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Mini-Review

Recent Advances in Halide Perovskite Resistive Switching Memory Devices: A Transformation from Lead-Based to Lead-Free Perovskites

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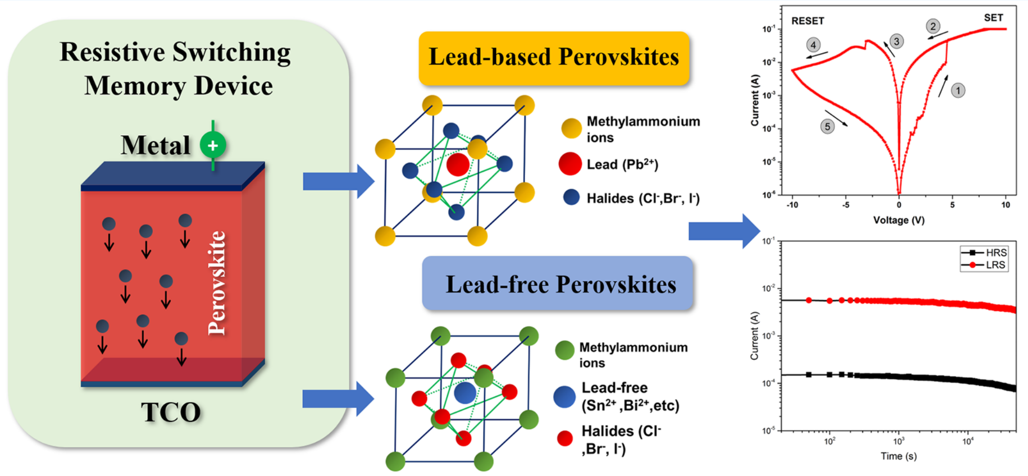

ABSTRACT: Due to their remarkable electrical and light absorption characteristics, hybrid organic−inorganic perovskites have recently gained popularity in several applications such as optoelectronics, lasers, and light-emitting diodes. Through this, there has recently been an increase in the use of halide perovskites (HPs) in resistive switching (RS) devices. However, lead-based (Pb-based) perovskites are notorious for being unstable and harmful to the environment. As a result, lead-free (Pb-free) perovskite alternatives are being investigated in achieving the long-term and sustainable use of RS devices. This work describes the characteristics of Pb-based and Pb-free perovskite RS devices. It also presents the recent advancements of HP RS devices, including the selection strategies of perovskite structures. In terms of resistive qualities, the directions of both HPs appear to be identical. Following that, the possible impact of switching from Pb-based to Pb-free HPs is examined to determine the requirement in RS devices. Finally, this work discusses the opportunities and challenges of HP RS devices in creating a stable, efficient, and sustainable memory storage technology.

1. INTRODUCTION

Advancements of memory storage and system memory have risen dramatically in recent years as technology progression in today's digital world has matured. This has led to the growth of volatile memory and nonvolatile memory (NVM), which are critical in the current memory technology. NVM devices typically preserve information whether they are turned on or off, whereas volatile memory does not behave in that manner. Flash drives, solid-state devices, random access memory (RAM), read-only memory (ROM), and optical discs are among examples of memory storage technology. In today's storage technology, the NAND flash memory is widely used in most electronics gadgets, where data are stored using electrical states.[1] However, the demand for faster writing speeds, better density, and lower costs drives fresh research into developing newer memory technologies such as resistive RAM (ReRAM). Thus,

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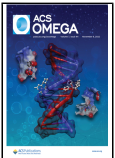

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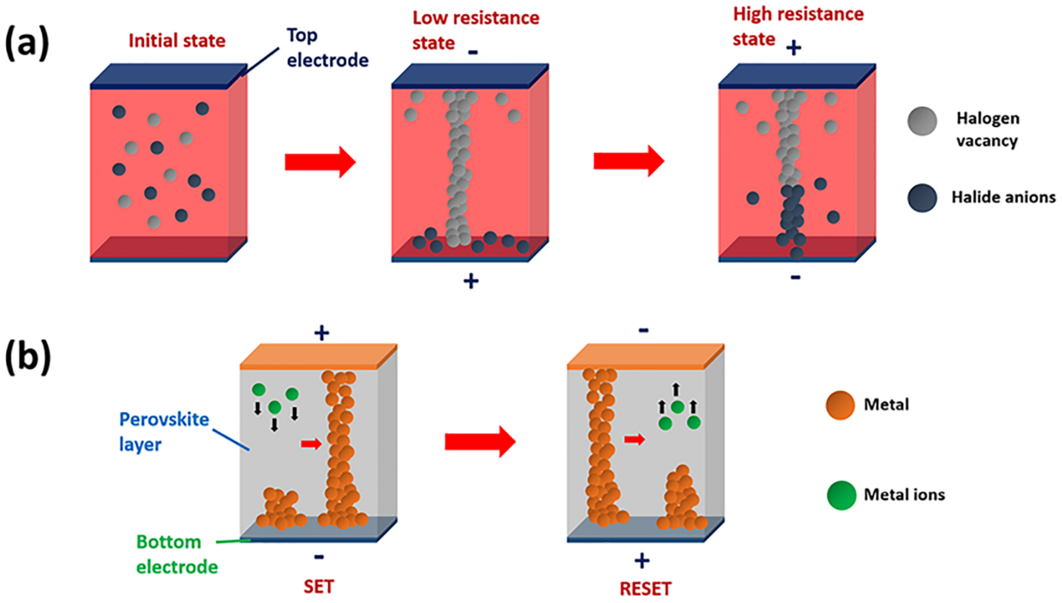

