Organic electronics for neuromorphic computing

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Abstract

Neuromorphic computing could address the inherent limitations of conventional silicon technology in dedicated machine learning applications. Recent work on silicon-based asynchronous spiking neural networks and large crossbar-arrays of two-terminal memristive devices has led to the development of promising neuromorphic systems. However, delivering a compact and efficient parallel computing technology, such as artificial neural networks embedded in hardware, remains a significant challenge. Organic electronic materials offer an attractive alternative for such systems and could provide biocompatible and relatively inexpensive neuromorphic devices with low-energy switching and excellent tunability. Here, we review the development of organic neuromorphic devices. We consider different resistance switching mechanisms, which typically rely on electrochemical doping or charge trapping, and discuss the challenges the field faces in implementing low power neuromorphic computing, which include device downscaling, improving device speed, state retention and array compatibility. We highlight early demonstrations of device integration into arrays and finally consider future directions and potential applications of this technology.

Artificial intelligence (AI) and deep learning algorithms are becoming increasingly important in many applications. While these algorithms resemble the workings of the human brain, they are traditionally implemented on a software level rather than emulated by hardware. Deep learning relies on artificial neural networks (ANNs) that are typically executed on computers based on the conventional von Neumann architecture, operating mostly sequentially. In contrast, the brain's hardware operates in a massively parallel fashion through a densely interconnected network of neurons. Communication between neurons is facilitated by chemical fluxes inside synapses that regulate the signal strength - or synaptic weight - that one neuron can pass to the next[1] and follow the Hebbian learning principle: neurons that fire together, wire together[2]. This plasticity is thus thought to form the basis of learning and memory, and to be largely responsible for information processing inside the brain. A consequence of this architecture is that the brain is extremely energy efficient compared to traditional computers, particularly for pattern recognition and classification tasks.

To execute neural network algorithms at a comparable energy efficiency and interconnectivity to that of the brain, it would thus be desirable to emulate synaptic functionality. Devices and circuits that possess the necessary characteristics were first described by Carver Mead in 1990, coining the term neuromorphic electronic systems[3]. As such, the idea of neuromorphic electronic systems has been around for decades, albeit with varying research attention.

On the one hand, asynchronous spiking neural networks based on silicon neurons and synapses, consisting of multi-element circuits[4], have been the focus of attention in emulating large-scale biological neural networks as well as in event-driven computing[5]. In these networks, information is encoded in spike timing and frequency[6]. Spiking networks have been utilised in some of the first commercial products, such as TrueNorth[7], NeuroGrid[8]and Intel Loihi[9].

On the other hand, following the seminal paper from Leon Chua, in which he theoretically described a memristor[10], and the first experimental demonstration of a memristor in 2008[11], there has been a significant amount of research to facilitate parallel computation, such as analogue vector-matrix multiplication[12] by forward-inference neural networks[6]. In this case, each synaptic weight in an artificial neural network is emulated by a hardware-based tuneable non-volatile resistive memory element (see Box 1). In contrast to spiking neural networks, information is encoded solely in the resistance state of the non-volatile memory device.

Memristors, or more accurately, memristive devices (no magnetic flux is involved which renders the term memristor not fully applicable[10]) are resistance switches that display a variable but non-volatile electrical resistance, depending on the history of applied voltage and current[19]. However, the concept of variable resistance devices goes back to the late 1960s/1970s[20,21] and such devices have been demonstrated to display synaptic functionality resembling Hebbian learning[22]. These two-terminal memristive devices allow for the construction of networks that can be used as non-volatile memory arrays as well as for processing information and carry out simple pattern recognition tasks directly in hardware, at relatively low energy cost[23,24]. The work of Strukov and colleagues demonstrated that neural network algorithms, traditionally implemented on a software level, could indeed be embedded in hardware itself, thus emulating the function and efficiency of the brain on a compact chip.

Although the use of an integrated network of memristive devices aims to efficiently emulate the parallel operation of the brain[6], ideal neuromorphic devices intended for forward-inference neural networks should: operate with low energy (to reduce power consumption); have a large linear and symmetric range of conductance states to facilitate "blind" synaptic weight update during learning; and efficiently perform parallel vector-matrix multiplication. State-retention time (a measure of how well a device can keep its state) requirements can vary significantly depending on the application but generally longer state-retention times are favoured (for example, for continuous learning the synaptic weights can be offloaded to an external memory whereas for train-once inference-only applications, the weights are stored long-term on-chip).

Furthermore, bio-inspired devices based on tuneable memory elements, require emulating additional brain-like functionality, such as spike-timing-dependent plasticity (STDP), spike-rate-dependent plasticity (SRDP) or short-term and long-term potentation[25,26]. As a result, detailed requirements of neuromorphic devices are highly dependent on the particular application as well as on the specific neural network architecture. Nevertheless, for efficient operation of hardware-based neural networks, several metrics are desired[19,27–29], as suggested for devices relying on organic electronic materials in Table 1.

Tuneable organic electronic materials and devices can serve as highly attractive alternatives to conventional memristive devices in specific neuromorphic applications, particularly for online learning where the synaptic weights are learned on-chip to make real-time predictions (i.e. inference). The specific nature of organic materials may offer novel and alternative switching mechanisms that are less stochastic while retaining low-energy operation and large dynamic range, enabling high training and inference accuracy.

Furthermore, organic electronic materials are generally inexpensive, can be integrated in low-cost manufacturing processes such as inkjet printing, and their chemical, electrical and mechanical properties can be tailored to the desired application by chemical synthesis[30]. As such, organic electronic materials have been recently utilised in a variety of neuromorphic device configurations, as well as proposed for biology related applications, potentially opening up a path towards efficient and adaptable brain-machine interfacing. In this Review, we discuss how organic electronic materials could meet some of the most important criteria required for neuromorphic computing: analogue conductance tuning via access to a large number of distinct conductance states while maintaining low power consumption. At the same time, we highlight key remaining challenges, which include device reproducibility, integration with conventional electronics, and the absence of a fully scalable fabrication process.

State-of-the-art organic neuromorphic devices

Similar to their inorganic counterparts, organic memristive devices have traditionally been developed for non-volatile memory applications and generally consist of a two-terminal "metal-insulator-metal" configuration that demonstrates two stable switchable conductance states[31–36]. Different configurations and active materials such as polymers[37], small molecules[38] and donor-acceptor complexes[33,34] as well as ferroelectric materials[39,40] have been proposed. In general, resistance switching in organic electronic materials is achieved by similar mechanisms as in inorganic materials, such as filamentary conduction[33,41], ionic charge transfer and electromigration[36] (Fig. 1). Most of these materials and devices, as well as their switching mechanisms, have been extensively described in other review articles[42–45]. Although binary (two-state) memory devices for neuromorphic computing have been demonstrated[4,46], here we will mainly focus on organic memristive devices enabling continuous (i.e. analogue) resistance tuning which is ideally suited for on-chip learning and inference using parallel multiply-accumulate operations[6,47] and has also been proposed in a related inorganic system[48]. At the same time, the resemblance of specific organic memristive devices with biological synapses (e.g. the coupling of ionic and electronic currents) constitutes significance, particularly for emulating biological neural network behaviour using artificial synapses, which is also briefly described.

