

Organic and perovskite memristors for neuromorphic computing

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ABSTRACT

Organic and perovskite memristors have superior characteristics both in material and structural perspectives, and therefore have been evaluated for possible integration as bio-realistic components of artificial intelligent hardware systems. This application will require the brain-inspired integrated systems that can process and memorize large amounts of complex information; requirements include highly uniform and reliable memristors that can be operated at low energy and integrated at high density. Here, we review the progress in development of organic and perovskite memristors to obtain various synaptic behaviors, with focus on material and underlying mechanism aspects. Then we address various approaches to meet the needs for constructing applications of neuromorphic computing, including low energy consumption, high uniformity and reliability of the memristors, and high-density integration. Lastly, we suggest future research directions toward realizing neuromorphic computing.

1. Introduction

Realization of artificial intelligence requires processing of vast amounts of complex information. However, when processing large amount of data, the existing von Neumann computing architecture faces limitations of speed and energy efficiency due to the von Neumann bottleneck and the end of Moore's law [1,2]. One way to solve this problem can be to emulate a biological brain [3]. A human brain is an extremely high-density neural network of $\sim 10^{12}$ neurons and $\sim 10^{15}$ synapses in a volume of ~ 2 L; it can process a huge amount of information in massively parallel and distributed manners, while consuming < 20 W [4,5]. The field of neuromorphic electronics seeks to emulate human brains at the hardware level [6]. Especially, artificial synapses have been developed to replicate biological synaptic functions and properties, because synapses contribute to learning and memory in the brain [1–4,6,7].

Among various kinds of synaptic devices, memristors as 2-terminal devices have been considered as critical components for neuromorphic computation due to their advantages of high-density integration in a cross-point array, multi-level memory, and good scalability [1,4,6,7], while they may require two-terminal selector devices to solve sneak path problem. Although they have been constructed mostly using inorganic

materials [7–9], organic materials and organic-inorganic hybrid perovskites (OIHPs) have been recently employed [1,4,6,10–12]. Organic memristors have advantages of mechanical flexibility, biocompatibility, so their applications can be expanded to wearable and implantable computing systems [1,6,13–23]. In addition, organic materials exhibit easy tunability of electrical, optical, and mechanical properties by molecular design for many purposes [4]. As for perovskites, they have attracted great interest in the neuromorphic devices, due to high on/off ratio for resistive switching, low cost, excellent optical and charge-transport properties, and mechanical flexibility. Since the first report on the perovskite memristors [12], they have been proved to be suitable for low-energy neuromorphic computation, because they demonstrate synaptic properties as a result of migration of halide ions, which have low activation energy of migration [11,24–30]. Thus, the perovskite memristors can emulate synaptic functions and properties which consume low power, so they can facilitate development of next-generation neuromorphic computing systems. However, investigation of organic and perovskite memristors has mainly been focused on demonstration of synaptic behaviors with material development and mechanism analysis [1,3,4,6,12,26,28,31–36], even though realization of brain-inspired computing requires extension of research toward the system level; i.e., in addition to complete emulation of synaptic

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functions and properties, memristors must be highly uniform and reliable, consume low energy, and be amenable to high-density integration for actual applications (Fig. 1). Despite the importance of this research direction, its direction and goals have not been sufficiently reviewed and discussed.

In this paper, we address the research progress of organic and perovskite memristors and provide guidelines for future research toward actual applications of neuromorphic computing. Firstly, we present the materials and mechanisms of organic and perovskite memristors that mimic synaptic characteristics. Secondly, we summarize investigations to demonstrate low-power memristors that resemble biological synapses, which only dissipate 1–10 fJ per synaptic event. Then we will review several approaches to improve uniformity and reliability of the memristors and to develop a high-density crossbar array; these investigations are prerequisite steps for constructing complex neural networks, and thus actual applications of them. Lastly, we suggest future research directions to realize neuromorphic computing.

2. Realization of neuromorphic computing with memristors

Hebb's rule (1949) explains the behaviors of human brains: "Cells that fire together, wire together". This means that neuronal activities from experience, thought, feeling, and physical stimuli trigger neurons to become connected in neural networks. That is, as input signals pass through synapses and neurons, the strengths (weights) of synaptic connection can change [1,4,6,37]. As a result, a complex biological neural network can be represented simply in an artificial neural network (ANN) as a set of synaptic weights; such simplified ANNs have demonstrated learning, cognition, and memorization that emulate those of biological brains. Thus, investigation has been conducted in the direction of replicating synaptic weight of synaptic function. Specifically, artificial synapses are required to possess controllable multi-state conductance (typically over 100 states), and ultimately, they need to have analog control of synaptic weight for processing huge amount of information like natural brain [1].