Figure 1. Diagrams of conductive filament processes based on (a) halide anions and (b) electrochemically active metal cations (metal^+).

there is now an urgency for more efficient, less priced, and high-performance memory storage. One potential contender involves the employment of resistive switching (RS) devices in which data are stored between low- and high-resistance states. RS occurs when the resistance of a dielectric abruptly changes in response to a strong electric field or current applied. With the expanding development of RS research, the technology exhibits great prospects for realizing RS devices in the mass market.[2]

Among the various types of materials studied for RS devices, perovskites are a newer class of nanomaterials that has been recently used in RS devices. Typically, perovskites have a general formula of ABX_3, where A and B are cations and X are anions in the perovskite structure. They are normally classified into two primary perovskite groups: perovskite oxides and perovskite halides. Through the many reported application uses of perovskites, photovoltaics (PVs) appears to be getting the highest benefit as the next-generation solar energy technology. This is usually done by harnessing the solar capabilities of halide perovskites (HPs). As a result, various disciplines have begun to investigate its possibilities, notably in RS devices. The use of HPs in RS devices has been initiated in recent years with multiple reported works displaying great potential. HP RS devices have advantages of simplicity, flexibility, and low processing temperatures over oxide perovskites RS devices.[3] Although there have been several reviews on HP RS devices, there has yet to be one on lead-free (Pb-free) perovskite RS devices, which are gaining prominence over lead-based (Pb-based) perovskites in various applications. In this work, only HPs in RS devices will be examined. This brief review will investigate the design and operating principles and current achievements in HP RS devices. Moreover, the possibilities in transitioning from Pb-based to Pb-free perovskites and prospects in this Pb-free HP RS sector will be explored.

2. OVERVIEW OF HPs IN RS MEMORY DEVICES

2.1. Design Structure of HPs RS Memory Devices. With its cheap manufacturing cost, low operation voltage, high ON/OFF ratio, and remarkable mechanical features, HPs have sparked a lot of interest for usage in RS devices. However, due to the limited lifespan and stability of the HP films, HP-based RS memory systems continue to face a variety of challenges. This section provides a high-level overview of HP-based design structures and its associated RS behavior. The universal design features a capacitor-like structure with a metal—insulator—metal (MIM) switching layer between the top and bottom electrodes in the RS memory. Inert electrodes such as Au, Pt, and W, as well as active electrodes (AEs) such as Ag, Cu, and Ni, were utilized. These electrodes served as the carrier's transport pathways and counter electrodes for reduction. However, active metals have a significant impact on RS behavior via electrochemical reactions of electrodes related with the formation and rupture of the conducting filament (CF).

When a positive bias is provided to the AE, the atoms of active metal atoms that will be ionized at the AE are diffused at the counter electrode and then reduced in the insulator layer. Yoo et al. used a simple solution approach to create a novel simple bilayer structure of Ag and $CH_3NH_3PbI_{3-x}Cl_x$ perovskite material on a fluorine-doped tin oxide (FTO) substrate.[4] The $Ag/CH_3NH_3PbI_{3-x}Cl_x/FTO$ device structure demonstrates bifunctional RS behavior with both digital and analogue features. The Ag filament generated by the redox reaction of the Ag electrode displayed bistable RS behavior with 10^3 times of dependable endurance and 4×10^4 s retention period. Later, Yan et al. revealed that the swap of Ag with Al electrode reached the first cycle ON/OFF resistance ratio of 10^9 in Pb HP memory.[5] They created a sandwich of $Al/CH_3NH_3PbI_{3-x}Cl_x/TiO_2/FTO$ structure and discovered that Al was the optimum electrode for instantaneous switching with a low operating voltage. Yan et al. also created a fiber-shaped NVM device on titanium wire with a $Ti/TiO_2/CH_3NH_3PbI_{3-x}Cl_x/Au$ structure, while revealing an ON/OFF ratio of 20 and a set voltage of 1 V.[6] However, no information about performance was provided. This investigation demonstrated that MIM memory cells based on HPs may be manufactured without the use of traditional electrode deposition.

Making a low-dimensional-layered HP can aid in the stabilization of HP-based RS devices. Because of their low stability, the RS characteristics of 3D HP memory devices

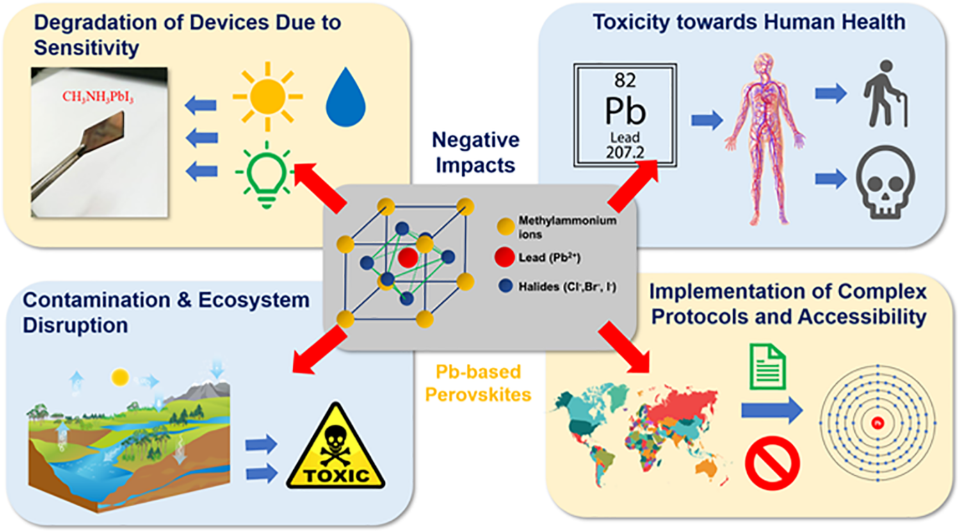

Figure 2. Diagram representing the negative effects of Pb in Pb-based perovskites.