In addition, the specific requirements for memory applications differ from those relevant to neuromorphic computing. More concretely, while both applications require relatively high switching speeds and cycling endurance, state-retention time requirements are much more stringent for memory applications than for neuromorphic computing (see Table 1), which is highlighted by the long state-retention of Write-Once-Read-Many times (WORM) devices[49]. On the other hand, it is essential for neuromorphic devices to display a large range of separable conductance states to efficiently perform neural network operations[6,28]. The unique conductance tuning mechanisms of a range of organic electronic materials and devices have been exploited to demonstrate a variety of neuromorphic devices. Resistance switching in most devices relies either on electrochemical doping or charge-trapping, as summarised in Table 2 and schematically depicted in Figure 1.

Electrolyte-gated redox-based switching. Most prominently, potentiation and depression effects are achieved by means of a gate electrode to gradually tune the device's conductance via electrochemical doping (Fig. 2b-c). This process can be achieved using an electrolyte (liquid or solid) that injects or extracts ions from the organic film, changing the doping (redox) state of the latter. Kaneto et al. already demonstrated in 1991 a device in which two different currents, electronic and ionic, flow in perpendicular directions. This concept was utilised for the conductivity tuning of an organic material with a solid electrolyte over 3 orders of magnitude[50]. Later, the work of Berggren[51] and Fontana[52] demonstrated liquid-electrolyte-based devices in which two stable conductance states were demonstrated with a large ON/OFF ratio between the low and high conductance states (10^5 in ref. [51] and ~10^2 in ref. [52]). A similar configuration with an electrolyte-gated conducting polymer was used to display a range of neuromorphic functions[53] while other polymeric materials, electrolytes[54–56] and nanowires[57] have also been reported.

More recently, a modification of an OECT-based system was demonstrated comprising a conducting polymeric gate in combination with a de-doped conducting polymer channel[58], i.e. a device architecture reminiscent of an organic battery where a counter redox reaction in the gate ensures electrical neutrality throughout the films, resulting in enhanced state-retention. Similar battery-like devices were also demonstrated using different polymers and solid electrolytes[54,59] and in a two-terminal configuration[60], however, these devices were lacking an ionic-conductor separation layer between the two electrodes to prevent recombination reactions causing undesirable self-discharge and limited state-retention. Although typically these devices display only a low and high conductance state, in some cases one of these states can be modulated in a continuous fashion by varying the gate potential or pulse frequency, thus enabling more than two conductance states, see Table 2 and Figure 2 a,b.

Overall, the easily tailored characteristics and large tuneable conductance range reported in a variety of electrolyte-gated organic materials and configurations, have thus far shown great promise for neuromorphic computing applications.

Charge-trapping based switching. Another widely used mechanism to display a memory effect and neuromorphic functionality in organic field-effect transistors is based on charge-trapping and was developed by Vuillaume et al[61] following previously reported bi-stable two-terminal organic memory research[62,63]. These devices rely on charge storage on metallic (e.g. Au) or non-metallic (e.g. ZnO)[64] nanoparticles that act as nanoscale capacitors embedded in an organic semiconductor such as pentacene or PMMA (Fig. 1d). The charged particles electrostatically repel the mobile holes in pentacene and thus effectively modify the source-drain behaviour, enabling resistance tuning.

Since the nanoparticles reside inside the bulk of the material rather than in a separate layer, the mechanism differs from conventional floating-gate transistors and related floating-gate organic memory devices[65,66]. Nevertheless, the channel still has to be switched on by a gate potential to perform a read operation[67]. The requirement to apply a gate voltage to operate the device was removed by connecting the gate with the drain to form a pseudo-two-terminal configuration (requires only a source-drain voltage), thus enabling the channel conductance to be modified as a function of the frequency of applied pulses[68].

Organic memory transistors based on charge-trapping are promising, especially since they offer relatively large channel resistance - desirable for low power computing - and long state-retention times (~10^5 s). However, small channels can fit only a limited number of nanoparticles, possibly limiting device performance in dense device arrays. It remains to be seen whether these devices can be scaled down while retaining sufficiently low operating noise and device reproducibility.

Opportunities and challenges for organic neuromorphic devices

As demonstrated by the successful large-scale commercialisation of organic light-emitting diodes and initial attempts to commercialise organic photovoltaics, organic materials span a wide spectrum of properties that could be advantageous for neuromorphic computing applications, such as their excellent ability to be tailored, chemically, mechanically as well as electrically, to specific requirements. These qualities were recognised early on and have led to a wide variety of unique demonstrations of organic memories and organic neuromorphic devices. Most prominently, the ability for the conductance to be tuned, combined with low energies required to do so, make organic materials specifically suitable for neuromorphic applications. However, at the same time, the nature of these materials introduces challenges, specifically regarding stability, integration and device-to-device variability. Some of these challenges have recently been addressed in the literature but several remain, as the field of organic neuromorphic computing is steadily growing[79].

Number of conductance states. Conventional organic and inorganic memristive devices often display only one low and one high conductivity state. Several memristive devices, such as phase change materials[80], resistive change materials and conductive bridge memristive devices have been demonstrated to display more programmable states[27], but are often stochastic and lack predictability, e.g. have high write noise. Biological synapses on the other hand, can change their conductance - or synaptic weight - in a virtually analogue fashion. In fact, hardware-implemented forward-inference neural networks generally require some form of analogue variation in the synaptic elements[81] while a large tuneable conductance range enhances artificial neural network accuracy and enables analogue computing[28].

The first demonstrations of organic neuromorphic devices with a large number of conductance states were based on tuning the amount of charge on the gold nanoparticles dispersed throughout the bulk of a transistor channel[61,67]. As the charged nanoparticles repelled mobile charge carriers inside the film, it was possible to accurately tune the channel conductance, with an ON/OFF ratio of about 10^3 – 10^4 [75]. Similarly to multi-level floating-gate memory, the charging of a gate electrode that was separated from the channel, also induced multi-level storage in organic memristive devices, although corresponding write voltages were relatively high (~40 V)[65].

