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Organic Synapses for Neuromorphic Electronics: From Brain-Inspired Computing to Sensorimotor Nervetronics

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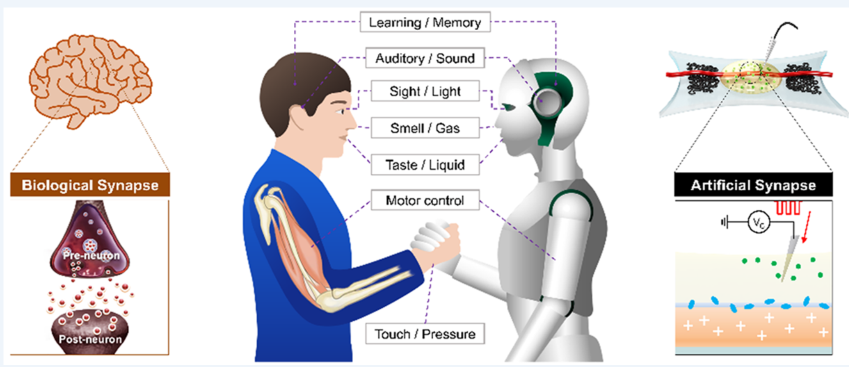

CONSPECTUS: Living organisms have a long evolutionary history that has provided them with functions and structures that enable them to survive in their environment. The goal of biomimetic technology is to emulate these traits of living things. Research in bioinspired electronics develops electronic sensors and motor systems that mimic biological sensory organs and motor systems and that are intended to be used in bioinspired applications such as humanoid robots, exoskeletons, and other devices that combine a living body and an electronic device. To develop bioinspired robotic and electronic devices that are compatible with the living body at the neuronal level and that are operated by mechanisms similar to those in a living body, researchers must develop biomimetic electronic sensors, motor systems, brains, and nerves.

Artificial organic synapses have emulated the brain's plasticity with much simpler structures and lower fabrication cost than neurons based on silicon circuits, and with smaller energy consumption than traditional von Neumann computing methods. Organic synapses are promising components of future neuromorphic systems. In this Account, we review recent research trends of neuromorphic systems based on organic synapses, then suggest research directions. We introduce the device structures and working mechanisms of reported organic synapses and the brain's plasticity, which are mainly imitated to demonstrate the learning and memory function of the organic synapses. We also introduce recent reports on sensory synapses and sensorimotor nervetronics that mimic biological sensory and motor nervous systems. Sensory nervetronics can be used to augment the sensory functions of the living body and to comprise the sensory systems of biomimetic robots. Organic synapses can also be used to control biological muscles and artificial muscles that have the same working mechanism as biological muscle. Motor nervetronics would impart life-like motion to bioinspired robots. Chemical approaches may provide insights to guide development of new organic materials, device structures, and working mechanisms to improve synaptic responses of organic neuromorphic systems. For example, organic synapses can be applied to electronic and robotic skins and bioimplantable medical devices that use mechanically stable, self-healing, and biocompatible organic materials. Biochemical approaches may expand the plasticity of the brain and nervous system. We expect that organic neuromorphic systems will be vital components in bioinspired robotic and electronic applications, including biocompatible neural prosthetics, exoskeletons, humanoid soft robots, and cybernetics devices that are integrated with biological and artificial organs.

1. INTRODUCTION

Bioinspired electronics is a technology to create functional electronic materials and sensory and robotic devices by emulating the structures and the functions of living things.[1−7] Development of future advanced bioinspired electronic devices and humanoid robots requires development of neuromorphic systems such as artificial brains for learning, calculating, and memorizing information and electronic sensorimotor nerves (sensorimotor nervetronics) for human-like sensory perception

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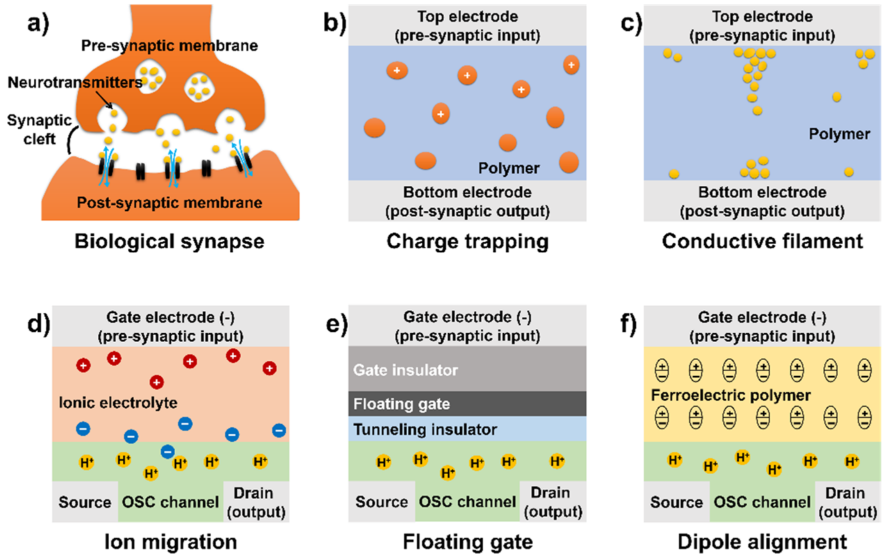

