphene ensures that the electrode will conduct normally. Although this substrate modification is reported in many other works to prepare stretchable memory devices, it has its shortcomings. While a prestrain is applied in a uniaxial direction, compressive stress in the orthogonal direction will cause mechanical failure of the graphene film when the prestrain is released. To overcome this problem, prestrain in the orthogonal direction can be applied simultaneously to offset the orthogonal compression. This biaxially pretrained substrate will form wrinkles in both directions, as shown in Figure 4F.

Maintaining mechanical integrity of the electrode and switching medium is important in ensuring the proper functioning of the stretchable memory device. Interface engineering of the substrate and electrode or the switching medium also plays a role. The surface roughness of electrodes plays a major role in determining the resistive memory effect. Tuning the interfacial roughness of reduced graphene oxide (r-GO) is critical in obtaining good resistive switching properties. r-GO films can be conveniently prepared by solution processing and spin coating. By tuning the sonication time of the r-GO solution suspension, surfaces with varying roughness can be obtained (Figure 4G). Surface roughnesses of 4.54 and 2.76 nm are obtained for sonication times of 3 and 18 h, respectively. Hence, the interfacial roughness of r-GO electrodes can be effectively modified by controlling the sonication time of the r-GO solution. As it turns out, this interfacial roughness plays a key role in tuning the resistive switching memory. The PDMS substrate with rough r-GO (r-rGO) will enable sharp morphology at the interface with the switching medium as compared with a flat surface or that of the smooth r-GO (Figure 4H). During electrical biasing, the enhanced localized electric field is prevalent at the tip of the sharp morphology, lowering the energy barrier to form carbon-rich filaments (Figure 4I), leading to resistive switching behaviors. On the contrary, no resistive switching behavior is observed with the smooth r-GO interface. Tuning the interfacial roughness of electrodes can be used as a universal strategy to prepare stretchable resistive switching memory devices.

PDMS is by far the most important polymeric material used for stretchable devices and has received much attention due to its unique properties like high transparency, electrically insulating, low surface energy, good weather resistance, and low toxicity. Despite that, it still suffers from insufficient mechanical properties due to the weak interaction of the PDMS polymer chains, reducing its effectiveness for practical applications that require high mechanical strength. There have been attempts to mitigate this, as published in several works, that blends in a variety of reinforcing fillers to improve the mechanical strength. Carbon black, carbon fiber, carbon nanotubes, and fumed silica are some examples of additives that have been blended into PDMS to improve its mechanical strength. However, due to the increasing size of these fillers, it causes a degradation in its electrical and optical properties. It is shown that the breakdown field strength reduces with increasing amounts of multwall carbon nanotubes (MWCNTs), where there is excess low-resistance points agglomerated in conductive MWCNTs under a high electric field. Nanosilica sol has been widely applied in coatings to improve transmittance and mechanical properties. Likewise, monofunctional silane and tetrafunctional silane (MQ) silicone resin has been used to improve the transparency of PDMS composites. Combining the merits of both fillers will stand to mitigate aforementioned shortcomings. About 30 wt% of nanosilica sol was added to PDMS, with varying amounts of MQ silicone resin as reinforcing fillers. It will be seen that the varying amounts of MQ silicone resin have a modulating effect on the thermal stability, mechanical, and optical properties of the PDMS composite. The preparation procedure is shown in Figure 4J. Due to the strong interaction between the hydroxyl groups of the silica sol and functional groups of the silicone resin (i.e., Si–OH and Si–O–CH$_3$), particle size was increased during the preparation steps. Clearly, the amount of MQ silicone resin added has a modulating effect on the size and morphology of the aggregated particles in the composite. Nanosilica sol can be observed as small particles, down to 1.3 μm in size, that is well dispersed on the PDMS (Figure 4L). Figure 4L clearly shows the evolution of the increase in aggregated particle size with increasing amounts of MQ silicone resin. It can also be seen that these particles are well embedded into the PDMS matrix, indicating good interfacial adhesion between the filler particulates and PDMS matrix. Up to 25 wt% of MQ silicone resin, the particles are intimately embedded within the PDMS matrix, implying the strong interaction between the aggregated particles and the PDMS chains. The addition of MQ silicone resin can significantly improve its mechanical properties (Figure 5A). The tensile strengths of these composites (with nanosilica sol) increase from 0.51 to
1.94 MPa for 10 and 25 wt% of MQ silicone, respectively (Figure 5B). These values are much larger than PDMS nanosilica composites without MQ silicone resin (0.24 MPa). Young’s moduli also show a similar increasing trend, with an increase from 0.13 to 0.2 MPa. Remarkably, the elongation before rupture increases from 473% to over 1000% for additions of 10–25 wt% of MQ silicone resin (Figure 5C). These reported values far exceed other works, reaching a breakthrough in the mechanical properties of PDMS composites. The strong interactions between the aggregated particles and PDMS chains with increasing amounts of MQ silicone resin account for the significant improvement in its mechanical properties. In addition, the
3D networks of the particles and PDMS matrix enable more load transfer and obstruct rupture effectively, resulting in improved mechanical properties.