The ultimate goal of neuromorphic electronics is to construct spiking neural networks (SNNs), in which information is coded as voltage spikes, as in biological nervous systems [1,4,16,31,38]. Biomimetic SNNs can be operated without the need for complex electrical stimulating equipment to generate overlapping programming pulses, so the system is simplified [1,16]. To fulfil this goal, artificial synapses that have synaptic weight controlled by spikes must be developed, so artificial synapses that demonstrate various synaptic properties such as short-term plasticity (STP), long-term plasticity (LTP),

spike-rate-dependent plasticity (SRDP), spike-duration-dependent plasticity (SDDP), and spike-timing dependent plasticity (STDP), have been studied; STP and LTP respectively indicate temporary and long-range change of synaptic weight. When the synaptic strength is modulated by the signal frequency and duration time, they are called SRDP and SDDP, respectively. Additionally, synaptic strength can be affected both by the time interval and temporal order between paired spikes, which is known as the basic principle of Hebbian learning. This modification of synaptic strength by temporal relationship between two spikes is called STDP.

Therefore, we will review recent work with organic and perovskite memristors that has been conducted to realize synaptic weight and various synaptic properties. For complete emulation of synaptic functions and properties to build brain-inspired computing hardware systems, the memristors must be highly uniform and reliable, use little power, and be suitable for high-density integration.

Artificial synapses are regarded as new components for artificial intelligent system that will overcome the huge power requirement (>15 MW) of von Neumann computing hardware systems [4]. Previous investigations of artificial synapses have focused on reducing energy consumption to levels comparable to biological synapses, and to show their potential for use in future low-energy neuromorphic devices.

The devices must be integrated to implement artificial intelligent systems, so uniformity and reliability of the device operating characteristics are important factors. Solution processing such as spin-coating and ink-jet printing, of organic and perovskite materials has low cost and large scalability. However, organic and perovskite memristors produced by these processes inevitably suffer from high device-to-device deviation and low reliability during operation, and these undesirable traits impede realization of actual neuromorphic applications. Therefore, several investigators have tried to improve the uniformity and reliability of the organic and perovskite memristors to the levels of inorganic counterparts.

Lastly, high-density integration similar to those of biological nervous systems should also be well-emulated to realize brain-inspired computing electronics. The density of synapses in a human brain is $\sim 10^9 \text{ mm}^{-3}$ [1,39]. For an ANN to process large amounts of complex amounts of information, like biological counterparts, integration of artificial neurons to this density is necessary.

This introduction has demonstrated several requirements of the organic and perovskite memristors for actual applications in neuromorphic computing systems. In this chapter, we will introduce previous research to achieve synaptic functions and properties, low-power consumption, high uniformity and reliability, low power consumption, and

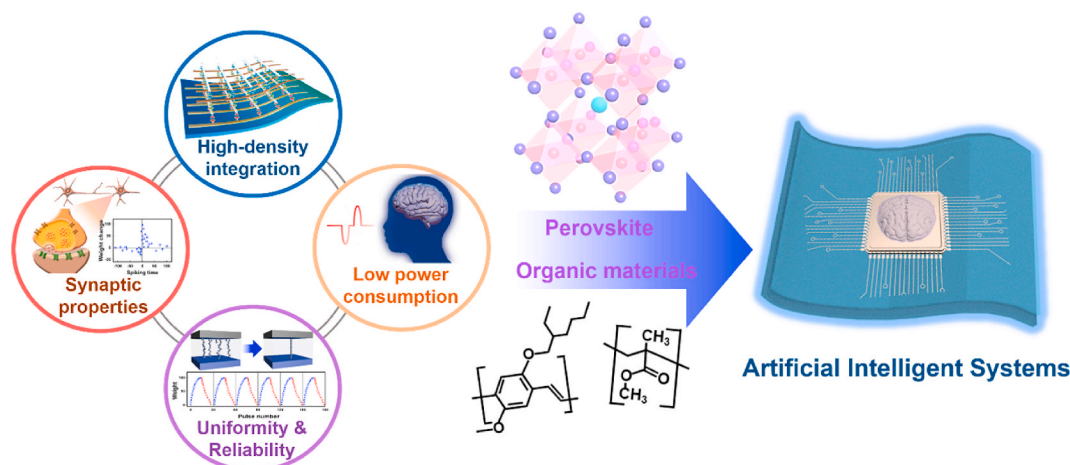


Fig. 1. Research directions of organic and perovskite memristors for next-generation artificial intelligent systems; replication of various synaptic properties [16] (Copyright 2020. American Chemical Society), high uniformity and reliability, low power consumption, and compatibility with high-density integration [71] (Copyright 2017. Springer Nature).

high-density integration.

2.1. Synaptic functions and properties

Organic-inorganic hybrid perovskite (OIHP) memristors were first introduced to emulate both the working principle and synaptic characteristics of biological synapses [12]. OIHP memristors were fabricated in bottom electrode/buffer-capped conducting polymer (BCCP)/OIHP/top electrode structure (Fig. 2a). When an electrical pulse is applied, ions migrate in the OIHP layer and are injected into the BCCP film, so the film conductance changes temporarily and persistently, respectively. As a result, the device can show multilevel memory, and various synaptic properties such as STP, LTP, paired-pulse facilitation (PPF), and STDP [12]. Many subsequent investigations have considered perovskite memristors that used various perovskite materials, e.g., lead-free [27,40,41], two-dimensional [26,28,42,43], and single-crystal [24,27,28] perovskites.