Most electrolyte-based redox coupling mechanisms were reported to display a gate potential-dependent tuning of the channel conductance[52,56,74,73,82] (Fig 2a) or demonstrated that a train of pulses could potentiate or depress the channel[53,55,56,74,69] (Fig 2b). These tuning mechanisms rely on the change in the redox state of the polymer due to the electrochemical potential induced by the gate electrode. Changing this redox state of the polymer results in a reduction or enhancement of the conductivity by increasing or decreasing the number of mobile charge carriers, respectively. At the same time, ions or other charge carriers have to compensate the induced space-charge in the film. Due to the physical movement of these charges, the process can be controlled relatively well by varying the gate potential and pulse frequency and in fact resembles the processes occurring in biological synapses. Nevertheless, for a given gate potential, the final conductance in the channel will still be the same, regardless of modulation by short pulses or a continuous gate potential.

In contrast, recently a near-analogue tuning of channel conductivity was demonstrated by accurate protonic doping of a polyethylene-imine that in turn was used to de-dope the conducting polymer[58]. This device operation resembles that of an organic battery[83] where both polymer electrodes are allowed to change their redox-state during switching, thereby ensuring charge neutrality in both electrodes. In between read and write operations, the two electrodes are electrically isolated from each other and prevent charge exchange, thereby enhancing state-retention considerably – the device effectively operates as a non-volatile memory. As a result, it was possible to accurately tune the conductance through the complete set of redox states of the polymer (Fig 2c) resembling conductivity modulation by p- or n-type doping of silicon.

While two-terminal devices commonly comprise organic memories that display only two conductance states, a few examples of multi-state devices exist. For instance, a redox coupling mechanism created a

gradual change in conductance by consecutive voltage stimulations[60] while a more traditional metal-polymer-metal configuration succeeded in displaying 100 conductance states[77]. In the latter, the authors used a Ti:PEDOT:PSS:Ti sandwich structure in which the interface between metal and conducting polymer was modified by the growth and migration of a Ti-compound, to display a gradual change in conductance. While these two-terminal based concepts benefit from possible smaller sizes and a more straightforward integration in a crossbar array and operation, effective utilization of this concept would rely on significantly decreasing the write noise during the predominantly stochastic switching.

Finally, a more exotic multi-state organic memory device was demonstrated based on optically modifying the charge transport inside a P3HT film blended with photochromic diarylethene[78]. Using ultraviolet and green light irradiation, the energy levels of the photochromic material inside the polymer matrix could be modulated. Modifying the highest occupied molecular orbital (HOMO) levels towards that of P3HT results in better charge transport. As such the authors were able to accurately tune the device to 256 distinguishable conductance states. This is an interesting concept for further development in neuromorphic applications, especially since light-assisted programming[85] could potentially overcome several limitations in electrical memristive-based crossbar arrays, such as unwanted (sneak) currents, without the necessity of sophisticated access devices[16] preventing cross-talk.

Short-term plasticity. In the brain communication between neurons is inherently dynamic and occurs over different timescales, ranging from milliseconds to months[1]. Changes in communication strength depend on the history of synapse activity and are known as synaptic plasticity. Short-term plasticity modulation facilitates a variety of computational functionality in the brain, while long-term plasticity effects are attributed to learning and memory[2]. Both of these functionalities have been reported in organic devices, with distinctions that predominantly lie in the specific application that is targeted.

Short-term plasticity is mostly useful in mimicking synapses and displaying synaptic functionality such as spike-timing-dependent plasticity (STDP), spike-rate-dependant plasticity (SRDP) and short-term to long-term plasticity. Devices and technologies targeted toward mimicking these specific synaptic functions are generally called artificial synapses and in addition from being able to aid understanding processes and dynamics inside the brain, these devices have also been found useful in spiking neural networks[4,7]. As with real synapses, artificial synapses are aimed at displaying a conductance that is a function of the history of previous applied pulses. In some cases, a leaky integrate-and-fire element is included as an artificial neuron, which resembles biological neuron functionality by controlling which signals pass to the next neural network layer. This is achieved through the integration of input signals until a threshold is reached, followed by the neuron firing a signal[4].

Spike-timing-dependent plasticity (STDP) is widely regarded as one of the fundamental mechanisms in biological neural networks that facilitates the modulation of the signal strength between two neurons inside a synapse. This process is based on the temporal difference between pre- and post- synaptic pulses arriving at the synapse. The shorter the time difference between those pulses, the higher the modulation of the synaptic weight. In contrast to error backpropagation[17], STDP is a local learning mechanism and thus far has been mainly demonstrated on a single device level in charge-trapping devices[68], electrolyte-gated three-terminal architectures[60,73,86] and two-terminal organic memristive devices[76,77]. Closely related to that, spike-rate-dependent plasticity correlates the modulation with the rate of pulses that arrive to the channel and was also demonstrated in several three-terminal configurations[67,76,87]. Finally, short-term to long-term plasticity can be described as the effect in which repeated stimulation will result in a long-term modulation of the conductance compared with a short-term plasticity effect for a single stimulation[88]. This transition was also imitated in an organic neuromorphic device by a gradual polymer-silver interface modification[76], see schematic in Fig 1a.

Particularly for use in spiking neural networks, as well as deeper understanding of biological neural networks, research on artificial synapses continues to inspire developments in mimicking biological synapses. However, for efficient on-chip learning it is essential to develop a global learning architecture next to the local STDP learning rule, while simultaneously increasing the state-retention times in these

organic neuromorphics devices, i.e. it is essential to develop artificial synapse devices capable of "long-term memory".

Long-term plasticity and state-retention. Long state-retention times and enhanced stability are desired for hardware-based artificial neural networks and related vector-matrix multiplication. In fact, long state-retention times were first reported during the development of non-volatile organic memories, where state stability is crucial to prevent data loss[39,49,89]. Specifically, WORM devices[49] have shown long retention times of up to 500 days[90], but more recent examples of multilevel optical (500 days[78]) or solution-processed azo-aromatics memories (11 days[91]) have also been demonstrated. Neuromorphic devices based on electrochemical doping allow operation in a larger conductivity range with more states than organic memories. To effectively use multi-level devices in hardware-based neuromorphic arrays, it is essential to increase the state-retention. A variety of research papers have included some form of state-retention measurements as highlighted in Table 2.

Conducting polymer-based organic electrochemical transistors generally operate in depletion mode[84]. This means that the channel is in a high conducting state until ions are introduced that cause the extraction of mobile holes and dedoping of the conjugated polymer backbone. When the gate voltage (V_{bias}) is switched off, these ions return to the electrolyte and the polymer is again doped, see Fig. 3a,e. This phenomenon in electrochemical transistors was first exploited to demonstrate basic short-term synaptic plasticity functions, such as short-term depression, adaptation, and dynamic filtering in a PEDOT:PSS based device[53]. Consequently, the device's state retention was based on the slow kinetics of ionic movement from the conducting polymer film back into the electrolyte and is thus considered volatile. Later, by slightly modifying the organic materials[92], certain non-volatile behaviour was also reported[74].