decrease in humid and ambient settings. To address this problem, 2D or quasi 2D HPs have been studied to improve the ON/OFF ratio and prevent the device from deteriorating in an ambient environment. Seo et al. reported that the utilization of 2D $BA_2MA_{n-1}PbnI_{3n+1}$ ($n = 1$) perovskite reveals the best resistive qualities when compared to quasi-2D and 3D perovskites.[7] This was due to the large Schottky barrier heights and thermal activation energy in improving RS performance. In other works, Jung et al. built Au/$CsPbBr_3$/ITO and Au/$CsPb_2Br_5$/ITO RS devices to explore their memory behavior.[8] $CsPb_2Br_5$ lower dimensionality raises the Schottky barrier height between the electrode and $CsPb_2Br_5$, resulting in a higher ON/OFF ratio than $CsPbBr_3$. The fabricated $CsPb_2Br_5$-based RS device retained RS behavior even at 140 °C. Kim et al. also created quasi-2D HPs, $(PEA)_2Cs_3Pb_4I_{13}$, as opposed to 3D $CsPbI_3$.[9] The $(PEA)_2Cs_3Pb_4I_{13}$ films were effectively synthesized by thermal evaporation and had a high ON/OFF ratio of 10^9, which was three times that of the $CsPbI_3$-based memory device. Furthermore, it produced a strong Schottky barrier, which increased the activation energy while decreasing the high resistance state current. For 2 weeks, the device based on $((PEA)_2Cs_3Pb_4I_{13}$ is very stable in an ambient environment at room temperature. Because of their excellent ON/OFF ratio and long-term stability for practical use, two-dimensional HP offers potential in RS memory. However, all of the works mentioned above typically involve the use of toxic Pb. To address the toxicity of the heavy metal Pb, 2D Pb-free HPs modified by doping or totally replacing Pb with Pb-free materials are worth researching.

2.2. Working Mechanism of HPs RS Memory Devices. Understanding the RS process is crucial for optimizing film preparation, creating device architectures, increasing device performance, and realizing large-scale integration. Many theories have been proposed to explain this RS behavior, including trap-controlled space charge-limited current, interfacial Schottky barrier, and CFs. For revealing the switching process of HP RS devices, the notion of CF formation/rupture is widely recognized. The switching mechanisms are typically classified as anionic and cationic, with the former being responsible for memory behavior. Figure 1 displays schematic drawings of two types of RS commonly found in perovskite RS devices. For the anionic mechanism, halide vacancies are equally spread throughout the perovskite layer before applying an electric field to a new device. However, the accumulation and migration of halide vacancies under the influence of an electrical field can result in the formation and rupture of CFs, resulting in recurrent RS behavior.[10] Meanwhile, electrochemical metallization is a cationic method that creates a RS effect in hybrid organic−inorganic perovskite memories. This method is reliant on the formation of metal cations in electrochemically active electrodes such as Ag or Cu. Metal ions that are highly mobile move to the opposite electrode and are neutralized by electron absorption. This process will be repeated until one or more metal filaments develop and the gadget switches from HRS to LRS. Sun et al. created an Ag/$CH_3NH_3PbI_3$/FTO structure in order to study the switching mechanism of Pb HP memory.[11] They determined that the thickness of the perovskite influences the switching process. Because the creation and migration of Ag^+ ions may result in the generation of AgX compounds, more halogen vacancies are produced in HP films, causing the Ag electrode to deteriorate.[12] They concluded that the switching mechanism of Pb HP memristors includes both metal and intrinsic vacancy CFs.

Furthermore, Schottky contact resistance is greatly influenced by Schottky barrier height, which determines switching behavior. Ion migration and charge trapping at the perovskite/electrode interface have been seen to change the barrier height, leading in a RS effect.[9,13]

3. RECENT ADVANCES OF Pb-FREE RS MEMORY DEVICES

3.1. HPs From Pb-Based to Pb-Free Perovskites. Due to their promising efficiencies and remarkable performance, HPs, particularly Pb-based perovskites, are extremely popular in perovskite PVs. Such technology has observed the use of HPs such as $CH_3NH_3PbI_3$, $CH_3NH_3PbCl_xI_{3-x}$, and $CsPbBr_3$.[14] As a result, these Pb-based perovskites were investigated for potential applications in harnessing their desirable characteristics. This led to many published studies which discovered promising outcomes when integrating Pb-based perovskites into RS memories. However, the usage of the Pb element was criticized since the production impact was detrimental to long-term use