Although obtaining a high tensile strength is desirable for elastomeric substrate, toughness is another property that is of importance. In the advent of wearable electronics and human-to-machine interface, skin-like mechanical properties are desirable, where there is an intrinsic self-limiting mechanical behavior in which the toughness increases at high stretching rates to prevent rupture. Such mechanical properties can be obtained by adding poly[(vinylidene fluoride)-co-trifluoroethylene] (P(VDF-TrFE)) as reinforcing fillers into PDMS matrix (Figure 5H). P(VDF-TrFE) have high toughness and strength, whereas PDMS has a low Young’s modulus. Combining these two materials will result in a composite with high strength and toughness. Such hybrid material is highly stretchable at a low strain but, becomes very tough at a large strain, similar to the human skin. This mechanical behavior can be controlled by tuning the amount of P(VDF-TrFE) nanofibers incorporated into the PDMS matrix. Embedding the nanofibers into the PDMS matrix can be accurately controlled using the electrospinning method, with varying amounts of the electrospun (ES) volume of P(VDF-TrFE) solution. As shown in Figure 5, the increasing amounts of ES volume of the nanofibers result in the increase in density of the nanofibers network. The mechanical behavior of these composites can be separated into three different phases (I, II, and III) which correspond to low loads, increased loads, and high load regions. These three phases describe the dynamic mechanical behavior in the microscopic scale. At region I, the stress–strain behavior is dominated by PDMS, with high stretchability. At region II, the nanofibers start to stretch out with increasing load. At region III, the nanofibers start to move past one another, accumulating rupture until failure occurs. These three regions can be seen in the step-like stress–strain curves, as shown in Figure 5I. Increasing the amount of P(VDF-TrFE) nanofibers significantly improved its strength, toughness, and Young’s modulus, albeit at the expense of stretchability (Figure 5). The Young’s modulus of the composite can be modified in the range of 0.04–0.3 MPa by controlling the loading amount of P(VDF-TrFE) nanofibers. The improved mechanical properties and demonstration of dynamic properties of this composite have shown to behave similar to the human skin, paving the way of using these types of composites as substrates for stretchable electronics.

Figure 5. A) Stress versus strain curve, B) Tensile strength and Young’s modulus and C) elongation at break of PDMS composites with varying amounts of MQ silicon resin. Reproduced with permission.[154] Copyright 2015, Elsevier. SEM images showing morphology of P(VDF-TrFE) nanofibers at ES volumes of D) 0.2, E) 0.4, F) 0.6, and G) 0.8 mL, respectively. H) Schematic of embedded P(VDF-TrFE) nanofibers acting as fillers in PDMS matrix. I) Stress–strain curves, J) % stretchability, and K) toughness of the PDMS composite with different ES volumes of P(VDF-TrFE). Reproduced with permission.[165] Copyright 2018, American Chemical Society.
3. Progress on Stretchable RRAM

In the earlier section, we reviewed and discussed the respective stretchable components of stretchable RRAM, including the electrode, dielectric layer, and substrate. For the fabrication and application of stretchable RRAM, the difficulty is not only that all parts of the device can be stretched, but also the internal stress of the interlayer interface and the stability of the switching state during the stretching process and after the recovery. To develop a satisfactory and expected stretchable RRAM, we should consider the mechanical properties of the device and the electrical reliability of the performance during application. Therefore, the feasibility of switching mechanisms, material reliability, and device construction should be carefully considered. In this section, we will introduce the representative research progress of stretchable RRAM from the perspective of material selection and structural design.

