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Recent Advance of Flexible Organic Memory Device

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ABSTRACT

With the recent emergence of foldable electronic devices, interest in flexible organic memory is significantly growing. There are three types of flexible organic memory that have been researched so far: floating-gate (FG) memory, ferroelectric field-effect-transistor (FeFET) memory, and resistive memory. Herein, performance parameters and operation mechanisms of each type of memory device are introduced, along with a brief summarization of recent research progress in flexible organic memory.

KEY WORDS

Ferroelectric, flexible, floating gate, memory, organic, resistive.

1. INTRODUCTION

Memory devices are now ubiquitous and inseparable from everyday life. From smartphones to computers, it is now impossible to find an electronic equipment without memory device. Since the advent of the first computer in 1946, the semiconductor industry has been centered on memory device, and significant developments in the field of memory device have been the driving force behind the development of modern IT industry [1], [2].

Memory device can be defined as an electronic device that stores data. By changing the electrical state between “programming” and “erasing” processes, memory device stores data as "0" and "1". Generally, there are two ways in which memory stores data: by trapping a charge carrier or by changing the conductivity of a switching layer [3], The former method changes the current of the channel layer by trapping charge carriers in a specific layer, while the latter changes the resistance of the insulating layer via structural changes in molecular arrangement or metal filaments formation.

Currently, the memory device market is dominated by the silicon-based floating-gate type, which is based on metal-oxide-semiconductor field-effect-transistor (MOSFET). Due to high electrical performance and the existence of an excellent passivation layer, silicon-based memory devices have been advancing with exponential growth in storage capacity [4], [5]. However, due to the recent introduction of foldable electronic devices and the limitations of MOSFET-based memory, based on rigid, brittle, and inorganic crystalline materials, flexible memory devices have successfully attracted a great deal of attention [6], [7].

To implement flexible electronic devices, each component of the device, semiconductor, dielectric, conductor, and substrate, should be mechanically flexible and processed at a low temperature.[8] Since the invention of conducting polymer in 1977, research has been focused on the use of organic small molecules or polymers as electrical components of semiconductor devices[9]. For instance, much effort has been made in the development of organic/polymeric semiconducting materials, and they achieved electrical performance (carrier mobility) comparable to or surpassing that of their inorganic counterparts [10]-[12]. Similarly, various materials including polymers, inorganics, or organic/inorganic hybrids have been employed as flexible dielectric materials for electronic devices [13]-[15]. For substrates, polymers such as polyethylene naphthalate (PEN), polyethylene terephthalate (PET), and