Table 1. Summary Table of RS Devices Integrating Pb-Free HPs

device structure	RS behavior	ON/OFF ratio	endurance (cycles)	retention (s)	ref
$Ag/PMMA/CsSnI_3/Pt/Ti/SiO_2/Si$	bipolar	$>10^3$	600	7×10^3	1
$Au/PMMA/CsSnI_3/Pt/Ti/SiO_2/Si$		$>5 \times 10^2$	120	–	
$Ag/CsSnBr_3/Pt$	bipolar	$>10^2$	–	–	15
$(ITO)/CsSnBr_3/Au$	bipolar, multilevel	$\sim10^5$	400	$\sim10^3$	17
$Ag/CsSnCl_3/ITO$	bipolar	10^2	10^5	$>10^4$	18
$Pt/CsSnBr_3/Pt$	bipolar	$>10^5$	50	10^4	19
$Ag(AgO_x)/Cs_3Bi_2I_{10}/FTO$	bipolar, multilevel	$\sim10^6$	250	$>10^3$	14
$Ag(AgO_x)/Cs_3Bi_2I_9/FTO$				$<10^3$	
$Pt/Cs_2AgBiBr_6/ITO$	bipolar, multilevel	500	–	1200	20
$Au/(CH_3NH_3)_3Bi_2I_9/ITO$	bipolar, multilevel	10^2	300	$\sim10^4$	21
$Pt/Cs_2AgBiBr_6/ITO$	bipolar	$\sim10^2$	>500	>2400	22
$Cu/(CH_3NH_3)_3Bi_2I_9/ITO$	bipolar	10^4	1730	3×10^5	23
$Ag/BA_2CsAgBiBr_7/Pt$	bipolar	10^7	$\sim10^3$	2×10^4	24
$Al/CsBi_3I_{10}/ITO$	bipolar	10^3	150	10^4	25
$Au/Cs_3Bi_2I_9/Pt/Ti/SiO_2/Si$	bipolar	$>10^7$	200	10^3	26
$Au/Rb_3Bi_2I_9/Pt/Ti/SiO_2/Si$			400		
$Au/Cs_2BiAgBr_6/ITO$	bipolar	10^4	10^2	10^3	27
$Ag/PMMA/(C_6H_5CH_2NH_3)_2CuBr_4/Pt$	bipolar, multilevel	10^8	2000	$>10^3$	28
$Ag/PMMA/Cs_3Cu_2I_5/ITO$	bipolar	10^2	10^2	$>10^4$	2
$Al/Cs_3Cu_2I_5/ITO$	bipolar	~65	200	10^4	29
$Au/Cs_3Sb_2Br_9$ /Au	bipolar	$>10^3$	200	2×10^4	30
$Al/PCBM/Cs_3Sb_2I_9/PEDOT:PSS:ITO$	bipolar	$\sim10^4$	10^2	$>10^4$	31

and sustainability. According to World Health Organization (WHO) guidelines, Pb exposure as low as 5 μg/dL would necessitate immediate medical attention because the consequences might be disastrous for humans. As such, the use of Pb-based perovskites presents concerns, even though Pb-based perovskites outperform their current technological equivalents.[15] The disadvantages of Pb perovskites are well-known to perovskite researchers, as shown in Figure 2, which includes

(i) extreme susceptibility to moisture, heat, and light, which results in rapid degradation and device instability (long-term usage of Pb-based HPs is known to impair the material because small changes in these parameters cause the layer to erode; hence, this will lead to reduced resistive efficiency in RS devices);

(ii) high toxicity to the environment due to contamination of natural resources and disruption of ecosystem maintenance (Pb leakage from Pb-based perovskite devices contaminates soil or water, inhibiting root growth owing to cell division disruption and possibly killing nearby animals due to central nervous system (CNS) damage);

(iii) neurotoxicity on the human CNS causing persistent damage, impairments, or death (direct effects could be morphological (interruptions of critical molecules during neuronal migration and development, impairment on synapse formation, and early differentiation of glial cells); also, pharmacological effects (triggers calmodulin-based processes and disturbance with neurotransmitter release) could be present);[16]

(iv) stringent safety and health requirements that vary by country, which may raise expenditures and limit accessibility (each country or region has different rules governing Pb acquisition, processing, disposal, and wellbeing, making it difficult to establish a common framework in Pb-based technologies).

Consequently, the research trend in RS devices shifted in the same path as perovskite PVs. Pb alternatives were highly investigated since they can replace Pb perovskites while maintaining resistive performance. These alternatives are expected to be far less harmful than Pb and may find use in the expanding trend of green technology. Thus, this has resulted in an increasing number of Pb-free perovskites that investigate the concept of sustainable, less hazardous, and environmentally friendly RS technology. In current research trends, Pb replacements in perovskite RS devices were researched with similar chemical, electronic, and physical characteristics such as Sn,[1,15,17−19] Bi,[14,20−27] Cu,[2,28,29] Ag,[20,22,24] and Sb.[30,31] This is evident in the growing number of scientific works that was found in integrating these Pb-free HPs as high resistive performance memories.

3.2. Types of Pb-Free Perovskites in RS Memory Devices. With the proliferation of HP-based RS devices, many published literature efforts have focused mostly on Pb-based perovskite RS devices. As previously stated, Pb replacements are gaining popularity in perovskite PVs.[31] As a result, one might anticipate a similar development with RS devices. Table 1 shows the increasing number of reported Pb-free perovskites investigated in RS devices. Although the amount of Pb-free perovskite RS publications has remained modest in recent years, there has been a pattern observed in the selection of specific Pb replacements. The usage of Sn and Bi was determined to be a preferred alternative in substituting Pb cations in Pb-free perovskite RS devices.[1,14,15,17−27] This has also similarly been proven in perovskite PVs in which Sn-based perovskites reign triumphant in Pb-free PVs due to the lower band gap in Sn and a better absorption coefficient than the frequently used $CH_3NH_3PbI_3$ perovskites. Moreover, the identical electronic structure and the existence of a lone pair of electrons in the s orbital makes Sn a popular choice in Pb-free perovskite PVs.