Figure 1. Schematic of (a) biological synapse, compared to organic artificial synapses with working mechanisms of (b) charge trapping, (c) conductive filament, (d) ion migration, (e) floating gate, and (f) dipole alignment.

and motor coordination by transmitting signals among sensory organs, the brain, and the motor organs.[1,8,9] Our brain and body use AC signals (action potentials), and their operations are affected by the rate at which action potentials are fired. Synaptic plasticity is important in learning, memory, sensation, and movement and is dependent on spike firing rate. For example, mechanoreceptors fire action potentials at rates that increase as pressure increases, and voluntary muscles contract with strengths that increase with the rate at which they are stimulated by action potentials.[1,8,9] Many existing bioinspired electronics and robotics use high-power operation or pneumatic/hydraulic systems that use DC signals,[5−7] but these devices have poor energy efficiency and are not readily compatible with integration in living systems. We believe that artificial synapse-based nervetronics would impart life-like computing, sensory detection, and movement to bioinspired robots and neuroprosthetics. We also believe that these traits can be obtained by mimicking the signal-transmission mechanism of biological neurons and their synaptic plasticity, such that the nervetronic devices respond to changes in the frequency and amplitude of action potentials. Devices with these abilities could change their response patterns for various durations; that is, they could "behave".

Organic synapses are promising components of neuromorphic electronic devices, including artificial brains and electronic sensorimotor nerves. Organic materials have many advantages such as applicability for both solution-printing and micropatterning or nanolithography processes, biocompatible mechanical properties that are desirable for future wearable and biocompatible electronics and robotics, and easy tuning of molecular properties. Organic synapses have simple device configuration and small energy consumption that is comparable to that of a biological synapse (∼1−10 fJ/synaptic event).[10] These are advantageous traits compared to neurons based on silicon circuitry, in which synapses are composed of several transistors and capacitors with complicated circuit configuration that consume relatively large amounts of energy.[11,12] Conventional inorganic bistable resistive memory (memristor) has limitations in emulating various synaptic behaviors due to abrupt switching of conductance states, so researchers have developed alternative types of artificial synapses based on metal oxide semiconductors.[13,14] However, inorganic materials are rigid and therefore not compatible with use in future wearable neuromorphic electronics. Organic neuromorphic memory and organic sensorimotor nervetronics that exploit these advantages have been demonstrated, and this progress can be expected to spawn diverse research on bioinspired soft robots and neural prosthetics.

In this Account, we review recent studies of organic neuromorphic electronics based on organic artificial synapses, from artificial brains to sensorimotor nervetronics, and suggest some perspectives for future research on organic neuromorphic electronics.

2. BIOLOGICAL AND ARTIFICIAL SYNAPSES

The nervous system of vertebrates can be divided into the central nervous system (CNS, the brain and spinal cord) and the peripheral nervous system (PNS, the sensory and motor organs and nerves). The CNS and the PNS work together to control an animal's behavior and body mechanics. Therefore, studies that emulate the CNS and PNS can develop bioinspired electronics and robotics that are equipped with a neuromorphic system of an artificial brain and sensory and motor systems.

A human nervous system is composed of $\sim10^{12}$ neurons and $\sim10^{15}$ synapses and propagates the action potentials between neurons through synapses and eventually transmits information to organs. Biological synapses are of two types of synapses: electrical and chemical.[15] Electrical synapses feature direct, rapid, and bidirectional electrical neural signal transmission at gap junctions (distance ∼3.5 nm) between neurons that are tightly attached to each other and can produce relatively simple synaptic behaviors.[15] Chemical synapses use neurotransmitters to transport neural signals at synaptic clefts (distance ∼20 to 40 nm) between neurons (Figure 1a).[15] Chemical synapses can conduct more complicated signal transmission than electrical synapses and can control synaptic strength precisely during learning and memory processes.[15] Neurotransmitters are released from vesicles in presynaptic membranes, then spread across the synaptic cleft and bind to receptors on the