The synaptic behaviors of the OIHP artificial synapses are affected by their structural dimensionality [26]. As it decreases from 3D to quasi-2D and 2D, and thus increases the quantity of organic insulating phenethylammonium (PEA) cations, activation energy for halide ion migration in the perovskite films increases [26]. As a result, in quasi-2D OIHP artificial synapses, the bulky PEA cations block back-diffusion of migrated anions after the voltage is removed, so these devices retain information longer than 3D OIHP devices. The 2D perovskite synapse that had the largest amount of insulating PEA cations had the smallest magnitude of postsynaptic current, and thus the lowest energy consumption of ~ 0.7 fJ per synaptic event, which is comparable to that of biological synapses (Table 1) [26]. Perovskite memristors have been mainly reported to operate by exploiting ion migration, but several studies have exploited growth of metallic filaments by using active metals such as Ag and Cu [10,40].

Organic memristors demonstrate synaptic behaviors by various mechanisms, including charge trapping [44–49], metallic-filament formation [15,16,18,19,21,23,50–62], ferroelectricity [63,64], and phase change [65]. In organic memristors, research has mostly focused on finding various materials and underlying mechanisms that can have synaptic properties (Table 1). Thus, a wide range of organic semiconducting and insulating materials [49,58,62,66,67] such as poly[2-methoxy-5-(2-ethylhexyloxy)-1,4-phenylenevinylene] (MEH-PPV) [61,68], poly(methyl methacrylate) (PMMA) [16,18,21,23], and poly(vinylidene fluoride-co-trifluoroethylene) (PVDF-TrFE) [63,64] have been adopted in plain forms. Additionally, biomaterials extracted from

nature have shown synaptic behaviors [51,55,60]; examples include glucose [69], carrageenan [50,53,56,57], collagen [70], and keratin [59].

To reduce the operating voltage range in the memristors, organic materials have been mixed with inorganic materials; i.e., organic materials as matrix, and inorganic materials as either charge-trapping parts to increase charge trapping [44–47] or ion reservoirs to facilitate ion migration [50,51,53,60,71]. These mixed-material organic memristors could demonstrate low energy dissipation. For example, Ag-doped carrageenan showed lower power consumption of $0.35 \mu\text{W}$ than bare carrageenan ($\sim 17.5 \mu\text{W}$) [50,56].

Organic memristors can be classified as diffusive and drift types. Diffusive memristors mimic STP, and drift memristors mimic LTP. The dynamics of the metallic filament (MF) in a PMMA polymer medium is affected by the polymer's molecular weight (free volume distribution), which affects MF diffusion [16,21]. This result can be exploited to consolidate diffusive and drift memristors in a single device by adjusting the molecular weight of the polymer (Fig. 2b) [16]. This artificial synapse with bio-realistic synaptic plasticity showed learning functions of SRDP and STDP in response to the electric spikes, like the biological counterparts.

2.2. Low power consumption

The energy consumption of a memristor depends on materials, operating mechanism, and device dimensions [1,4]. OIHPs have potential uses as low-energy neuromorphic devices, because they mainly exploit ion migration, which is a low-energy mechanism, and the migrating ions are halides, which have inherently low activation energy for ion migration (e.g., $\text{Br}^- \sim 0.2$ eV and $\text{I}^- \sim 0.6$ eV) [12,25,26,28,29,72], but effort has been devoted to further reducing energy consumption. In 3D polycrystalline thin films, ions are assumed to move along grain boundaries; this motion causes high leakage current, which increases energy consumption (Fig. 3a) [26,28]. To decrease this source of leakage current, two directions of investigation have been tested [30]. One is to reduce the number of grain boundaries by fabricating single crystals [24,27,28]; the other is to adopt low-dimensional (2D and quasi-2D) perovskites that use insulating bulky (e.g., PEA) cations to efficiently suppress migration of halide ions [24,26,28]. The investigation taking both research directions was reported (Fig. 3b); the memristor with 2D single-crystalline perovskite dissipated 400 fJ/spike synaptic event [28].