Using a specific biasing scheme, however, it was possible to induce a non-volatile memory effect for a few hours, in a similar organic electrochemical transistor[51]. More recently the addition of a ferroelectric layer, was demonstrated to have a long lasting memory effect[93], typical for ferroelectric materials[39]. In a polyaniline-based memristive device, the retention time was increased to about 10^3 seconds while simultaneously increasing switching rates by reducing the length of the conductive channel[72]. Physically limiting the movement of ions involved in the redox process, was also reported to increase the retention time[73]. In this case, however, the resulting films remain in an electrochemically meta-stable state since the ionic charges are effectively stuck in the polymer due to drift/diffusion disparity between the anions and cations.

For long-term retention, it is more suitable to ensure electrical neutrality throughout the films in the device. This can be achieved by allowing a counter redox reaction and was demonstrated in a two-terminal device with ethylviologen diperchlorate within a solid polyethylene-oxide electrolyte on top of a triphenylamine-containing polymer[60]. The cation in the solid electrolyte of this device can uptake an electron, while in the polymer side, an electron is removed, opening up a mobile hole, see Fig. 3b,f. Essentially, this device can be considered as an electrochemical battery with two redox systems and mobile ions compensating space-charge. The disadvantage of a two-terminal configuration, however, is that any pulse between the top and bottom electrodes, either write or read, results in bias through the complete redox system, thereby disturbing it. Thus, despite being electrically neutral, the reported device still lacks a high state stability, i.e. is volatile to a read operation.

In a similar battery-like configuration, but now comprising a three-terminal configuration and polythiophene as conducting polymer, a separation of the read and write circuits decoupled the redox reactions during conductance tuning (write) (see Fig. 3c,g) from the conduction state measurement (read) in the polymer film[54] and demonstrated an enhanced two-state stability[59]. While the conductivity modulation decreased marginally over time it was still possible to distinguish the two states. This modulation was thought to be the result of recombination reactions at the interface between the two sides of the redox system (orange dashed trace in Fig. 3g). In fact, by adding a charge separation layer between the two sides, thus forming a device similar to an electrochemical battery, comprising of an

electrolyte allowing only ion motion, it was possible to prevent interface recombination reactions and enhance state-retention for multiple states[58], see Fig. 3d,g.

As demonstrated in the previous sections, the electrochemical doping of conducting polymers allows for accurate tuning through a large range of oxidation and reduction states. However, a further reduced polymer is more susceptible to oxygen doping and consequently returns to an oxidized state. This inherently limits state-retention in semiconducting polymers but could be prevented by appropriate encapsulation, which reduces the effect of oxygen doping, or appropriate level tuning to stabilize the reduced polymer.

Charge-trapping is also an inherently meta-stable mechanism. However, the first reported devices based on charge-trapping already displayed a retention time of more than 10^3 seconds[61]. The main difference with conventional floating-gate transistors is that the gold nanoparticles are inside the polymeric channel (i.e. the electronic charge conduction path is through the film with the particles) which limits the state stability and state-retention of these charge-trapping devices. By using a monolayer of functionalised gold nanoparticles however, the authors were able to demonstrate a higher retention of up to 10^5 seconds[75].

Cycling endurance. For neuromorphic arrays to be useful for prolonged periods of time, next to state-retention, it is important to study whether device performance deteriorates over time, e.g. during extensive read/write cycling. However, these characteristics are not yet fully established, and endurance requirements are not yet clear. In flash memory devices for instance ~10^4 cycles are common, whereas SRAM and DRAM can be cycled for over 10^{16} times[19]. To investigate device performance, endurance measurements are usually done by cycling between conductance states or over the complete dynamic range. Although not many studies on organic neuromorphic devices include complete endurance cycling (i.e. until failure), successful cycling was demonstrated ranging from 20 cycles in two-terminal memristive devices[77] up to 800 cycles in charge-trapping devices[75]. Redox-gated architectures were demonstrated from 50 cycles in early work[70] up to 10000 cycles, more recently[72]. See also Table 2. The optical memory-based device, on the other hand, showed state-retention times of up to 10^7 seconds with close to no deterioration after 70 cycles[78]. These preliminary results show great promise for long-term operation of neuromorphic devices and suggest that there is no intrinsic obstacle to achieve organic devices with high cycling endurance.

Energy consumption. Energy consumption per synaptic - or switching - event is closely linked to read stability. In fact, read stability of two-terminal devices is inherently linked to the energy necessary to write or erase a state: low switching energies result in reduced stability, since a reading operation of that state could disrupt the state itself. This trade-off is commonly circumvented by using a lower voltage to read a state and an elevated voltage or current to write to a new state. Conventional inorganic memristive devices have reported writing energies in the order of 1-1000 picoJoules[19,27] with typical device dimensions between 0.1 – 5 μm, depending on the specific mechanisms and materials. Ultimately, switching energy should be below 100 pJ to be competitive with traditional CMOS logic circuits[29].

Energy losses in the electrode lines leading to the devices in an array should also be taken into account. What this means is that a device located in the centre of an array should receive a certain minimum voltage which can be relatively high depending on the switching mechanism. Additional resistive losses in the electrode lines leading to that device may cause significant higher energy costs[19]. For most filament-forming resistive switching devices a large energy difference between potentiation and depression is observed, due to the high resetting current that is required. This was also the case for a reported polymer-based filament-forming resistive-switching mechanism, that required an energy between 0.1 – 100 pJ to switch, depending on the pulse number and whether it was lowering or enhancing the conductance[94].

Redox coupling in polymeric materials is inherently a low activation energy process where the voids between polymer chains, i.e. the free volume, facilitate the energetically cheap and reversible ion exchange, and thus enable low energy switching. As a result the energy for a single spiking operation resulting in a short-term modulation was found to be in the range of 10 pJ for a ~50 μm^2 device[73]. For a complete write-read-erase operation of a 100 x 100 nm polymer device the energy was estimated to be less than 10 pJ [59].

In an effort to demonstrate ever decreasing switching energies, a hybrid organic-inorganic perovskite memristive device was recently described having switching energies in the order of femtojoules per 100 nm^2 [95] which is comparable to the 10 fJ per synaptic event in the brain[27]. These low values are generally closely correlated with a lower state stability, as a low switching energy implies a switching process close to reversibility. In such conditions, slow ion kinetics are expected to ultimately result in a return to the original state, but this remains to be explicitly investigated.

In a battery-like artificial synapse with a higher state stability, the minimum energy necessary to reliably switch a 100 μm^2 device was measured to be less than 0.4 pJ when the voltage source was tuned to the open-circuit potential of the device using a potentiostat. The energy was also found to scale with channel area, leading to an estimated absolute minimum switching energy for a 300 x 300 nm device of 35 aJ[58]. The large difference in energy when compared to other approaches might originate from the fact that in a battery-like artificial synapse changing states requires only slightly charging a capacitor which does not require much current. Furthermore, enhanced state-retention has the advantage that it enables to define separate conductance states with a smaller difference in conductance between adjacent states.