3.1. Intrinsically Stretchable RRAM

On the fabrication of stretchable RRAM, the electrodes and dielectrics that can be stretched intrinsically are considered to be the best choices. Most of the inorganic materials are generally considered to be brittle or lack of toughness. Due to their physical and chemical stabilities, researchers are trying to obtain the tensile properties by material engineering on inorganic materials, even though the stretchable RRAM with only a smaller strain is obtained. Recently, Shang et al. introduced an all-oxide-based RRAM device that could withstand a mechanical strain of up to 2.12% using the amorphous–nanocrystalline hafnium oxide (HfO$_x$).

As shown in Figure 6A, with indium tin oxide (ITO) as the electrode, ITO/HfO$_x$/ITO RRAM has been fabricated, in which the HfO$_x$ dielectric exhibited an amorphous–nanocrystalline-mixed morphology. The ITO/HfO$_x$/ITO RRAM showed a certain strain stability and stable resistive switching performance. The mechanical deformations and corresponding strains of the ITO/HfO$_x$/ITO RRAM were evaluated by bending the devices with different radii (Figure 6B). The strain as a function of bending radius was estimated with the equation $e = t/2R$

where $e$ is the calculated strain and $t$ and $R$ is the device thickness and bending radius, respectively. The experimental and fitted results of strain-bending radii are shown in Figure 6C. The set and reset voltages of ITO/HfO$_x$/ITO RRAM were measured at different radii (Figure 6D). The ITO/HfO$_x$/ITO RRAM could switch ON and OFF with the strain less than 2.12%. In addition, the endurance performance of the ITO/HfO$_x$/ITO RRAM was tested with the pulse mode at the strain of 1.06% for 1200 cycles (Figure 6E). As shown in Figure 6F, the retention times of the HRS and LRS of the ITO/HfO$_x$/ITO RRAM were also measured at the strain of 1.06%. As the ITO/HfO$_x$/ITO RRAM only works at the small strain level, it is difficult to meet the real stretchable application.

Compared with the inorganic hafnium oxide used as dielectric material mentioned earlier, organic polymers have inherent advantages in the research and application of stretchable electronics. In fact, the organic polymers have been reported in RRAM research. Chen and coworkers developed a series of organic-based stretchable electrical memory devices using chemically synthesized polymers. As shown in Figure 6G, a new stretchable diblock copolymer of maltoheptaose-block-polyisoprene (MH-b-PI) was synthesized. MH-b-PI was demonstrated with the charge-trapping capability and stretchability due to the hydrophilic oligosaccharides block and polyisoprene block, respectively. With the MH-b-PI and PDMS as the dielectric and substrate, respectively, the CNTs/MH-b-PI/Al configured stretchable memory device was fabricated (Figure 6H). The network CNTs and wrinkled Al electrode exhibited a stable conductivity during the stretching characterization. The switching performances of the ON/OFF current ratio and set voltages of the MH-b-PI-based stretchable memory were analyzed. As shown in Figure 6I, the ON/OFF current ratios stabilized around 10$^6$ with the mechanical strain from 0% to 100%. The set voltages were decreased during stretching. In addition, the MH-b-PI-based stretchable memory still works after 1000 cycles with 40% strain. The application of these intrinsically stretchable materials makes the preparation of the stretchable RRAM feasible.

In addition to the aforementioned use of a stretchable dielectric to prepare a stretchable memory, a stretchable RRAM can also be prepared using the composite consisting of a traditional stretchable polymer matrix and the metal nanoparticle as a dielectric layer. Yang et al. proposed a stretchable synapse memory device with the synaptic function features including the long/short-term plasticity and potentiation/depression characteristics. As shown in Figure 7A, the stretchable memory was fabricated by laminating the separately prepared top electrode, medium layer, and bottom electrode together. The Ag nanoparticle/thermoplastic polyurethanes (TPU:AgNPs) composite with the high elastic property acted as a medium layer. The PDMS was selected as a supporting layer. As shown in the photograph of Figure 7B, the Au/TPU:AgNPs/Au memory device exhibited good elasticity and stretchability. To identify the electrical performance of the Au/TPU:AgNPs/Au memory device, the current–voltage characteristics were measured at the original state and at a 35% strain state (Figure 7C, inset photograph). With the direct current (DC) voltage sweep applied on the Au/TPU:AgNPs/Au memory device, the current–voltage curves exhibited a history-dependent asymmetric resistive switching behavior at both original and stretched states (Figure 7C). In addition, to further study the basic synaptic function of the Au/TPU:AgNPs/Au memory device at the original and stretched states, the potentiation/depression characteristics were evaluated by the consecutive negative/positive pulses operation. As shown in Figure 7D, the Au/TPU:AgNPs/Au memory device displayed similar potentiation/depression characteristics both at the original state and at a 35% strain state. The mechanism of the resistance switching of the stretchable Au/TPU:AgNPs/Au memory device was proposed as the formation of Ag conductive filaments in the medium layer.