Organic memristors composed of conjugated polyelectrolyte

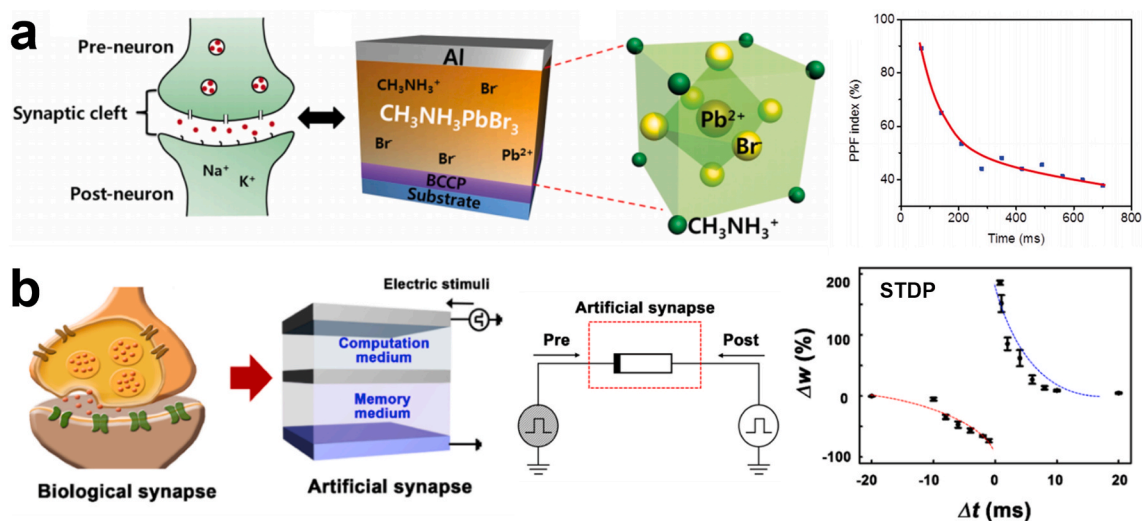


Fig. 2. Emulation of synaptic behaviors of (a) perovskite [12] (Copyright 2016. John Wiley & Sons, Inc.) and (b) organic memristors [16] (Copyright 2020. American Chemical Society). Schematics of biological synapses (left), device structures (middle), and curves of resulting synaptic properties (right) (a: PPF, b: STDP).

Table 1
Summary of organic and perovskite memristors.

Mechanism	Material			Power [W] consumption	Multistates	PPF	SRDP	STDP	P/D	Application	Ref.
	Substrate	Electrodes	Active								
Metallic filament	PEN ^{a)}	ITO/Ag	PMMA	$\sim 8 \times 10^{-8}$	0	0	0	0	–	–	[16]
Metallic filament	PEN	ITO/Ag	PMMA	$\sim 7 \times 10^{-6}$	–	–	–	–	–	–	[21]
Metallic filament	SiO ₂	Pt/Ag	Protein	$\sim 6 \times 10^{-10}$	0	–	0	–	–	–	[55]
Metallic filament	SiO ₂	Pt/Ag	Carrageenan	$\sim 2 \times 10^{-5}$	0	0	–	–	–	–	[56]
Metallic filament	PES ^{b)}	Al/Cu	pV3D3 ^{c)}	$\sim 5 \times 10^{-4}$	0	0	–	0	0	Face classification	[58]
Metallic filament	SiO ₂	Al/rGO ^{d)}	MEH-PPV	$\sim 2 \times 10^{-4}$	0	–	–	–	–	Pressure sensory and recognition memory	[61]
Metallic filament	Si/SiO ₂	Ag/Pt	(Cs ₃ Bi ₂ I ₉) _{0.4} -(CsPbI ₃) _{0.6}	$\sim 2 \times 10^{-7}$	0	–	–	0	0	–	[86]
Charge trapping	PES	Cu	Pentacene/PVN ^{e)}	$\sim 3 \times 10^{-3}$	0	0	–	0	0	–	[49]
Ferroelectricity	Ni	Ni/Cu	P(VDF-TrFE)	–	0	–	–	0	–	–	[63]
Ferroelectricity	Glass	ITO/Au	CuPc ^{f)} /P(VDF-TrFE)/PFO ^{g)}	$\sim 2 \times 10^{-5}$	0	–	–	–	0	–	[64]
Redox reaction	SiO ₂	Ag/Ta	PEDOT:PSS ^{h)}	$\sim 7 \times 10^{-6}$	0	–	0	0	0	Learning-forgetting-relearning	[62]
Redox reaction	PET ⁱ⁾	Ta/Pt	EV(ClO ₄) ₂ ^{j)} /TPA-PI ^{k)}	$\sim 3 \times 10^{-6}$	0	0	0	0	0	Learning-forgetting-relearning	[66]
Ion migration	SiO ₂	Au/ITO	Nafion	$\sim 7 \times 10^{-10}$	0	0	–	0	0	Pressure sensory /recognition and learning	[67]
Ion migration	PET	ITO/Mg	Collagen	$\sim 3 \times 10^{-5}$	0	0	0	0	0	–	[70]
Ion migration	Si	Si/Al	Conjugated polyelectrolytes	$\sim 2 \times 10^{-14}$	0	0	0	–	–	–	[32]
Ion migration	Glass	Au	Single-crystalline CH ₃ NH ₃ PbBr ₃	$\sim 6 \times 10^{-10}$	0	0	0	–	0	Nociceptor	[24]
Ion migration	SiO ₂ /quartz	Au	Single-crystalline Cs ₃ Sb ₂ Br ₉	$\sim 3 \times 10^{-6}$	0	–	0	–	–	Letter recognition	[27]
Ion migration	Glass	ITO/Al	CH ₃ NH ₃ PbBr ₃	$\sim 1 \times 10^{-12}$	0	0	0	0	–	–	[12]
Ion migration	Glass	ITO/Al	(PEA) ₂ PbBr ₄	$\sim 4 \times 10^{-14}$	0	0	0	–	0	Pattern recognition	[26]

^{a)} Poly (ethylene maphthalate).