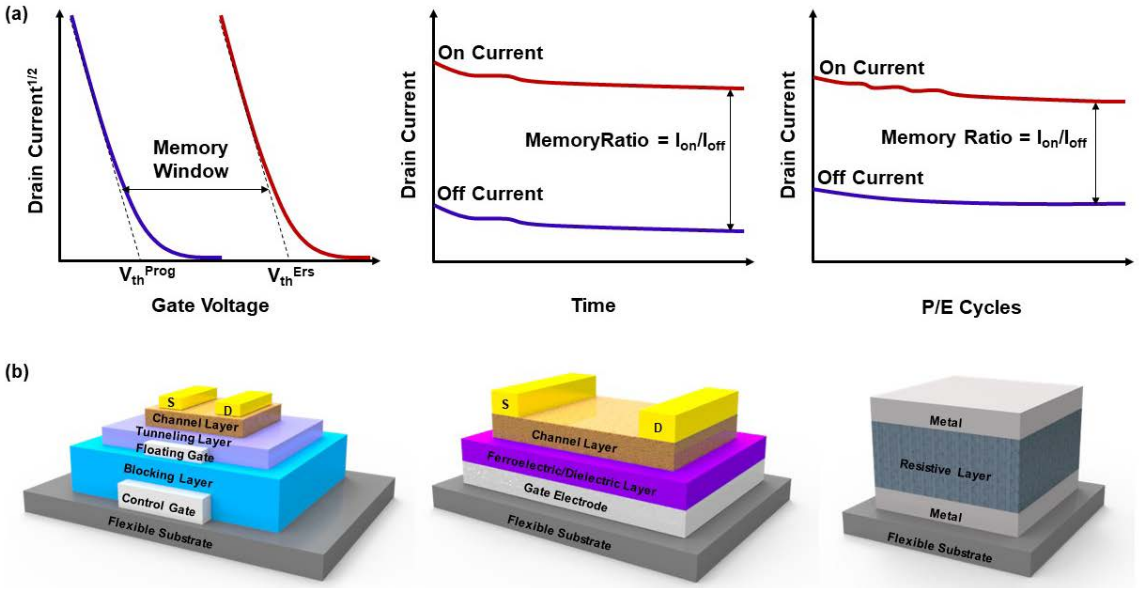

Figure 1. (a) Schematic graphs of memory performance: memory window (left), retention time (center), endurance (right), and memory ratio (center, right). (b) Schematic diagram of flexible organic memory: floating-gate memory (left), ferroelectric field-effect-transistor memory (center), resistive memory(right).

polyimide (PI), or thin metal foils have been used for the fabrication of flexible electronics [16]-[19].

In this review, recent research efforts on the development of various types of flexible organic memory device are summarized. First, representative figures of merits for flexible memory devices are presented. The paper is then divided into three types of memory devices based on the data storage mechanism: i) floating-gate (FG) memory, ii) ferroelectric field-effect-transistor (FeFET) memory, and iii) resistive memory. Data storage mechanisms are briefly reviewed for each memory type, and the materials, device structure, and performance parameters for each flexible memory device are presented.

2. PERFORMANCE PARAMETERS

In order for flexible memory devices to achieve high performance, they must exhibit high memory capacity, good reliability, and flexibility. To evaluate the performance of memory devices, a few parameters, including memory window, retention time, endurance, etc. are characterized (Figure 1(a)).

Memory window is defined as the difference of the threshold voltage (V_{th}) between programmed and erased state, showing memory capacity (Figure 1(a)). Therefore, the larger the memory window is (hence, the more distinct between programmed and erased states), the better the memory device is.

The reliability of a memory device can be characterized in terms of retention time, endurance, and memory ratio. Retention time and endurance

Table 1. Memory performance parameters for various organic flexible memory devices

Year	Memory Type	Substrate	Memory Window (V)	Retention Time (s)	P/E Cycle	Operating Volatge (│V│)	Ref
2011	Floating gate	PEN	2.5	10^5	10000	5	[24]
2012		PES	10.0	10^5	1300	70	[25]
2015		PET	32.0	-	700	70	[26]
2017		Mylar	~ 5.0	10^5	1000	~ 10	[27]
2017	Ferroelectric field-effect transistor	PEN	9.6	10^4	150	10	[31]
2018		PEN	17.2	10^4	600	15	[32]
2019		PI	38.0	10^4	200	40	[33]
2019		-	5.6	5×10^4	400	5	[34]
2016	Resistive	PET	-	3×10^3	200	1.6	[37]
2017		PET	-	10^4	100	4	[38]
2017		PET	-	10^4	-	4	[39]
2018		PET	-	10^8	400	> 1	[40]
2019		PET	-	10^4	-	> 3	[41]