Sn-based perovskites have generated good RS results that are comparable to the output standard obtained by Pb-based perovskites. The ionic radius of Sn^{2+} is substantially closer to that of Pb^{2+} (225 to 202 pm, respectively) than it is to Bi. Theoretically, Sn should remain as the primary replacement for Pb in perovskite RS devices. The development of a flexible Pt/

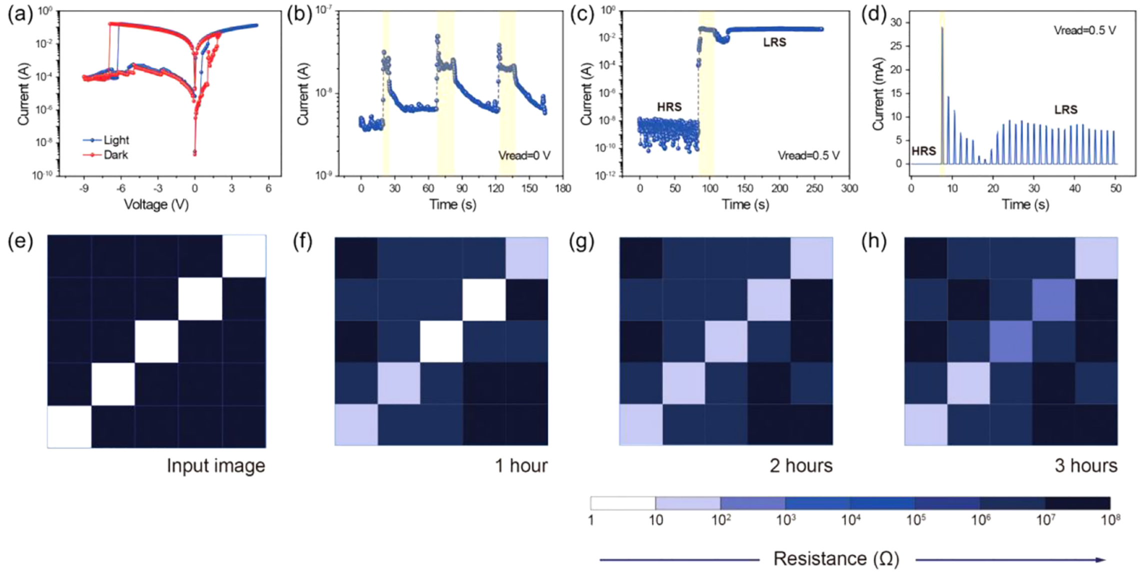

Figure 3. (a) *I*−*V* characteristics of the Cu/$(CH_3NH_3)_3Bi_2I_9$/ITO RS device. Plot representing the photoresponse of Cu/$(CH_3NH_3)_3Bi_2I_9$/ITO RS device at (b) zero bias and (c) 0.5 V. (d) Duration of illumination. (e−h) Schematic diagram of the temporal retention of the Cu/$(CH_3NH_3)_3Bi_2I_9$/ITO RS device in a 5 × 5 square matrix. Reproduced with permission from ref 23. Copyright 2021 Royal Society of Chemistry.

$CsSnBr_3$/Pt device demonstrates that Sn may offer outstanding resistive characteristics.[19] With a good ON/OFF ratio of 10^5, relatively low operating voltages, and bending flexibility of up to 200 cycles. However, the endurance cycles of the said devices can only run up to 50 cycles, implying that stability may be a concern. The stability of Sn-based perovskites must be considered since it has been established that Sn(II) is prone to oxidation due to moisture and heat, resulting in the formation of Sn(IV).

Surprisingly, as observed in Table 1, Bi is mostly used than Sn in fabricating perovskite RS devices. Bi^{2+} (230 pm) has the same ionic radius as Pb^{2+} (202 pm), making it possible to substitute Pb in the perovskite structure.[32] Bi-based perovskites provide a far more stable type of perovskite than Pb-based perovskites. Poddar et al. have demonstrated an outstanding Bi-based perovskite RS device.[23] The fabrication of Cu/$(CH_3NH_3)_3Bi_2I_9$/ITO RS device has a good 1730 cycle repeatability, a long electrical durability of 3 × 10^5 s, and a high ON/OFF ratio of 10^4. Furthermore, their picture storage in a 5 × 5 square matrix (Figure 3e − 3h), which replicates the real-life process, demonstrates a temporal retention of roughly 3 h with no substantial data deterioration. Additionally, Sn-based and Bi-based perovskite RS devices appear to be less reproducible and durable.[1,14,17,19,21−23,25,26] This is evident from the shorter endurance cycles, which may explain why all forms of perovskite RS devices still require further research for an efficient long-term use when compared to other types of materials that can easily outperform them such as inorganic materials.

Alternatively, double perovskites could be potentially applied and studied in perovskite RS devices. Double perovskites have much higher resistive properties than single perovskites. So far, the combination of Bi and Ag suggests the possibility of fabricating efficient RS devices. According to Kim et al., the incorporation of butylammonium (BA) into $CsAgBiBr_7$ resulted in a material dimension reduction to 2D $BA_2CsAgBiBr_7$.[24] The shift from 3D to 2D double perovskites boosts the ON/OFF ratio roughly 5-fold (ON/OFF ratio of 10^7), which is an intriguing finding in this work. The aging impact of the RS device was further investigated using relative humidity of 30−50%, demonstrating that the double perovskite is highly stable. Another double perovskite work reported on the solution processed $Cs_2BiAgBr_6$ film shows promising RS results.[27] This device (Au/$Cs_2BiAgBr_6$/ITO) shows excellent RS properties owing to the defect states caused by the filamentary conduction of bromide vacancies, V_{Br}. With low operating voltages of −0.4 and 1.0 V, endurance of 10^2, and retention properties of 10^3 s, the RS device performance exceeds a few currently reported Pb-based perovskites. This analysis depicts the research potential for using double perovskites as the primary preferred choice for Pb-free perovskites in RS devices.