^{b)} Poly (ether sulfone).

^{c)} Poly (1,3,5-trivinyl-1,3,5-trimethyl cyclotrisiloxane).

^{d)} Reduced graphene oxide.

^{e)} Poly (2-vinyl naphthalene).

^{f)} Copper phthalocyanine.

^{g)} Poly (9,9-dioctylfluorene).

^{h)} Poly (3,4-ethylenedioxythiophene):poly (styrenesulphonate).

ⁱ⁾ Poly (ethylene terephthalate).

^{j)} Ethyl viologen diperchlorate.

^{k)} Triphenylamine-based polyimide.

artificial synapses have shown synaptic properties down to 10 mV, and thus enable ultralow energy consumption of ~ 20 fJ per synaptic event, similar to biological energy consumption (Fig. 3c). The ultralow energy consumption was enabled by a combination of two working mechanisms: dipole reorientation and ion migration [32]. Recently, protein nanowire memristors that operate at the biological voltages of 40–100 mV have been developed; the nanowires facilitated Ag⁺ reduction, which accelerated MF formation in the memristors, so various synaptic characteristics could be demonstrated even at voltages of 40–100 mV that are used in biological systems [55].

2.3. High uniformity and reliability

To realize practical neural networks, the synaptic device arrays must be highly uniform and operate reliably. Specifically, high accuracy of ANNs requires reproducible conductance changes in memristors, and low variation in operating voltage among the cells in the array [9]. In organic memristors that use the MF growth mechanism, the MFs grow randomly, so the reliability and the device-to-device uniformity are inherently restricted [73,74]. Generally, MF formation in the organic matrix entails two successive steps: ion injection at an interface between

an active metal and a polymer, then ion migration under an electric field [75,76]. The reliability and uniformity of the organic memristors can be increased by i) localizing the ion injection at the interface between active metal and organic layers, ii) confining the ion transport in the organic matrix, or iii) regulating the electric field to restrict MF overgrowth.

To localize the ion injection at the active metal and the polymer interface, the interfacial area between the active metal and the polymer insulator should be constrained [75,76]. In inorganic systems, photolithography and lift-off processes have been widely used to pattern these interfacial regions to localize the ion injection [77,78]. However, for fabrication of organic memristors, these processes cannot be used because they do not meet the requirement for solvent orthogonality.

As a method to localize ion injection in an organic memristor, active metal islands were formed on an inert electrode (Au and indium-tin-oxide) film by Ostwald ripening, instead of fabricating the whole active electrode (Ag) film (Fig. 4a) [18]. The device with the metal islands grew a limited number of MFs, so the resulting devices had improved uniformity and reliability compared to the typical organic memristor.