Charge-trapping devices traditionally operate with relatively high voltages (~10-40 V) and consequently display high switching energies, although some examples of two-terminal bi-stable organic memory devices exist that operate with lower voltages between 3 and 5 V[96,97]. More recently it was demonstrated that by optimising the fabrication process of a nanoparticle organic memory field-effect transistor, the voltage required to switch was lowered to 1 V and the related energy could be reduced to 2 nJ[98].

Further reducing the size of organic devices was also proven to decrease the necessary programming energy significantly, down ~1 fJ for an electrolyte-gated polyethylene oxide nanowire, the lowest value reported to date[57]. As such, organic neuromorphic devices have demonstrated great potential in enabling low switching energy, an essential characteristic for future low-power neuromorphic computing. However, whereas low-power devices are highly desirable, the energy required to operate the complete neuromorphic system is the most relevant metric, e.g. a low-power device which cannot be implemented in an energy-efficient neuromorphic system, is not useful. Therefore, when developing single devices, their integration into and the efficiency of the final neuromorphic system must be carefully considered.

Integration and biocompatibility

One common issue with organic electronics is that large-scale integration of devices into useful applications may suffer from low device reproducibility and relatively low yield. Still, a growing number of successful implementations of integrated devices is being demonstrated. Large-scale integration of organic neuromorphic devices such as previously demonstrated in flexible organic memory arrays[89] has been reported by Yang et al. in a flexible 3D-network configuration[94] (Fig. 4c). This network consisted of three stacked copper-doped polymeric layers acting as a non-volatile memory array. In single artificial synapses coupled with a selector device, correlated learning following a spike-timing-dependent plasticity mechanism were demonstrated. Another integrated network was created by phase separation of block co-polymers[99]. Here, the authors were able to simultaneously potentiate and depress an organic film by creating separate conductance paths in three dimensions, effectively self-forming two memristive devices on a crossbar-point between four electrodes.

Apart from large (crossbar) arrays, several implementations of functional organic neuromorphic circuits have been reported such as a single-layer perceptron consisting of two[100] or three[82] organic

neuromorphic devices that could classify inputs via supervised learning. A perceptron emulates biological neuron functionality by summing and mapping the input signals to the output signal[101]. The former architecture also included the activation function based on an organic field-effect transistor and two organic resistors[100] but training of the memristive devices was done off-line. In contrast, on-chip training was implemented to perform NAND and NOR functionality using simple threshold currents[82] as well as more complex circuitry combining organic memristive devices with CMOS based-neurons[102]. More elementary learning functionality, such as simple associative learning (e.g. Pavlov's Dog), was also reported in a variety of research papers[58,69,100,103,104].

An important feature of organic redox-based neuromorphic devices is the electrolyte, which allows novel architectures to be designed. One example is an architecture with one channel and multiple gates. The spatiotemporal coupling of applied signals to the latter controls the state of the channel. Using this concept, information processing functions (e.g. orientation selectivity) inspired by the mammalian visual system were demonstrated[105,106] and later mimicked in a different polymer[107]. The electrolyte can also be used to couple one gate to multiple channels, in a way that is reminiscent of global control of neurons in the brain, obtained by parameters such as hormones, ion concentration and temperature[108] (Fig. 4b). This property can be useful in large device arrays where a common bias is necessary. Other, more general integrated ionic-based logic gates such as inverters and NAND gates were reported that can be potentially useful for integration with organic neuromorphic circuitry[109].

Furthermore, the soft mechanics and biocompatibility of organic materials render organic memristive elements and arrays highly appealing for interfacing with biology (Fig 4a,d). Such interfacing was demonstrated in a wide variety of applications, such as implants[110,111], (Fig 4a), biosensors[84] and biomimetics[112], as well as bio-inspired and biomaterials-based memories[113]. At the same time, observation of short-term potentiation and synaptic operation in organic memristive arrays shows that these devices could operate with biological cells placed on top of the array[87] (Fig. 4d). Recently, organic ion gel-gated transistors were used in a flexible organic artificial afferent nerve to detect movement and pressure for use in neurorobotics and neuroprosthetics[114] (Fig 4e). Additional development could further enhance the connection between synaptic memristive arrays and adaptive control of physiology and processes of cells, tissue and organs; and vice versa to enhance site-specific sensing and monitoring[111,115].

Despite some progress in terms of integration of organic neuromorphic devices, several challenges lie ahead to achieve large-scale programmable and functional neuromorphic arrays. Although the activation function, implementing biological neuron functionality, was emulated by organic field-effect transistors[100], it is not yet well-established how this functionality will be implemented on large scale arrays with multiple hidden layers. Furthermore, sneak currents and voltages (unwanted conducting paths) through crossbar array-based networks, are a common problem that require solutions in the form of rectifying behaviours inside the materials or additionally integrated access devices[16] such as in dot-product engines[12].

Outlook

Organic materials have the potential to be successfully exploited for neuromorphic computing due to low-energy switching and excellent tuneability in addition to their low-cost and biocompatibility. Specifically, access to a large conductance range and corresponding number of states is highly attractive for hardware-based forward-inference neural networks and related vector-matrix multiplication which could enable fully parallel read and write neural algorithm accelerators. Furthermore, the ability to tailor the electrical, chemical and mechanical characteristics of organic compounds could allow for the development of near-ideal materials that possess long-term stability, linear switching characteristics and can switch at low energies all while enabling integration in various form factors.

In spite of recent successes, more research is necessary to overcome current limitations for organic neuromorphic devices to succeed. Read and write noise should be sufficiently reduced and state-

retention further increased, especially for long-term operation. Encapsulation could help increase device stability and cycling endurance to desired levels.

While device-to-device variability remains an issue during fabrication, improvements are expected from high-quality manufacturing facilities. Still, apart from a few recent successful demonstrations of large-scale integration, organic neuromorphic arrays might suffer from low device reproducibility (particularly for devices downsized <1 μm^2) and consequently array failure. This requires further investigation and development. Nevertheless, the large conductance range of redox-coupled devices can be used to determine an acceptable but reduced range for a large number of inferior devices that are fabricated in the same array. Combined with linear symmetric switching[116], this could enable sufficient array performance.