With the progress of material synthesis technology and composite engineering, new materials or composites with expected properties were prepared and promoted the development of electronic devices. Yi et al. reported an intrinsically stretchable RRAM with the elastic conductor and mechanical deformable insulator medium layer. The elastic conductor was prepared by mixing the GaInSn liquid alloy and PDMS prepolymer. The GaInSn liquid alloy exhibited a calabash bunch network.
in the PDMS matrix. As shown in Figure 7E, the resistance variation of the stretchable GaInSn@PDMS electrode with a strain range from 0% to 105% was characterized. The resistance variation less than 4.13% at the stretching strain of 108.14% indicates the promising reliability for the stretchable electrode. The aluminum-based MIL-53 metal-organic framework nanofilm was selected as the deformable dielectric. As shown in Figure 7F, the mechanical flexibility and structural elastic deformation of MIL-53 were endowed from the variation of the dihedral angles. Based on the stretchable MIL-53 nanofilm and GaInSn@PDMS liquid metal, the stretchable Ag/MIL-53/GaInSn@PDMS RRAM was fabricated. As shown in Figure 7G, the RRAM device displayed the bipolar resistive switching behavior from a stretching strain range from 0% to 10%. The cyclability of Ag/MIL-53/
Figure 7. A) Schematic illustration of the fabrication process of the stretchable Au/TPU:AgNPs/Au memory devices with PDMS as substrate. B) Photographs of the Au/TPU:AgNPs/Au memory devices at the original and stretched state. C) The photograph and the corresponding $I$–$V$ characteristics of the Au/TPU:AgNPs/Au memory at the original state and at a 35% strain state. D) The potentiation/depression characteristics of the Au/TPU:AgNPs/Au memory with the positive/negative pulse voltage without and with 35% strain. Reproduced with permission.[65] Copyright 2018, The Royal Society. E) The resistance variation of the stretchable GaInSn@PDMS electrode with a strain range from 0% to 105%. F) Crystal structure of metal–organic framework MIL-53 (left) and the structural deformation with external mechanical stresses. G) The typical $I$–$V$ characteristics of the stretchable Ag/MIL-53/GaInSn@PDMS device with the strains range of 2–10%. H) The stability and I) the retention performance of the stretchable memory devices at the fixed stretching strain of 10%. Reproduced with permission.[64] Copyright 2019, Wiley-VCH.
GalInSn@PDMS RRAM was demonstrated with the pulse-mode operation at a 10% strain for 50 cycles (Figure 7H). In addition, the retention performances at both HRS and LRS of the stretchable RRAM device were recorded (Figure 7I). The resistance of Ag/MIL-53/GalInSn@PDMS RRAM can be retained for 10^4 s under dynamic stretching–relaxing (with a 10% strain).

3.2. Structurally Enabled Stretchable RRAM

The representative progress of stretchable memory devices depends on the stretchable components including the conductor and medium layer that have been introduced above. However, the stretchable memory devices made using the stretchable components have the interfacial problem due to the differences in Young’s modulus between each layer. Therefore, developing stretchable electronic devices using structural engineering strategies such as wrinkled, mechanical hybrid, and serpentine-structured configurations is still important. Hou et al. developed a stretchable memory device with the switchable graphene oxide/poly(3-hexylthiophene) GO-P3HT complex (Figure 8A) as the medium layer. As shown in Figure 8B, with the ITO-coated PDMS substrate, the GO-P3HT thin film as switching layer, and the Al top electrode, the Al/GO-P3HT/ITO-PDMS structural device was fabricated. They proposed that the mechanical stretchable and resistance state stable features of the Al/GO-P3HT/ITO-PDMS memory device during stretching were on account of the interlayer sliding of GO-P3HT nanosheets between. The schematic diagram and photograph of the Al/GO-P3HT/ITO devices with PDMS substrate at the stretching strain of 50% are shown in Figure 8. The Al/GO-P3HT/ITO-PDMS device was evaluated with I–V characteristics at a 50% strain state and exhibited a bipolar resistive switching behavior (Figure 8E). In addition, the Al/GO-P3HT/ITO-PDMS memory device showed the stable set/reset voltage and ON/OFF ratio with the continuous stretching-relaxing operation from 0% to 50% for 200 cycles (Figure 8F).