Conventionally, to confine ion transport in an organic medium, the

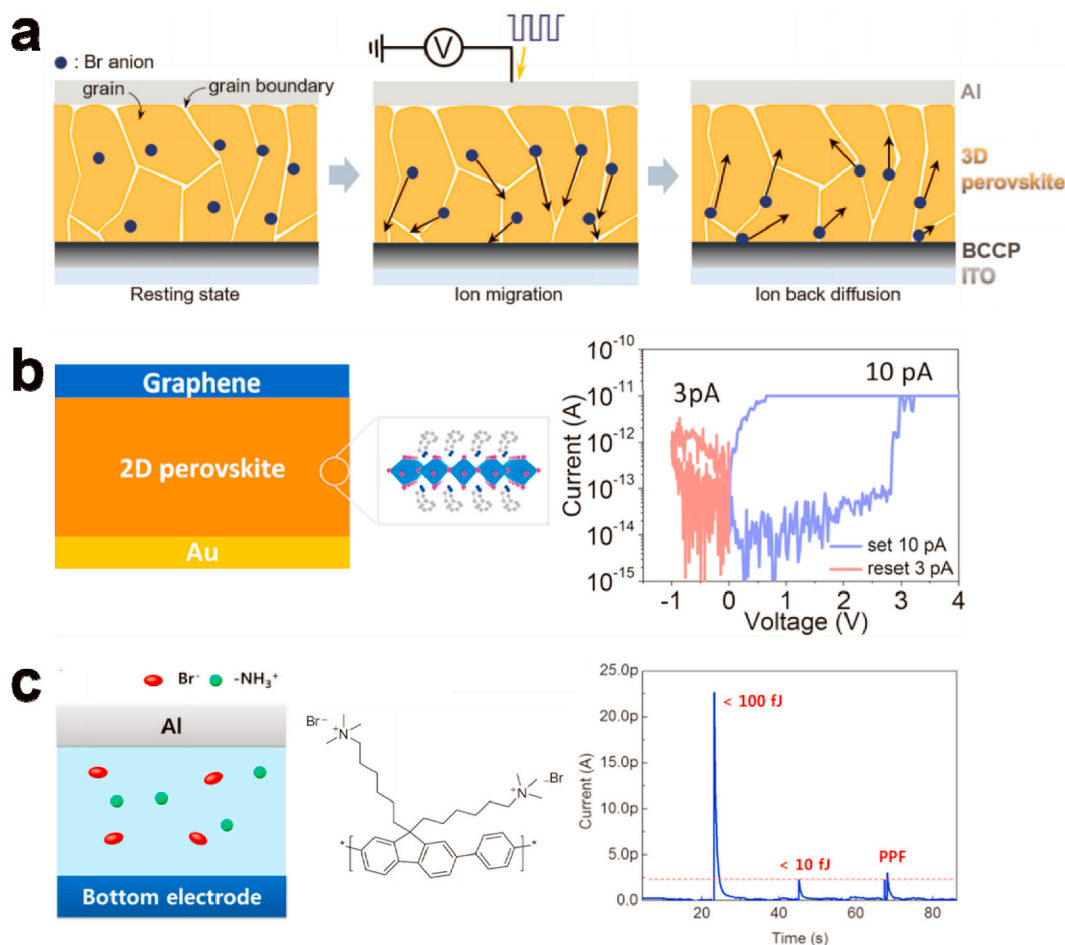


Fig. 3. Low energy consumption of perovskite and organic memristors. (a) Schematic of halide ion migration along grain boundaries that causes high leakage current, and thus increases energy consumption [26]. Copyright 2019. John Wiley & Sons, Inc. Schematics of device structures, chemical structures, and curves demonstrating low energy consumption of (b) perovskite [28] (Copyright 2017. American Chemical Society) and (c) organic memristors [32] (Copyright 2018. Elsevier B.V.).

local electric field was strengthened by using protruding cone-shaped electrode contacts (Fig. 4b) [53,61,79,80]; they facilitate MF growth by shortening of the ion migration path and strengthening the local electric field (violet arrows in Fig. 4b).

Other approaches predefine the localized ion-migration path in the organic matrix [15,19]. Ion transport can be selectively suppressed according to the distribution of the free volumes in the polymer, so MF growth can be confined to follow the ion migration path which was defined by photocrosslinking [15] or molecular alignment [19]. Cross-linkable poly (vinyl cinnamate) polymer as an organic matrix predefined the localized ion-migration path (LIP), then after formation of the first MF in the crosslinkable polymer film during electroforming process, the film was crosslinked by UV exposure (Fig. 4c) [15]. In the crosslinked region, the free volumes were highly shrunken, so growth of MFs impeded: the ion-migration path was effectively defined along the first MF growth path in the crosslinkable polymer, so further MF growth occurred along this path (the violet arrow in Fig. 4c). Organic memristors obtained using this method had reliable responses for use as an artificial synapse in neural networks [15].

Ion migration can also be controlled by aligning the polymer (Fig. 4d) [19]; ion migration follows the aligned molecules (the violet arrow in Fig. 4d). A liquid-crystal polymer (LCP) was used as organic medium, and the molecular alignment of LCP was controlled by a mechanical rubbing process. In a memristor that used non-aligned LCP, MF growth was stochastic, as in typical MF memristors, whereas in the device that used aligned LCP, MF formation followed the free-volume

distribution, which was determined by alignment of LCP molecules [19].

In organic memristors, random overgrowth of CFs also causes the switching variation. Initially, a compliance current during the writing process has been used to regulate the electric field and the resultant CF growth [81,82], but when this method was used, the devices stored charges, so the overgrowth of CFs was not effectively suppressed. For practical applications that require reliable operation, the electric field can be regulated by serial connection of an external load resistor [83]. Furthermore, an organic memristor that had self-compliance current was developed by inserting an interfacial load polymer layer in the single memristor (Fig. 4e) [23]; the polymer serves as an internal resistor which regulates the electric field, and as a result, MFs form uniformly and reliably.

Despite these approaches, use of organic memristors in neural networks remains difficult. Ultimately, to realize complex neuromorphic systems for use in artificial intelligence, the organic memristor should be switched by a single MF, and a method to reliably control the MF in the device should be developed. Development of practical applications of perovskite memristors requires that they show uniform and reliable operation, but the device-to-device uniformity and reliability have not been analyzed yet. These studies must be conducted.