Other essential functionality such as the activation function and related mapping of the input signals to output signals is still lacking - the path toward an efficient all-organic neuromorphic system is not yet clear. Eventually, it is very likely to be important that organic tuneable memristive elements can also be integrated in inorganic or other conventional CMOS applications in order to broaden their appeal and adaptability. For instance, compact organic neuromorphic cores can be built-in within a digital CMOS processing unit to perform the most intensive vector-matrix operations, while communication between the neural cores would be executed by the digital CMOS processing units. Further integration into Back-End-of-Line (BEOL) processing (requiring materials to withstand elevated ~350-400 °C temperatures) would have to be investigated and devised. An efficient organic-inorganic neuromorphic system, leveraging the advantages of both technologies, could serve as motivation for this pursuit. At the same time switching mechanisms in organic devices can also form an inspiration for inorganic devices, as was recently demonstrated in an Li-ion based inorganic synapse for analogue computing[117]. In applications in which dedicated local functionality and low energy are important, organic electronic materials can serve as highly complementary to inorganic CMOS-based neuromorphic arrays.

Finally, switching speed, which is essential during the training phase, is not well understood. The fundamental device mechanisms and their dependence on the nature of the materials used must be investigated in order to determine what limits switching speed and how to improve it. An accurate physical model of these devices, which is extremely useful in this respect is still missing. Such a model would allow to accurately simulate the complete behaviour of arrays of devices, greatly accelerating design feedback loops.

More exotic weight update mechanisms, such as those based on optically adjusting the weights[85] could make organic neuromorphic devices unique compared to other technologies, with some advantages, such as the elimination of sneak currents as mentioned earlier. Other unique properties of organic electronic materials, such as low-temperature processing and inkjet manufacturing capabilities could potentially enable low-cost, disposable and simple neuromorphic-based lab-on-chips, that could be utilised for smart point-of-care devices. We speculate that developments in organic neuromorphic computing could lead to and be exploited for enhanced hybrid biological/organic functionality such as trainable and adaptable brain-machine interfaces[111], biosensor networks, robotic skin[118] or adaptable local control of prosthetics[114].

Reference 3
First mention of the term neuromorphics which started the field.

Reference 10
First theoretical description of the memristor.

Reference 20
Experimental demonstration of a non-volatile analogue memory.

Reference 50
First demonstration of hybrid elelectronic/ionic switching in a conducting polymer.

Reference 53
This article demonstrates synaptic functionality in an electrochemically gated conducting polymer device.

Reference 57
This article shows an artificial synapse that switches at femtojoule energy consumption.

Reference 62
This article demonstrates the first solution-processed bi-stable organic memory device based on charge storage.

Reference 94
A three-dimensional integrated artificial synapse network is demonstrated in this article.

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Competing financial interests
The authors declare no competing financial interest.

Box 1. Vector-matrix multiplication and learning can be efficiently emulated using hardware-based artificial neural networks.

Artificial neural networks connect an input layer (e.g. pixels in an image, list of data etc.) to an output layer using hidden layers, see the image below. Each node is connected to the nodes of the next layer and as such these networks rely on multiplication of large matrices and vectors for inference (i.e. prediction) as well as learning. Connections between the nodes (synaptic weights) can be modulated to train the network to perform the desired operation. Vector-matrix multiplication (the basis of ANN algorithms including convolutional neural networks[13], deep belief networks[14] and multilayer perceptrons[15]) can be efficiently implemented in hardware, e.g. in a crossbar array using analogue resistive memory elements[6] (indicated here by a variable resistor), by making use of Ohm's law and Kirchhoff's law. More concretely, vector-matrix multiplication performed in software $y_m = \sum_i w_{i,m}\, x_i$ is emulated via current-voltage operations as $I_m = \sum_i G_{i,m}\, V_i$, where $G_{i,m}$ is the conductance of the neuromorphic device at the i,m node, V_i is applied voltage at the i input, and I_m is the read out current at the m output. To avoid device crosstalk, each neuromorphic device is typically paired with an access device[16]. Learning in these networks generally relies on the backpropagation method, a supervised learning algorithm in which the synaptic weights are iteratively adjusted in accordance to the gradient of the error function[17]. These have been demonstrated to show promise in efficiently emulating artificial neural networks in recent commercially significant domains such as deep learning[18].

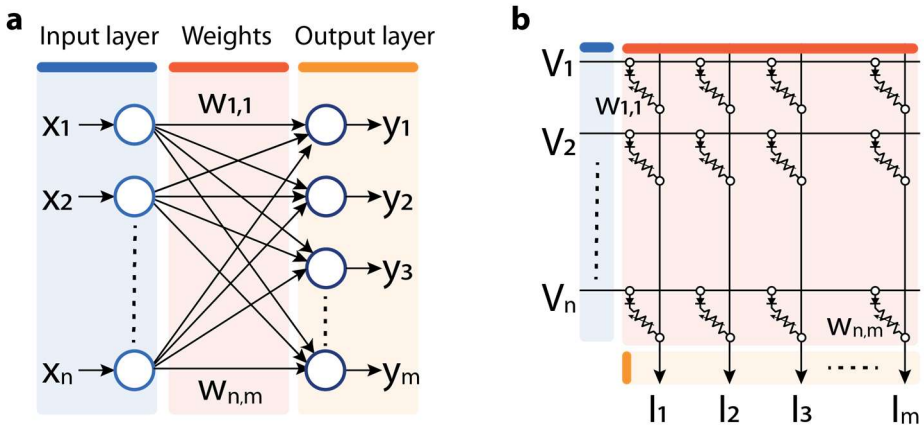

Table 1. Desired and recommended metrics for organic neuromorphic devices.

Parameter	**Value**
Size for integration	<1 μm^2 for dense/compact arrays
Number of states	~100 separable states or ~6 bit
Conductance Tuning	Linear and symmetric
Switching noise	<0.5% of weight range
Switching energy	<100 pJ per switching event
Write/read speed	<1 µs
State retention*	10^3 - 10^8 s
Write Endurance (cycles)**	~10^9 (online-learning)
Temperature stability***	Array operating temperature

** Long state retention is essential for hardware-based neural network inference using non-volatile memory elements, e.g. for a dot-product engine[12] but not as critical for online-learning where synaptic weight values can be stored elsewhere after training.*

*** High write endurance is more important for online-learning than for inference-only (e.g. dot-product engine), i.e. for the latter the write endurance requirements are less strict.*

**** Device temperature stability is important to consider since neuromorphic arrays may heat up significantly during array operation. The relevant temperature range depends on materials and system-level architecture. Note that device integration into inorganic/organic 3D stacks may require material stability at elevated processing temperatures, e.g. ~350-400 °C for Back End of the Line (BEOL) processing.*

Table 2. Switching mechanisms and materials used in organic neuromorphic devices, sorted by publication date (earliest at the top).