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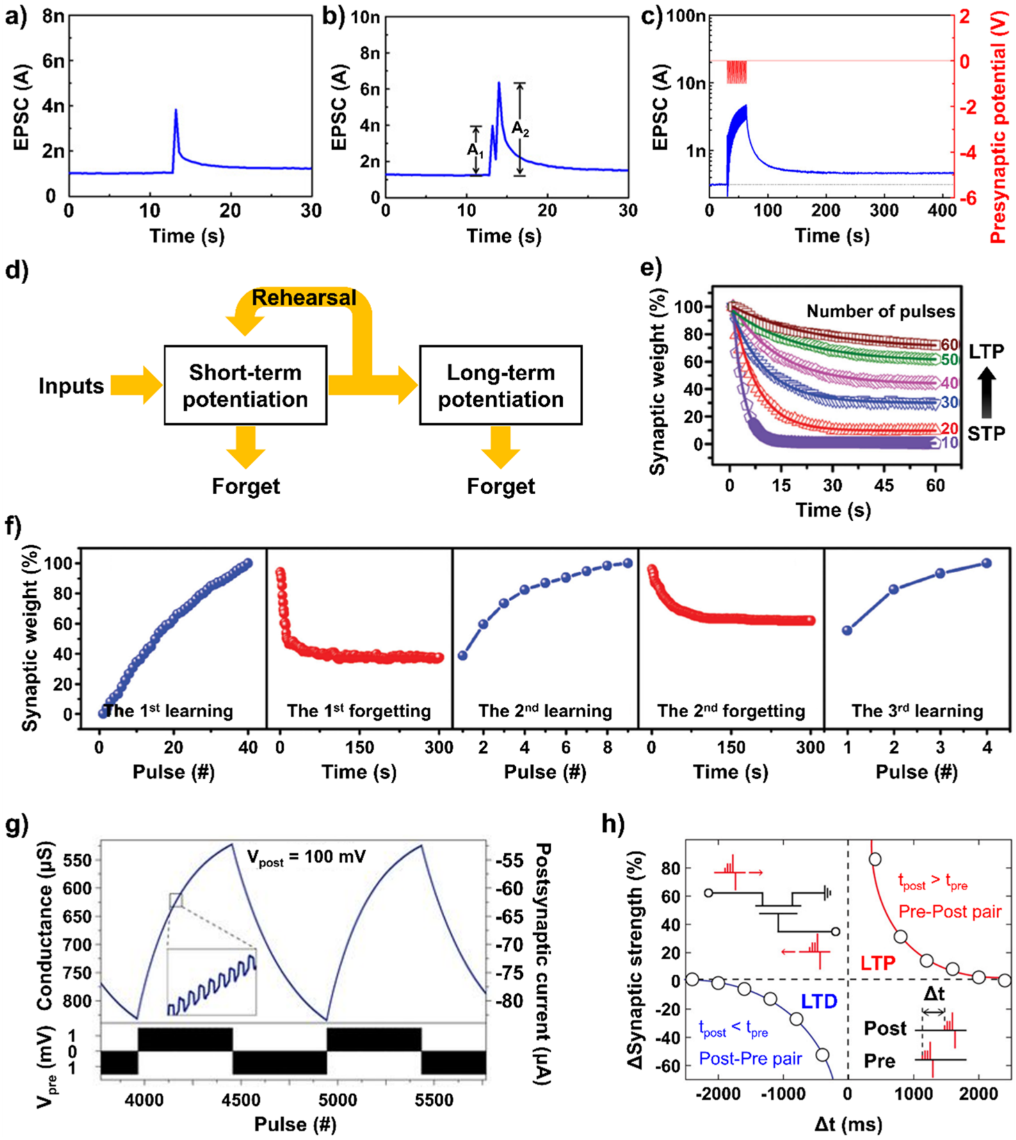

Figure 2. Synaptic responses of organic synapses. (a) Excitatory postsynaptic current (EPSC). (b) Paired-pulse facilitation (PPF, A_2/A_1). (c) Long-term potentiation (LTP) triggered by 30 spikes. Panels a–c reproduced with permission from ref 10. Copyright 2016 American Association for the Advancement of Science. (d) Schematic of STP–LTP transition. (e) Retention curves for synaptic weight after different number of applied pulses, showing STP–LTP transition. (f) Learning–forgetting–relearning behavior. Panels d–f reproduced with permission from ref 22. Copyright 2016 Wiley. (g) The potentiation and depression of synaptic weight. Reproduced with permission from ref 26. Copyright 2017 Nature Publishing Group. (h) Spike-timing-dependent plasticity (STDP). Reproduced with permission from ref 31. Copyright 2019 American Chemical Society.

postsynaptic membrane.[15] This process triggers ion channels to open and to conduct ions through the postsynaptic membrane. As a result, the membrane potential changes. If the synapse is excitatory, the membrane is depolarized, responds to the stimulus, and relays it to the next neuron. If the synapse is inhibitory, the membrane is hyperpolarized and suppresses relay of the signal. These types of synapses control synaptic activity appropriately[15] and result in phenomena such as memory, learning, cognition, emotion, action, homeostasis in the brain, generation and transmission of signals in sensory organs, and muscle activation in motor organs.