2.4. High-density integration

To increase integration density to match that of a 3D complex biological neural networks (synapse density in human cortex $\sim 10^9 \text{ mm}^{-3}$),

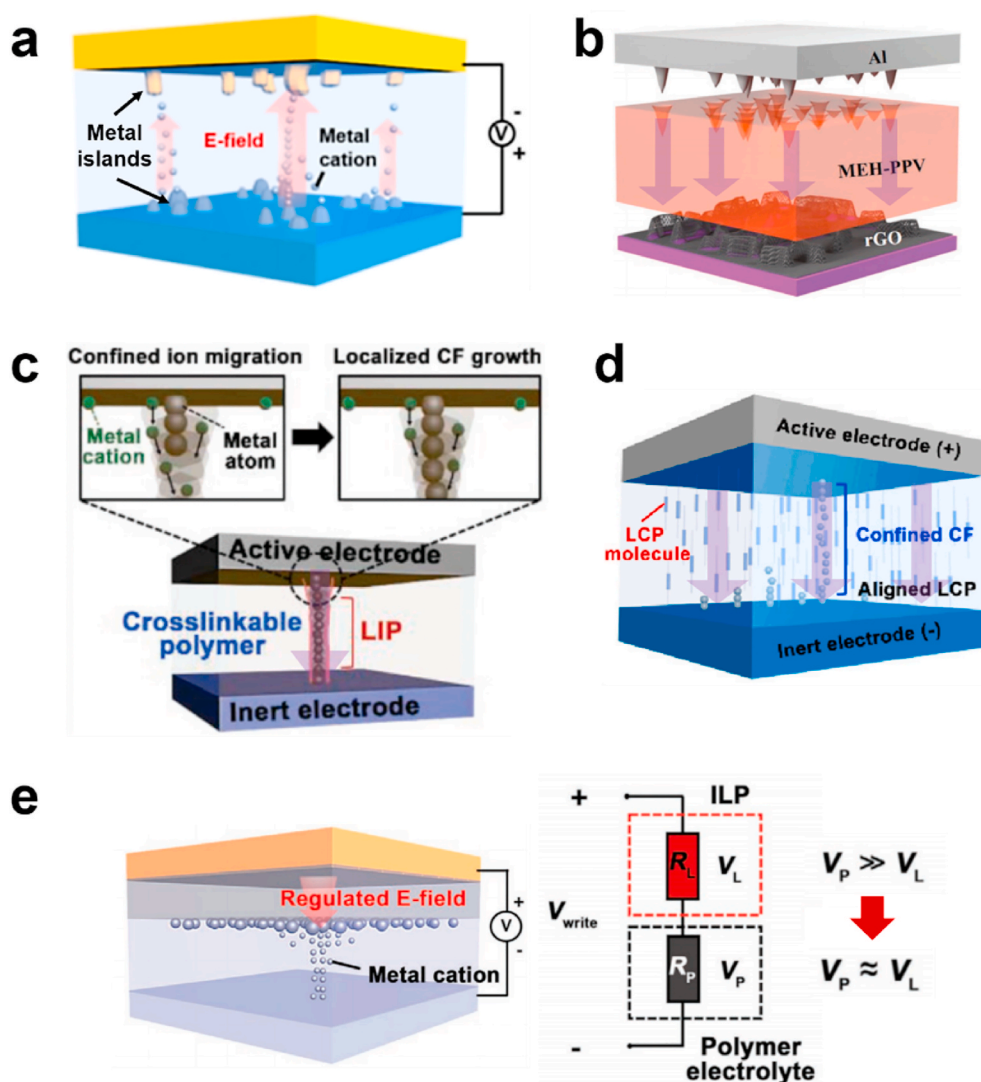


Fig. 4. Improvement of the reliability and uniformity of the organic memristors by (a) localizing ion injection at the interface between active metal and organic layers [18] (Copyright 2019, American Chemical Society), (b–d) confining the ion transport in the organic medium (violet arrows indicate ion migration path in the memristors), and (e) regulating electric field to inhibit MF overgrowth. b) Reproduced with permission [61]. Copyright 2020, Wiley-VCH. c) Reproduced with permission [15]. Copyright 2020, Royal Society of Chemistry. d) Reproduced with permission [19]. Copyright 2020, Elsevier B.V. e) Reproduced with permission [23]. Copyright 2020, Wiley-VCH. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