By placing or embedding the high Young’s modulus islands on a low Young’s modulus elastomer as a substrate is an alternative for the stretchable electronic device fabrication. Liu et al. presented a mechanical hybrid substrate for achieving the stretchable RRAM device. As schematically shown in Figure 8G, the high-modulus rigid SU-8 flakes were embedded in a low-modulus PDMS to form the mechanical hybrid substrate. The RRAM devices with Ag as the top electrode, zeolitic imidazolate framework–8 (ZIF-8) as the medium layer, and Au as the bottom electrode were prepared on the rigid SU-8 flakes of the mechanical hybrid substrate. Figure 8H shows the microscope photograph of the RRAM devices. The resistance switching behavior of the Ag/ZIF-8/Au RRAM was characterized and is shown in Figure 8I. By grounding the Au electrode and applying the voltage bias to the Ag electrode, the Ag/ZIF-8/Au RRAM showed the setting and resetting voltages of 2.2 and −0.85 V, respectively. Benefited from the mechanical hybrid substrate, the Ag/ZIF-8/Au RRAM device was tested at different strains from 0% to 50% and showed no obvious change of HRS and LRS at stretching states (Figure 8J). In addition, the retention measurement demonstrated that the resistance states of Ag/ZIF-8/Au RRAM device could remain stable for 3000 s at stretching strains of 0%, 30%, and 50% (Figure 8K). This structural engineering strategy of the mechanical hybrid substrate is valuable for stretchable electronics.

In addition to the aforementioned mechanical hybrid structure, the serpentine structure was also reported and demonstrated as an effective strategy to obtain stretchable electronics.[37,40,67,173] Son et al. developed a multifunctional wearable device containing the stretchable RRAM that was used for data storage.[67] The RRAM devices with TiO2/Au nanoparticles (AuNPs)/TiO2 as the medium layer and Al as both the bottom and top electrodes were first fabricated on the rigid wafer and subsequently transferred on a PDMS substrate. As shown in Figure 9A, the microscope image of the serpentine-structured stretchable RRAM device at the stretching strain of 25% showed that only the serpentine electrode in the stretching direction shows obvious deformation. The corresponding finite element modeling (FEM) of strain distribution at the stretching strain of 25% demonstrated that the intersections of serpentine electrodes, i.e., RRAM, have no obvious strain (Figure 9B). As shown in Figure 9, the serpentine-structured stretchable RRAM devices can attach on and deform along with human skin. The serpentine-structured stretchable RRAM was evaluated with I–V curves with the strain range from 0% to 25% and showed a stable electrical operation (Figure 9E). The stable cyclability of the serpentine-structured stretchable RRAM was demonstrated at a 30% strain level for 1000 cycles (Figure 9F).

In addition, the indium zinc oxide (IZO)-based stretchable RRAM was also constructed with the similar serpentine structure.[37] The stretchable RRAM was fabricated with the Au/IZO/Au configuration with polyimide encapsulations. Figure 9G shows the schematic drawing and the optical microscopic image of the IZO-based stretchable RRAM. The I–V curves of the IZO-based stretchable RRAM in Figure 9H exhibited a typical bipolar resistive switching performance with the set and reset voltage of 0.96 and −1.6 V. The write–read–erase–read cycles of the IZO-based stretchable RRAM were recorded and demonstrated its feasibility (Figure 9I). In addition, the sequential microscope images and corresponding FEM of the strain distribution of IZO revealed the advantages of the serpentine-structured electrodes design (Figure 9J).

4. Wearable RRAM

Nowadays, the electronic elements including power supply, display, and sports performance monitoring devices are usually integrated into the fabrics/textiles to meet the smart wearable requirement.[32,174–176] The vision behind wearable computing has led to the development of electronic textiles that inevitably require wearable memory that will be mounted or formed on fabric.[35,177] The considerations in terms of comfort, the wearableability of the electronic garment substrates, washability, and reliability require more investigations.