Up to this point, only a few kinds of Pb-free perovskites other than the commonly used Bi and Sn, such as Cu and Sb, have been investigated.[2,28−31] The use of these reported rare Pb-free perovskites is still ongoing, with a growing number of publications. Mao et al. reported the production of monocrystalline Pb-free $Cs_3Sb_2Br_9$ perovskite nanoflakes in their paper.[30] This first experimental RS device work using Sb demonstrates nonvolatile bipolar RS properties with an electric field of 2.2 × 10^5 V m^{-1} for threshold switching, resulting from V_{Br} migration. Cu has also been used in Pb-free perovskites RS devices. The Ag/PMMA/$Cs_3Cu_2I_5$/ITO memristor produced has excellent RS characteristics (ON/OFF ratio = 10^2, endurance = 100 cycles, and retention = > 10^4 s).[2] The inclusion of a poly(methyl methacrylate) (PMMA) layer was proven to be critical for RS performance because it prevents the perovskite layer from interacting with the Ag electrode to create silver iodide, AgI_x, which is a typical issue in perovskite RS devices. Most importantly, this presented work has been examined for their long-term potentiation (LTP) and long-term depression (LTD) values, which suggest successful usage as a biological synaptic activity recognition rate of up to 94%. Yet, further study and more data is required to establish the utilization of other Pb alternative cations. This suggests that the subject of investigating various types of Pb-free perovskites is still unexplored and may

Table 2. Comparison Table of Some Recent High=Performance RS Devices Employing Pb-Based and Pb-Free Perovskites

device structure	perovskite type	(ON/OFF) ratio	endurance (cycles)	retention (s)	ref
$Ag/BA_2CsAgBiBr_7/Pt$	Pb-free	10^7	$\sim 10^3$	2×10^4	24
$Ag/PMMA/(C_6H_5CH_2NH_3)_2CuBr_4/Pt$	Pb-free	10^8	2000	$>10^3$	28
$Au/CH_3NH_3PbI_3/Au/SiO_2/Si$	Pb	10^8	10^3	$>10^4$	33
$Ag/(PEA)_2Cs_3Pb_4I_{13}/Pt/Ti/SiO_2/Si$	Pb	10^9	200	2000	9

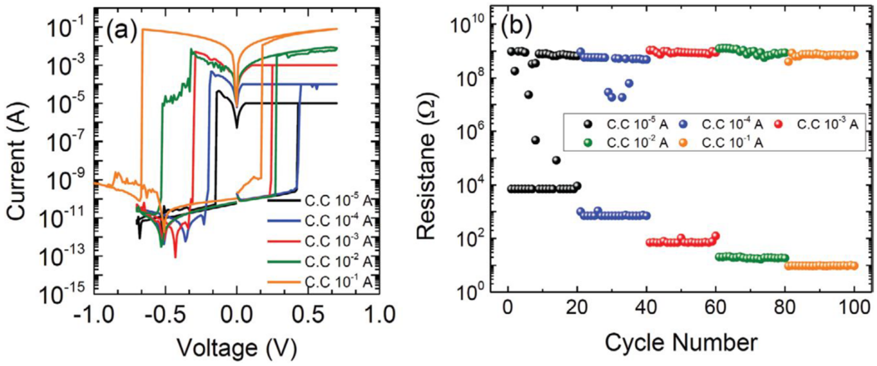

Figure 4. (a) $I-V$ plot indicating multilevel data storage of $Ag/PMMA/(C_6H_5CH_2NH_3)_2CuBr_4/Pt$ RS device. (b) Endurance properties of the $Ag/PMMA/(C_6H_5CH_2NH_3)_2CuBr_4/Pt$ RS device with a read voltage of +0.07 V. Reproduced with permission from ref 28. Copyright 2020 Wiley-VCA Verlag.

provide a strong research opportunity in advancing perovskite RS technology.

3.3. Challenges of Pb-Free RS Memory Devices.

Although Pb-free perovskites are gaining traction in the design architecture of RS devices, a few essential parameters must be evaluated and analyzed to determine an effective RS device. A high ON/OFF ratio is required because it decreases current leakage in RS devices and ensure process optimization in the RS process. The endurance characteristic indicates how many cycles the device can interchange between resistance states (SET and RESET operation) in storing data before deteriorating. Furthermore, the retention period indicates how long the device can keep that resistance condition. Other deciding factors would include operating voltage values and range, flexibility potential (bending cycles) for wearable electronics, and temperature dependence on the active perovskite-based RS devices. Table 2 compares a few current high performance RS devices that use Pb-based and Pb-free perovskites.

According to our extensive review of the literature carried out on perovskite RS devices, Pb-based RS devices appear to have comparable overall performance to Pb-free RS devices. This is not the case with perovskite PVs, where Pb outperforms Pb-free perovskite PVs in terms of power conversion efficiencies (PCEs). The PCE values of Pb-free perovskite PVs are nowhere near those of Pb-based perovskite PVs. Even the most promising emerging Pb-free perovskite, Sn, has low PCEs when compared to Pb-based PVs. Thus, Sn becomes less appealing to researchers until high PCEs can be achieved to be on the same level as Pb-based perovskites.