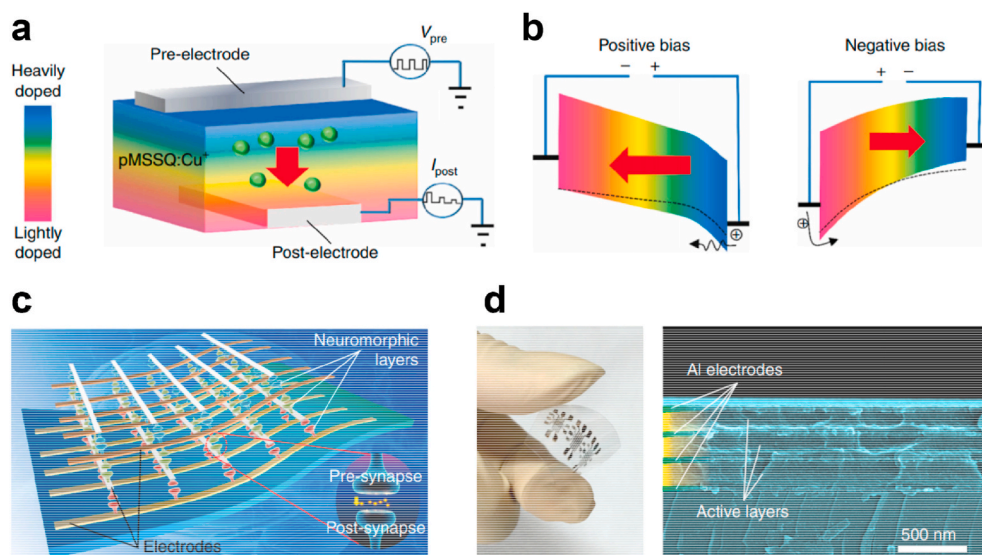


Fig. 5. High-density integration of the organic memristor arrays. Schematic illustration of (a) device structure, (b) operating principles for rectifying property, and (c) 3D integrated array by using organic memristors. (d) Optical (left) and scanning electron microscope (right) images of the 3D organic memristor array [71]. Copyright 2017, Springer Nature.

3D integration of the synaptic devices may be a favorable approach. Flexible organic memristors have been 3D integrated (Fig. 5) [71]. Memristors have a simple fabrication processes and device structure, so they are compatible with use in crossbar arrays [7,71]. Operation of such arrays to achieve accurate resistance programming and reading requires suppression of sneak path current causing crosstalk between adjacent component memristor cells. The insertion of a rectifying characteristic into the artificial synapses is beneficial for constructing the array. The rectifying property was realized by using asymmetric doping concentration in the active layer of poly (methylsilsequioxane (pMSSQ)). The device structure was Al (bottom electrode)/lightly-doped active layer/highly-doped active layer/Al (top electrode) (Fig. 5a). Doping pMSSQ with Cu^+ reduced hole-injection barrier between pMSSQ and the Al electrodes. That is, at the heavily-doped interface (the top electrode part), the hole injection barrier was lower between pMSSQ and the Al electrode than at the lightly-doped bottom electrode (Fig. 5b). As a result, with positive bias at the top electrode (forward bias), the current level was relatively high, whereas with negative bias at the top electrode (reverse bias), current level became very low. By using the memristors, flexible 3D artificial synapse networks were developed (Fig. 5c, d). This 3D integrated system showed good potential toward high-density information storage and information processing, as achieved by biological neural networks. This research may provide a new structure for construction of highly-integrated biomimetic neuromorphic systems.

To demonstrate high-density nanoscale perovskite memristors, 16×16 cross-point array was fabricated by using sequential vapor deposition with nanotemplates [84]. Sequential vapor deposition was exploited to deposit uniform and large-scale resistive switching layer of perovskites. However, this cross-point array suffered from crosstalk problem causing from sneak current during reading operation. Thus, external diodes are added individually to each unipolar-type perovskite memristor in an array, to reduce this cross-talk interference [85]. In addition to diodes, selectors can be integrated to suppress sneak current paths.

3. Conclusions

We have reviewed recent progress in development of organic and perovskite memristors which have superior characteristics both in material and structural perspectives. Thus, investigation of organic and perovskite memristors has mainly been focused on with material development to achieve synaptic behaviors, and on determining the operating mechanisms. Neuromorphic computing requires integration of components into neural networks as well as the achievement of simple synaptic functions and properties in a single device, so we also covered attempts to satisfy requirements for construction of actual applications of neuromorphic computing. These requirements include development of highly-uniform and reliable memristors that consume low power, and that are compatible with high-density integration. However, these kinds of studies are still lacking, so this is an open field for further research.

Besides, even though organic and perovskite memristors are less chemically stable than inorganic memristors in ambient conditions, but investigation to improve materials and the resultant device stability has rarely been conducted. In the perovskite field, a dual phase perovskite (e.g. $\text{AgBi}_2\text{I}_7\text{-Cs}_3\text{Bi}_2\text{I}_9$) was adopted for structural and mechanical stability [40,86]. In addition to this, various approaches to increase the chemical stability of organic and perovskite memristors should be conducted for actual applications. Ultimately, to adopt online learning in the artificial neural networks consisting of the memristors, high endurance memristors should be developed to tolerate at least hundreds of thousands of learning processes.

Together with investigations constructing artificial central nervous systems covered so far, artificial peripheral nervous systems [31,35] can also be developed with the organic and perovskite memristors. Previously, the memristors have been focused on long-term retention time (LTP), but due to the rapid progress of the memristors with various

materials and underlying mechanisms, the memristors are able to demonstrate various synaptic decay time scales [32], like biological counterparts. Thus, these can fulfil various requisites in components of artificial nervous systems that can detect, process, and memorize large amount of complex information and respond to them, as biological neural networks do. These research trials might find new ways to build highly integrated actual neuromorphic systems that emulate natural forms.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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