Bae et al. reported a polydopamine (PDA)-intercalated fabric memory with the RRAM configuration for wearable devices.[177] As shown in Figure 10A, the Al/PDA-coated yarns and the insulated cotton yarns without coatings were interwoven repeatedly to form the fabric-based RRAM. The magnified scheme of the RRAM based on two crosslinked PDA/Al-coated yarn is shown.
in Figure 10B. With the creative design, the wearable fabric-based RRAM was obtained and the photograph of the wearable RRAM is shown in Figure 10C. The electrical characteristics of the as-prepared wearable fabric-based RRAM were further measured. Figure 10D shows the electrical forming process of the conductive filaments in the Al/PDA/Al-configured RRAM. The write and erase of the two-terminal Al/PDA/Al RRAM could mimick the function of the synapse. As shown in Figure 10E, the typical I–V curves revealed the expected set and reset procedure of the Al/PDA/Al-configured RRAM. In addition, Kang proposed
a nonvolatile nylon thread (NT)-based memory device with a dip-and-dry process. Figure 10F shows the NT-based resistive memory with three segmented lengths corresponding to the LRS, HRS, and LRS, respectively. These three parts also exhibited a long retention capability (Figure 10G). In addition, the NT-based memories were also integrated into textiles for the wearable application (Figure 10H). This integrated fabric memory could withstand 50% tensile deformation (Figure 10I). These initial perspectives on textile-based memory promoted the research enthusiasm on the wearable data storage devices and presented us the varied possibility of future memory.

Figure 9. A) The microscope images and B) the finite element modeling (FEM) of strain distribution of the serpentine-structured stretchable memory devices at a stretching strain of 25%. Photographs of the stretchable memory devices under C) no strain and D) tension. E) The $I-V$ characteristics of the serpentine-structured memory devices at different strains. F) Resistance cyclability between LRS and HRS for 1000 stretching cycles. Reproduced with permission. Copyright 2014, Springer Nature. G) Schematic drawing and optical microscopic images of the IZO-based stretchable RRAM. H) $I-V$ curves of the resistive switching of the RRAM. I) Write–read–erase–read cycles of the stretchable RRAM. J) Sequential microscope images of the IZO-based RRAM and corresponding FEM of strain distribution of IZO. Reproduced with permission. Copyright 2019, AAAS.
5. Conclusions and Perspectives

In this article, we first introduced the advantages including the simple sandwich structure and high operation speed, low power consumption, high-density data storage, and scalability of RRAM compared with other nonvolatile memory such as ferroelectric, magnetoresistive, phase-change random-access memory. The requirements and expectations of the portable and wearable electronic devices on the stretchable RRAM are discussed. The fundamental components, materials selection principle, and configuration design strategy of the preparation of the stretchable RRAM are systematically summarized. Particularly, the basic electrical and mechanical properties of stretchable components including the electrodes, intermedia, and substrate for RRAM are reviewed. New materials with the intrinsically stretchable features toward the realization of stretchable RRAM are discussed. Novel configuration design strategies to achieve the stretchable RRAM are summarized and evaluated. These representative works have made tremendous progress and opened up the initial ideas for the stretchable RRAM fabrications. The successful development of stretchable RRAM devices will be attractive for advanced memory computing, artificial synapse, and neuromorphic systems for use in artificial intelligence and promote the revolutionary development of storage technology.

Although substantial research achievements have advanced the development of stretchable RRAM, there are still many problems related to the stability, continuous/scale-up fabrication, and reliability issues to be solved for the application of stretchable RRAM. To further promote the development of the stretchable RRAM, the following issues require critical attention. 1) The deeper understanding on the fundamental switching mechanisms and the failure mechanisms of stretchable RRAM are needed to guide the optimization of the devices. 2) The optimized physicochemical parameters including the large ON/OFF ratio, high write/read speed, lower switching voltage, and high-density data storage of the stretchable RRAM devices are expected to be reliable. 3) The facile and low-cost fabrication technologies for the stretchable RRAM are considered as critical for its large-scale production. The compatibility of the wearable
RRAM with the electronic textiles is expected. 4) Regarding improving the device durability, and intrinsic electrical defect-free and mechanical robust insulator/intermedia layer is required. Proper encapsulation or passivation on the stretchable RRAM which does not impede the mechanical property will be required. Thermal stability and thermal management should be considered carefully to avoid thermal runaway and leaky pathway formation upon long-term cycling applications. In addition, the development of self-healing stretchable components including the conductor, insulator, and substrate is a possible approach to improve the device durability of stretchable RRAM. 5) The link circuitry such as wordlines and bitlines and the leakage current paths of the crossbar array during the integration between the stretchable RRAM and high-density circuits have to be considered. Finally, in the sustainable development viewpoints, the applied materials for stretchable RRAM are expected to be environmentally friendly and even recyclable.

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Conflict of Interest

The authors declare no conflict of interest.

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