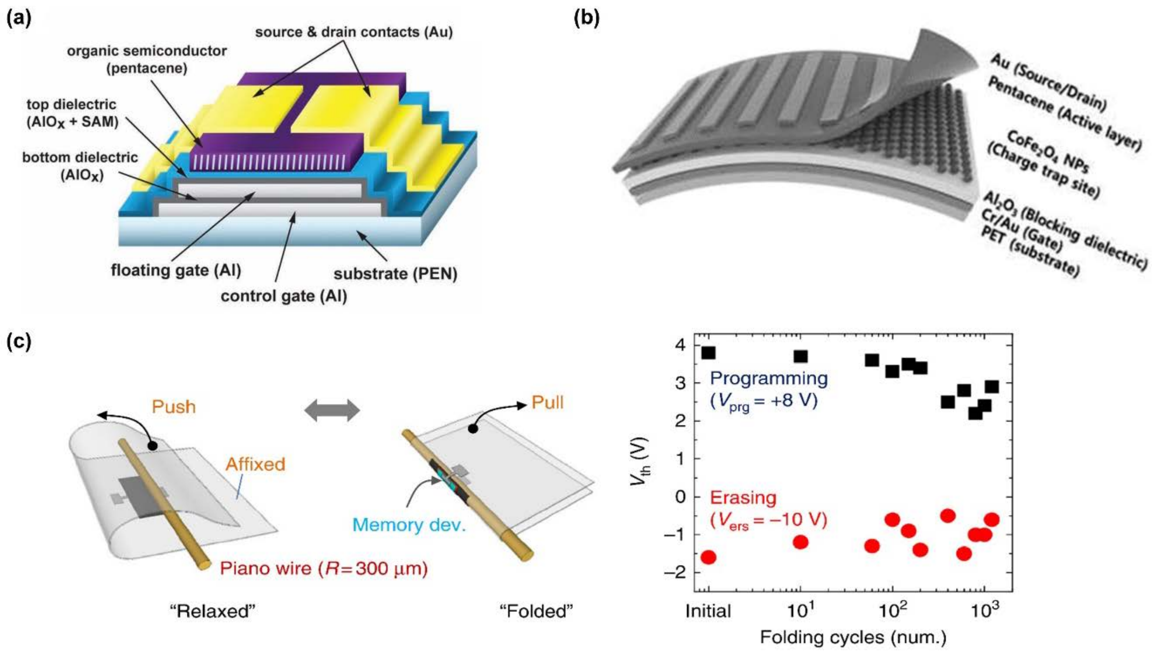

Figure 2. (a) Schematic diagram of the non-volatile organic memory on flexible substrates (PEN) with a potentiostatically anodized aluminum oxide dielectrics. A n-octyltrichlorosilane self-assembled-monolayer (SAM) forms the interface to the pentacene organic semiconductor. (a) Reproduced with permission.[24] Copyright 2011, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (b) A schematic image of the flexible nano-floating-gate memory device based on 8 nm $CoFe_2O_4$ nanoparticles on PET substrate. (b) Reproduced with permission.[26] Copyright 2015, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (c) A schematic diagram showing the method for alternating between "relaxed" and "folded" states for measurement of memory characteristics after repeated folding cycles (left). Threshold voltage of the memory on ultrathin Mylar substrates measured upon programming/erasing operations after repeated folding cycles at 300 μm bending radius vs. the number of folding cycles (right). (c) Reproduced with permission.[27] Copyright 2017, Springer Nature.

show how well memory device retains its data state over time and under repeated programming/erasing (P/E) cycles, respectively. For the commercialization of organic memory devices, more than 10 years of retention time and 10^6 times of measured endurance with minimal degradation are required [20]. Memory ratio is defined as the ratio of drain-on current and drain-off current with certain bias stress time or cycle number. Therefore, the memory ratio is determined between on- and off- current after long retention time or multiple P/E cycles, as shown in Figure 1(a).

Flexible memory requires an ability to be rolled up or bent without data loss. Therefore, in order to demonstrate the flexibility of a memory device, the reliability characteristics of the device, after bending a certain number of times on the bending axis with a specific radius, should be shown.

3. FLOATING-GATE MEMORY

Among various memory devices, floating-gate structure is the best known and most often utilized due to it being the most developed structure and well-established data storage mechanism. A floating-gate type memory device is generally composed of a control gate, blocking layer, floating gate, tunneling layer, and active layer (Figure 1(b)). Although the structure of floating-gate memory is similar to that of a field-effect-transistor, there is a floating gate and tunneling layer for storing data in the memory device. The tunneling layer makes it possible to exchange charge carriers between floating gate and active layer by Fowler-Nordheim tunneling mechanism [21]. In terms of charge storage position (electret layer or nanoparticles), floating-gate memory can be categorized as electret memory or nano-floating-gate memory (NGFM) [22], [23].

Kaltenbrunner et al. reported flexible, low-voltage organic non-volatile memory device (Figure 2a) [24]. The memory device is comprised of anodized aluminum oxide as dielectric, and pentacene as organic semiconductor. The fabricated memory device on PEN substrate operated with low programming/erasing voltage of -5 V and 5 V with long retention time of > 10^5 s.

Rani et al. employed a multilayer of reduced graphene oxide (rGO) and 3-aminopropyltriethoxysilane (APTES) as the charge trapping layer for flexible organic memory [25]. The device was fabricated based on PES as a substrate, crosslinked polyvinylpyrrolidone (PVP) as blocking and tunneling dielectric, APTES/rGO as a charge trapping layer, and pentacene as an organic semiconductor. The resulting organic memory device showed reliable memory operations with long retention time of 10^5 s and minimal loss of charge storage capability.