Principle	Material / electrolyte	# states / tuning *	Memory mechanism	State retention** / Demonstrated cycles***	Ref.
Electrochemical doping	poly(3-methyl thiophene) / poly(ethylene oxide-propylene oxide) + $LiClO_4$	4 / continuous	Redox counter reaction + separation read/write	~hours / -	[50]
	PANI / PEO	>2	Slow kinetics	- / 10^4 (ref. 72)	[52,69–72]
	MEH-PPV / $RbAg_4I_5$	8 / continuous	Diffusion disparity	~20ms for STP and >240 h for LTP / -	[73]
	PQT / PEO + $EV(ClO_4)_2$	2	Redox counter reaction + separation read/write	14 h**** (ref. 59) / >100 (ref. 54)	[54,59]
	PEDOT:PSS / NaCl	>2	Slow kinetics	<1 s / -	[53]
	PEDOT:PTHF / NaCl	continuous	Slow kinetics	<1 s / -	[74]
	BTPA-F / PEO + $EV(ClO_4)_2$	continuous	Redox counter reaction	~seconds / -	[60]
	P3HT / P(VDF-HFP) P3HT / P(VDF-TrFE)	>2	Slow kinetics	<10 s (ref. 56) / -	[55,56]
	P3HT / PEO	continuous	Slow kinetics	<5 s / 60	[57]
	PEDOT:PSS / NaCl PEDOT:PSS / Nafion	512 / continuous	Redox counter reaction + separation read/write	100 s / >15	[58]
Charge trapping	Pentacene (Au)	continuous	Charge trapping nanoparticles	24 h / 800 (ref. 75)	[61,67,68,75]
	DNTT (Al)	continuous	Charge trapping nanosheet	~1 s / -	[66]
Ion migration	Ti/PEDOT:PSS/Ti Ag/PEDOT:PSS/Ta	continuous	Compound formation	~ seconds (ref. 76) / -	[76,77]
Light-assisted reaction	P3HT / diarylethene	256	Energy level modification	>500 days / 70	[78]

** If mentioned in the original work, the number of demonstrated conductance states is given. Some devices display a low and high conductance state, where one of the two can be modulated in a continuous fashion by varying the gate potential or pulse frequency (marked as >2).*

*** State-retention strongly depends on the definition, e.g. the number of defined states, and on the state the device is discharging from.*

**** Cycling until device failure (endurance) has not yet been demonstrated for most organic neuromorphic devices. The numbers cited here represent the minimum number of cycles during which the device was operating successfully.*

***** Although the two conductance states were distinguishable during 14 hours of operation, the conductance of both states (ON/OFF) decreased by several orders of magnitude.*

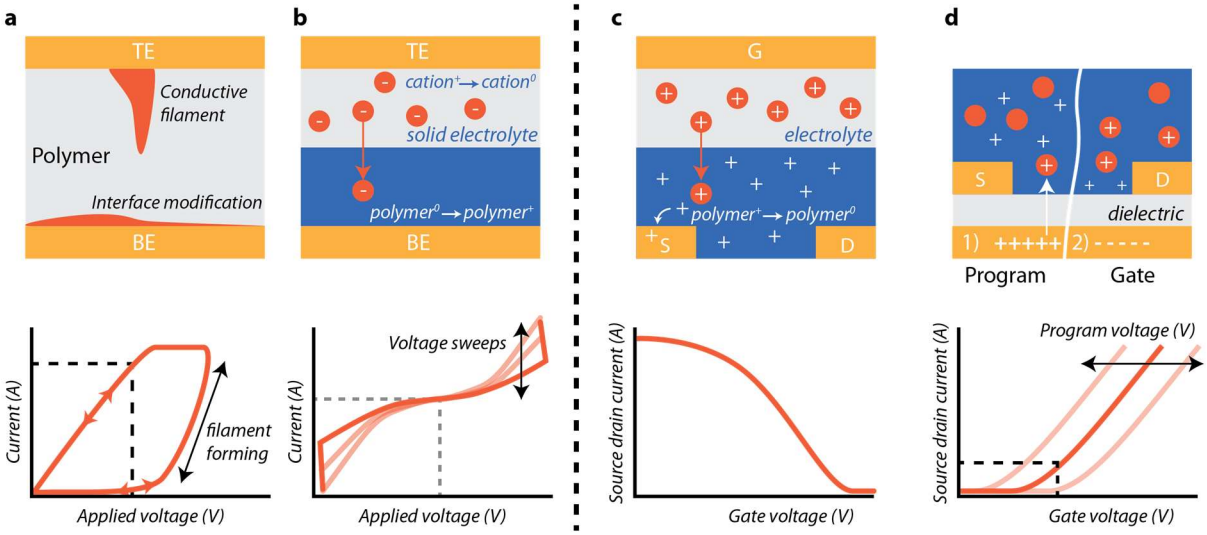

Figure 1. Overview of conductance switching mechanisms in organic electronic materials. (**a**) Two-terminal organic memory based on conductive filament formation and/or bias-dependent interface modification. (**b**) Two-terminal redox-based switching with a counter redox reaction (**c**) organic electrochemical redox-based switching. (**d**) charge-trapping based switching.

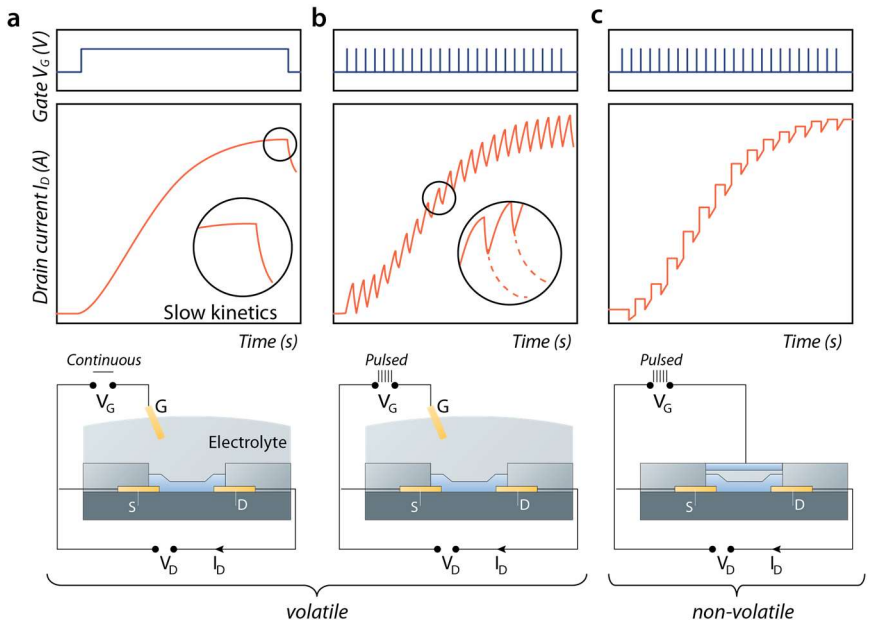

Figure 2. Conductance tuning methods for electrolyte-gated redox-based neuromorphic devices. (**a**) continuous gate (**b**) pulse train (**c**) pulse train with polymeric electrode gate and decoupling of read and write operations. Below the gate voltage vs time and drain current vs time graphs, schematic representations of the respective electrochemical devices are depicted. Part of this figure is reproduced from reference[84].