Pb-free perovskites have a long way to go before they can compete with Pb-based perovskites in PVs. Despite that, this is debatable with the recent development of HP RS devices presented in this mini review because both devices perform admirably in comparison. Pb and Sn-based perovskite RS devices have roughly equivalent output properties, which initially provide good long-term prospect of either element. Nevertheless, keep in mind that the stated work on Pb-free RS devices is currently limited and may necessitate more research before making such claims. In currently reported works for a Pb-based RS device, Kang et al. created an excellent RS device ($Au/CH_3NH_3PbI_3/Au/SiO_2/Si$) which has exceptional resistive capabilities.[33] This unipolar RS device has a compliance current of 200 μA and was built in a cross-bar array design (1 × 8 cross-bar array memory cells). Their technology has a high durability of 1000 cycles with an average ON/OFF ratio of 10^8 and a lengthy retention duration of more than 10^4 s. Similarly, Cu-based perovskite RS device ($Ag/PMMA/(C_6H_5CH_2NH_3)_2CuBr_4/Pt$) was fabricated by Kim et al. that shows the potential of Pb-free perovskites.[28] Their electrochemical change metallization (ECM) method, which takes place in the RS device, results in a very high ON/OFF ratio of 10^8, a repeatability of 2000 cycles, and more than 10^3 retention characteristics. Also, the RS device was capable of multilayer storage, in which reset voltages gradually increased from lower compliance current to higher compliance current (10^1 to 10^5 A) owing to the extended filament mechanism (Figure 4).

Nonetheless, there is still room for development for Pb-free perovskites to stand out from Pb-based perovskites in RS devices. To justify the usage of Pb-free components, the resistive properties of Pb-free RS devices must maintain or even exceed those of Pb-based RS devices. Other in-depth modifications to Pb-free perovskite RS devices, such as investigating the effect of morphological structure, the addition of enhancement layers or dopants, the interaction between the top electrodes and the active layers, and the mechanism that builds the filamentary conduction of RS devices, would still need to be critically analyzed. Also, the availability of some Pb-free elements in natural resources, such as Sn (0.0002%), Bi (0.00002%), and Ag (0.000007%), is substantially lower than Pb (0.0013%). This makes it more expensive for Pb-free elements to be extracted and produced. To resolve this, the use of other Pb-free materials, such as Cu (0.0055%) and Mn (0.095%), can be sought after and explored to reduce operating costs while being less damaging to the environment. Likewise, while the usage of Pb-free perovskites may seem appealing, there are other types of RS devices that have significantly more proven data, efficiency,

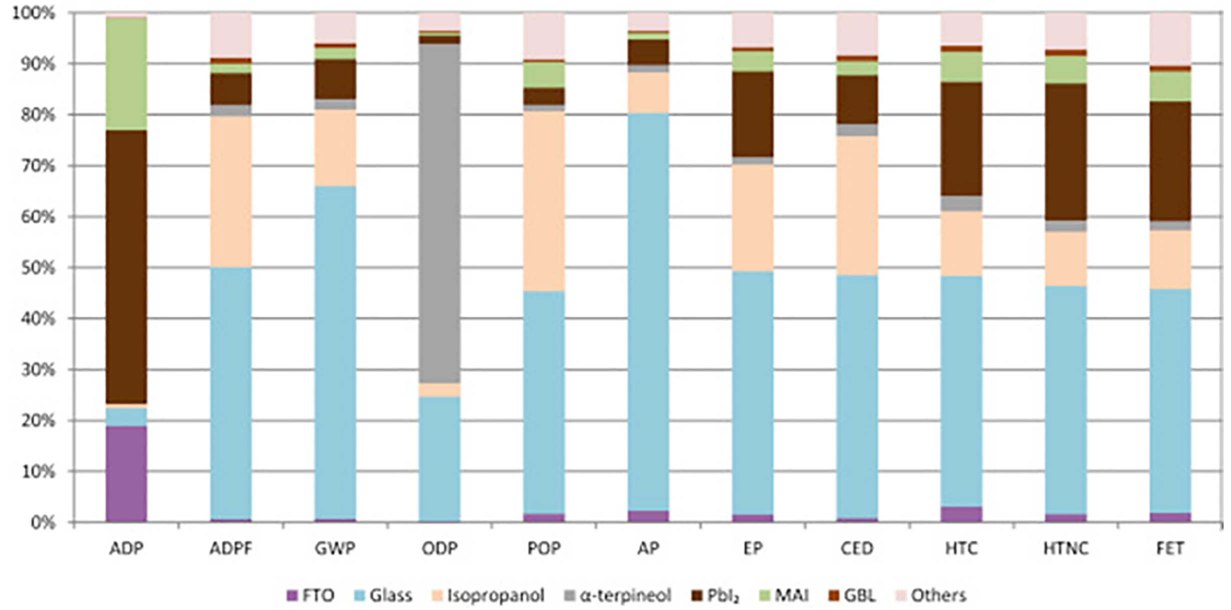

Figure 5. Material source effects of the carbon stack perovskite module, arranged by input factors. Reproduced with permission from ref 37. Copyright 2018 Elsevier.

low manufacturing costs, abundance, stability, and far less toxicity, such as inorganic materials. This reinforces the need for Pb-free perovskites to outperform competing nonperovskites RS devices, such as the popular metal oxides-based memories.[25] Overall, the memory storage capability of Pb-free perovskites still has a long way to go before it can compete in the commercialization of RS devices.