Jung et al. employed cobalt ferrite ($CoFe_2O_4$) nanoparticles as charge trap sites for the fabrication of

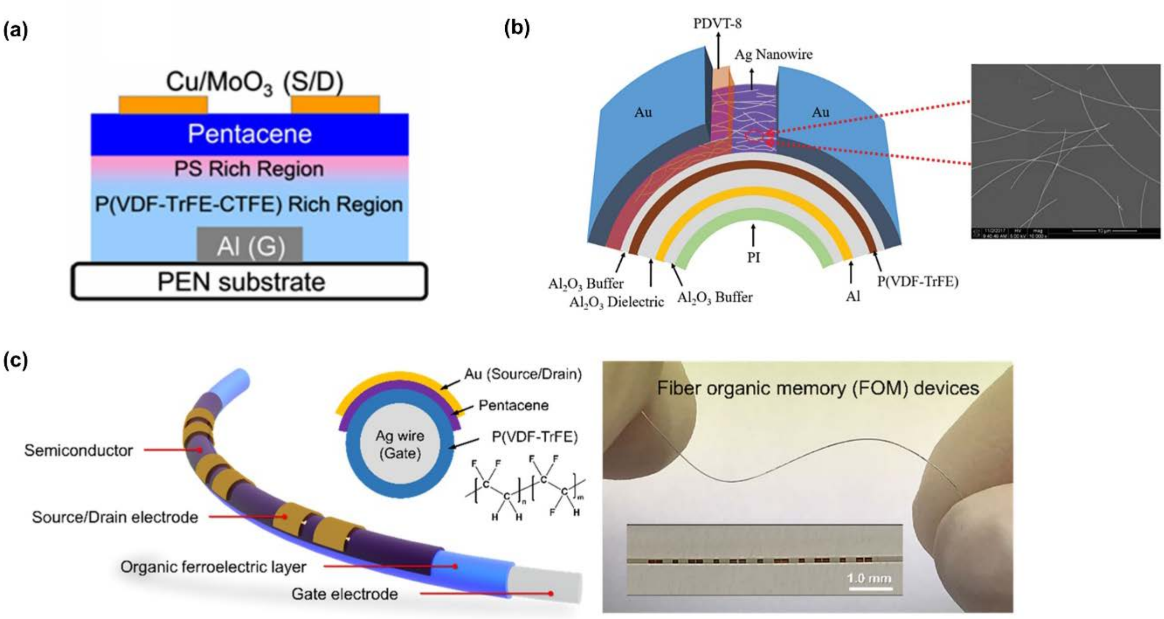

Figure 3. (a) The schematic structure of the flexible FeFET nonvolatile memory in a bending state with a curvature radius of 5.5 mm. (a) Reproduced with permission.[31] Copyright 2017, AIP Publishing. (b) Schematic diagram of the device architecture of an ultra-short channel FeFET non-volatile memory device and an SEM image of Ag nanowires, which shows that the silver nanowires are randomly stacked together to form a network structure, and the size of the grid is about 3–10 mm. (b) Reproduced with permission.[33] Copyright 2019, The Royal Society of Chemistry. (c) Schematic image of the device architecture of the fiber organic memory (FOM) (left). Cross-sectional FOM structure indicating the major materials and optical photograph of a flexible FOM device array (right). Thicknesses of each layer: P(VDF-TrFE), 260 nm; pentacene, 50 nm; Au electrodes, 50 nm. (c) Reproduced with permission.[34] Copyright 2019, American Chemical Society.

NGFM devices, using pentacene as p-type semiconductor (Figure 2(b)) [26]. The effects of cobalt ferrite size, in terms of energy level and particle-particle interaction, were investigated. The optimized memory device retained a memory window of 32 V after 700 P/E cycles and 26 V after 500 bending cycles. Especially, alternative tunneling dielectric layer (oleate) surrounding the cobalt ferrite nanoparticles afforded exceptional charge trapping/release capability with fast switching behaviors.

Lee et al. reported ultra-flexible organic flash memories based on polymer dielectrics, prepared by initiated chemical vapor deposition (iCVD) (Figure 2c) [27]. The fabricated memory device exhibited both industry-compatible operating voltage (~10 V) and relatively long-projected retention time. Furthermore, the flash memory devices on various flexible substrates, such as PET, Mylar, and paper, showed bendability down to a radius of 300 μm.