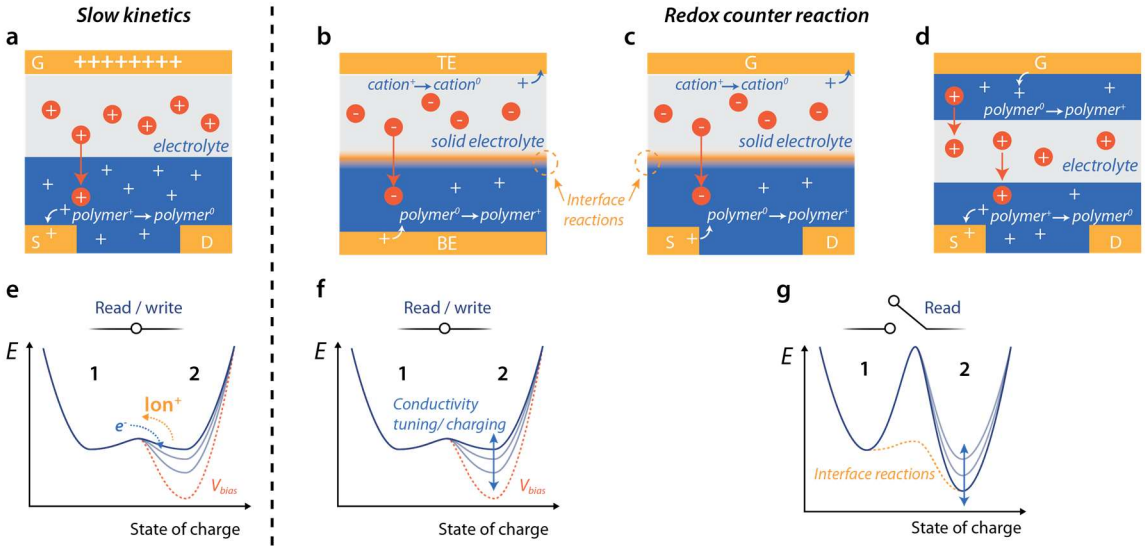

Figure 3. Non-volatility in electrolyte-gated redox-based neuromorphic devices. (**a**) electrochemical transistor-based devices rely on slow kinetics to retain a conductance state. (**b-d**) redox counter reaction-based (battery-like) devices rely on a counter reaction in either the electrolyte in a two-terminal configuration (**b**) or three-terminal gated configuration (**c**) or comprising a conducting polymeric gate (**d**) to ensure electrical neutrality and enhance stability. (**e-g**) related energy versus the state of charge. (**e**) In electrochemical transistors a bias will change the electrochemical potential of the polymer films (dashed orange traces in e-f panels) but offers little stability due to the low energy barrier for self-discharge. (**f**) In two-terminal redox counter reaction-based mechanisms depicted in b, the write and consequently, the read action, is similar to conventional electrochemical transistors but state-retention is enhanced when no voltage is applied. Interface reactions can introduce local self-discharge (**g**). In three-terminal gated electrochemical devices (c and d), the read action is decoupled from write action (similar to f) which prevents self-discharge via a large energetic barrier (g). However, without appropriate separation of the electrodes involved in the redox reaction, for example by an electrolyte, undesired interface reactions (dashed orange trace) lower the barrier for local self-discharging. The battery-like structure depicted in (d) is therefore expected to have the longest state-retention due to the electrolyte separator and large energetic barrier for self-discharge.

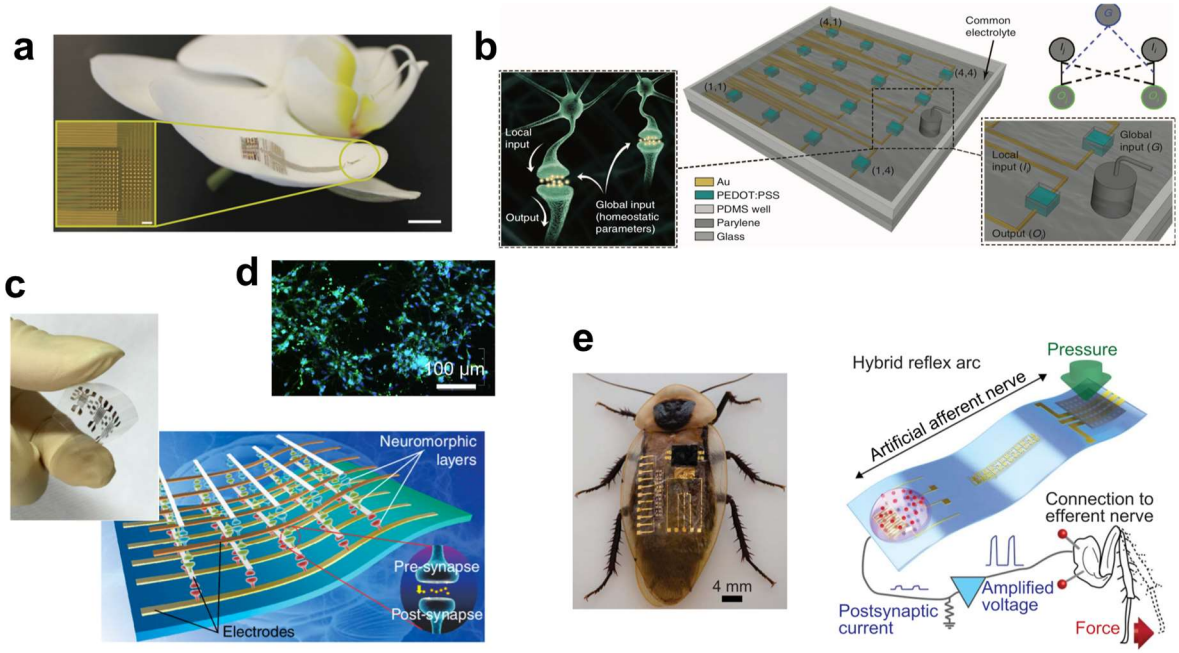

Figure 4. Examples of integration and functionality. (**a**) Flexible implant structure conforms to the surface of an orchid petal (scale bar, 5 mm). Inset, optical micrograph of a 256-electrode NeuroGrid (scale bar, 100 µm). PEDOT:PSS covered electrodes are 10 × 10 µm^2. From reference[110] (**b**) Global gate induced effects on an organic neuromorphic device array, from reference[108] (**c**) three-dimensional flexible synaptic array, from reference[94] (**d**) neurons on top of an organic memristive device array, from reference[87]. (**e**) artificial flexible afferent nerve connected to biological nerves in a cockroach, from reference[114].