3.4. Strategies in Choosing between Pb-Based and Pb-Free Perovskites. Based on the observation of recent RS works, the resistive performance of both Pb-based and Pb-free perovskite RS devices appears to be similar, with no significant changes in resistive properties. Thus, choosing either pathways would yield similar RS results. The only difference lies in Pb-free perovskites which continue to show more environmental promise than Pb-based perovskites. Because it is more ecologically friendly, the eco-friendly strategy will be able to attract investors, manufacturers, and customers. Fundamentally, Sn and Bi have the potential to replace Pb. However, the references on Sn and Bi for Pb-free perovskites are significantly fewer than that of Pb-based perovskite RS devices.[1,14,17,19,21−23,25,26] Moreover, further variations of other Pb-free elements, such as Ge, In, Mn, and Te have yet to be investigated. These untapped Pb-free perovskites may provide more options to justify its use than Pb-based perovskites. To achieve such realization, Pb-free perovskite RS devices must be improved through its resistive characteristics (ON/OFF ratio, durability, retention, operating voltages, real-world applicability) over Pb-based perovskites. If not, they will continue to fall behind because Pb has a significantly greater RS output and is becoming more favored, particularly in PVs. Furthermore, the use of Pb-free perovskites has a significant environmental, social, and governance (ESG) impact on enterprises. The ESG criteria are a set of requirements for the behavior of a corporate that socially concerned investors use to analyze possible investments. As the world fights climate change and aims to minimize carbon footprints, the adoption of Pb-free perovskites would boost the industry's image through effective social and economic responsibility. Thus, the emphasis on good ESG responsibility can manage the organizations for long-term success.

Several review works has briefly shown the benefits of implementing Pb-free than Pb-based perovskites in RS memories.[34−36] However, vilifying the use of Pb element without an exhaustive investigation would necessitate serious rethinking. There has been no research, particularly on the negative impact of Pb-based perovskite RS devices in the manufacturing sector. Nevertheless, looking at the Life Cycle Assessment (LCA) of perovskite PVs can provide some optimizm in RS devices technology as the design structure is similar. The LCA framework is used to evaluate the environmental implications of manufacturing systems and industry actions. Several studies have found that Pb is not the primary harmful contributor in perovskite PVs. According to Alberola-Borràs' LCA analysis, the essential interest is the energy used in the synthesis and heating of the precursor solution, not the Pb component.[37] The most harmful substance is mostly glass substrate (above 43%), which accounts for the biggest proportion in Figure 5. As a result, optimizing the entire design of perovskite PVs is more essential than eliminating entirely the use of Pb.

Moreover, Pb-based RS devices are likely to provide a comparable LCA output of Pb-based PVs. Besides this, up to 99% Pb may be recycled, giving Pb-based perovskites an advantage over Pb-free perovskites. Generally, a comprehensive LCA analysis reaffirms that[38]

(i) on the basis of the tiny amount utilized, Pb or Sn has little influence on toxicity (because of the little amount utilized, neither Pb nor Sn have a significant environmental impact);

(ii) perovskite PVs are safer than other PV technologies such as GaAs, dye-sensitized solar cells (DSSCs), Si-based, CdTe, copper indium gallium selenide (CIGS), organic, and quantum dot solar cells.

This proves that there is a need for a thorough LCA study of Pb-based RS devices which should be urgently conducted. To the best of our knowledge, there is no reported LCA analysis on Pb-free RS devices. Hence, it is predicted that future LCA studies might possibly mirror the results of the LCA studies on perovskite PVs. For the meantime, researchers should address the following questions when selecting between Pb-based and Pb-free perovskites for RS devices:

(i) Would the resistive characteristics of constructed Pb-based and Pb-free perovskite RS devices differ significantly? Significant fluctuation in the operating voltage window, temperature tolerance, degradation of the perovskite layer, or filamentary conduction routes might be examples of this.

(ii) If the resistive performance of Pb-free perovskites is worse for RS devices, is the trade-off of adopting Pb-free perovskites with inferior RS performance worthwhile when compared to Pb-based perovskite RS devices? In some reports, Pb-free perovskites are reported to have worse application performance than Pb-based perovskites. If the performance drop is minor, and the long-term advantages exceed this disadvantage, then the use of Pb-free perovskites can be rationalized.

(iii) Are the chosen Pb-free perovskites stable in terms of external variables (light, heat, and moisture) or is it the same as Pb-based devices? Some Pb-free materials, such as Sn, are not stable in contrast to metal oxides or perovskite oxides. They also may be on the same performance level as Pb-based devices.

Ultimately, it is not improper to choose any path, as both types of materials have benefits and drawbacks. With the expanding number of related perovskite RS publications, the situation may be further clarified as we are still in the initial research stage. More investigations on perovskite RS devices would still need be carried out to observe how perovskite RS technology overcomes other currently positioned memory storage technology in the market.

4. CONCLUSION AND FUTURE OUTLOOKS

In conclusion, the most recent improvements of HP RS devices have been reviewed. The employment of Pb-based and Pb-free perovskites has long been a source of contention among academics and scientists. Most research data about the shortcomings of Pb-based perovskites have been brought over by existing perovskite PVs analysis to perovskite RS devices. On the other hand, the resistive characteristics of Pb-based and Pb-free perovskites appear to be identical as presented in this mini-review. This means that using Pb-based or Pb-free perovskites would produce similar results in terms of good ON/OFF ratio, endurance, and retention characteristics. Nonetheless, the number of works in this field is still minimal and would need more extensive research to further confirm this performance comparative studies. Although Pb-free perovskites appear to be a more fascinating alternative as a considerably less harmful material than Pb-based perovskites, several LCA findings indicate otherwise. Thus, the use of Pb is not entirely a bane in most applications. However, the importance of LCA studies specifically for HP RS devices should be conducted to confirm this hypothesis. With the current research progress in RS device technology, Pb-free perovskites may be expected to outperform Pb-based perovskites in the future. For the time being, Pb retains the top rank since it is easier to procure, less expensive to produce, and becoming more stable. More prospects would need to be provided through additional studies on Pb-free perovskite RS devices. This includes RS improvements, affordability, stability, toxicity, and elements that have yet to be examined in RS technology, such as Mn, Te, and In. Most importantly, both perovskites should outperform other inorganic materials in well-established experiments to justify its use in RS devices. Ultimately, both perovskites have very strong possibilities and may find their own usage in memory storage technology in the near future.

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