4. FERROELECTRIC FIELD-EFFECT-TRANSISTOR MEMORY

FeFET memory employs ferroelectric materials as dielectric layer, where data is stored based on semi-permanent polarization upon the application of external electric field. Due to the rapid polarizing speed and tolerance to external environments of ferroelectric materials, FeFET memory devices exhibit fast switching speed and notable read/write endurance. Among various ferroelectric materials, flexible FeFET memory devices have recently been focused on poly(vinylidene fluoride-trifluoroethylene) (P(VDF-TrFE))-based materials, because of their solution processability and relatively large polarization [28]-[30].

Xu et al. reported flexible ferroelectric organic nonvolatile memory based on poly(vinylidene fluoride-trifluoroethylene-chlorofluoroethylene) (P(VDF-TrFE-CTFE)/polystyrene (PS) dielectric (Figure 3a) [31]. Especially, authors enabled low-voltage operation via vertical phase separation of P(VDF-TrFE-CTFE)/PS, where an ultrathin PS buffering layer covers the surface of the P(VDF-TrFE-CTFE) layer through a one-step spin-coating process. The resulting memory device exhibited P/E voltage of ±10 V, reliable P/E endurance of > 150 cycles, retention time of > 10^4 s, and excellent mechanical bending durability at a curvature radius of 5.5 mm.

Similarly, Xu et al. reported flexible ferroelectric organic field-effect transistor nonvolatile memory [32]. In this contribution, authors inserted an ultrathin AlOx layer between the semiconductor film and the ferroelectric polymer (P(VDF-TrFE-CTFE)) film to enhance the charge carrier mobility. The resulting memory device on PEN substrate showed high mobility of 6.5 cm^2·V^{-1}·s^{-1}, a memory window of 17.2 V, a high memory ratio up to 2×10^5, and a reliable switching endurance over 600 cycles at P/E voltages of ±15 V. It

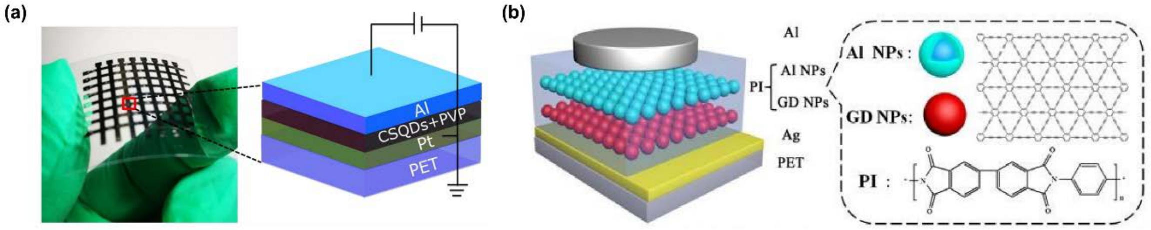

Figure 4. (a) Photograph of the flexible Al/CSQDs-PVP/Pt/PET resistive switching memory device and schematic conductive diagram of experiments. Cathodes were grounded for the tests. (a) Reproduced with permission.[37] Copyright 2016, American Chemical Society. (b) Schematic illustration of the fabricated resistive random access memory. The insets are the structure diagram and the molecular structure of the used materials. (b) Reproduced with permission.[39] Copyright 2017, The Partner Organisations.

also showed mechanical flexibility and maintained memory characteristics after 2000 times of bending with a radius of 5.5 mm.

Li et al. attempted to overcome the limitations of organic ferroelectric memory transistors – low current density, low operating speed, and poor mechanical stability (Figure 3(b)) [33]. By employing short channel length, authors achieved memory device with large current density of up to 6.32 mA·cm^{-2} and fast operation speed. Furthermore, vertical current flow across the semiconducting layer reduced the influence of relatively rough surface morphology of ferroelectric polymer (P(VDF-TrFE)), affording enhanced electrical performance, as well as mechanical stability under bending radius of 27 mm. The developed flexible memory device in this study showed potential for the application in wearable and flexible electric devices.

Kang et al. reported low-voltage organic transistor memory fiber on a thin and flexible metal wire (Figure 3(c)) [34]. By forming a thin, uniform nano-grained organic ferroelectric (P(VDF-TrFE) film on the wire, excellent switching stability (~100 cycles), long retention time ($> 5 \times 10^4$ s), and low-voltage operation (< 5 V) were achieved. Furthermore, the fiber-shaped memory devices were sewn in a stretchable textile fabric, and the resulting memory fiber exhibited negligible degradation of memory window even under a constant diagonal strain of 100 %. This study clearly showed the potential of wearable fiber-type memory device.

5. RESISTIVE MEMORY

Resistive memory generally consists of a metal-insulator-metal (MIM) structure. Unlike the other two types of organic flexible memory, resistive memory is easy to scale down the production process because of its simple two terminal-based structure. Resistive memory stores data through the conductivity change of insulator material via various mechanisms, such as metal filament formation, space charge and trap, and conformational charge, etc. This switching pattern in the conductivity of insulator materials, between high resistance state (HRS) and low resistance state (LRS), leads to the data storage in resistive memory [35], [36].

Zhang et al. synthesized Co_9Se_8 quantum dots (CSQDs) and employed them in the fabrication of flexible resistive switching memory device (Figure 4(a)) [37]. By blending CSQDs and PVP, high dispersity and stability as well as uniform film quality were obtained. The resulting memory device with Al/CSQDs-PVP/Pt/PET structure showed high current on/off ratio of 10^5, low operating voltage of 1.6 V, and good stability. The device also showed decent flexibility, maintaining a high current on/off ratio of 10^5, even under a bending radius of 7.1 mm.

Rosales-Gallegos et al. reported nonvolatile rewritable organic memory devices, employing nanocomposites of nitrogen-doped multi-walled carbon nanotube (NCNT) and poly(3,4-ethylene dioxythiophene):poly(styrenesulfonate) (PEDOT:PSS). [38]. The bistable resistive switching characteristics, originated from the creation of oxygen vacancies, were obtained at very low NCNT content in the polymer matrix. The fabricated memory device exhibited memory ratio of 10^3, retention time of 10^4 s under operating voltage of ± 4 V, and a few hundredths of write-read-erase-read cycles. Furthermore, the NCNT-based composites could readily be applied on flexible substrate (PET) for the fabrication of flexible memory device.

Jin et al. employed graphdiyne (GD), a new carbon allotrope with a 2D structure, for the fabrication of resistive random access memory (RRAM) devices (Figure 4(b)) [39]. Flexible memory device was fabricated on PET substrate with Ag bottom electrode, where GD nanoparticles as well as Al-Al_2O_3 core-shell NPs were inserted into polyimide insulating film. Existence of two kinds of electron trapping sites enabled a three-stage operation of memory device with long retention times of $> 10^4$ s.

Casula et al. reported resistive memory device based on parylene-C resistive layer between Ag electrodes on PET substrate [40]. The resulting device showed high current on/off ratio of 10^8 and long retention time of ~10^8 s at low operating voltage (< 1 V). Furthermore, the flexible memory device maintained good retention characteristics, even after 500 bending cycles, demonstrating excellent mechanical reliability.

Zhao et al. reported RRAMs based on organic bulk heterojunction active layer [41]. An active layer formed by blending N2, 7-bis(4-methoxyphenyl)N2, N7-bis (2-spiro [fluorene-9,9′-oxanthracene]-spiro[fluorene-9,9′-oxanthracene]-2,7-diamine) (X55) and 1-(3-methoxycarbonyl)propyl-1-phenyl[6,6]C61) (PCBM) resulted in higher ternary memory performance than those formed by pure X55, PCBM, or bilayer X55/PCBM. Furthermore, the bulk heterojunction active layer could be applied on flexible substrate, affording reliable mechanical flexibility even after 5000 times of bending cycles or a bending angle of 62°.

6. CONCLUSION

With the release of several foldable smartphones last year, a new era for flexible electronic devices has begun. Since the semiconductor industry has been primarily driven by the development of memory devices, the realization of flexible memory is of great importance. In this regard, this paper reviews three distinct types of flexible organic memory devices in terms of operation mechanism, performance parameters, and recent achievements. As the first review on flexible organic memory, this paper aims to inspire further research in the development of high-performance and stable flexible organic memory. In order to achieve this goal, more efficient materials, as well as fabrication methods on flexible organic memory, should be investigated.

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