Researchers are trying to mimic the signal transmission mechanism and various kinds of synaptic plasticity of biological synapses in the brain. The goal is to demonstrate neuromorphic electronics that precisely control synaptic functions related to learning and memory. Organic synapses can be used to emulate the neurotransmission mechanisms and functions of biological synapses to achieve a neuromorphic system. By applying the presynaptic signals to an organic artificial synapse, the conductivity of active materials of the organic synapse is changed by various mechanisms such as charge trapping and ion movement. As a result, the device generates postsynaptic electrical responses of excitatory or inhibitory postsynaptic current, which are related to synaptic potentiation or depression, respectively. Depending on the presynaptic stimulation, organic artificial synapses can emulate the brain's short-term memory, long-term memory, spike-dependent plasticity, Hebbian learning, and pattern learning.

Studies have also been conducted on sensorimotor nervetronics composed of artificial sensory and motor organs and organic nerves and synapses that mimic the sensory and motor nervous systems of the human body.[8,9] Artificial synapses have been achieved by mimicking the mechanism of chemical synapses, because they are the locations at which

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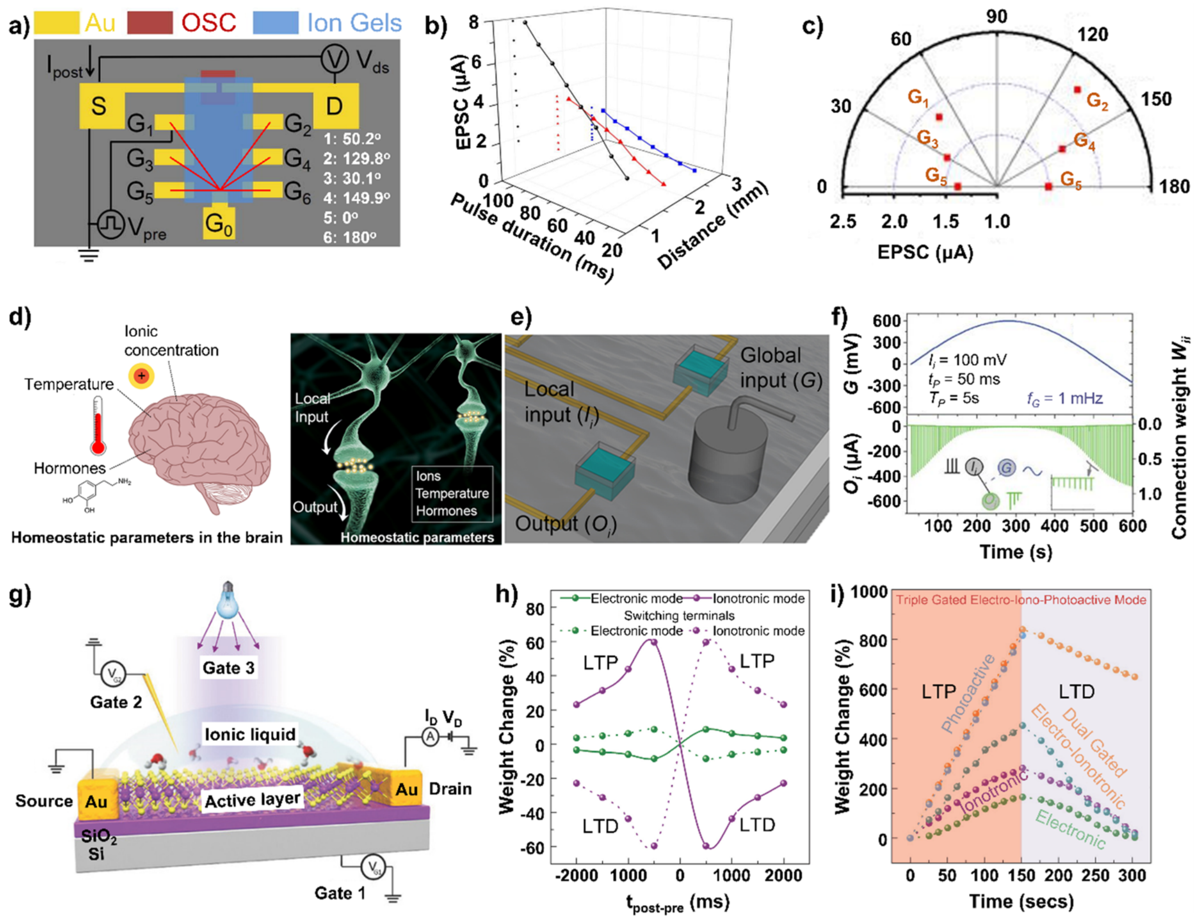

Figure 3. Examples of organic artificial synapses with diverse functionalities. (a) Multigate organic synaptic transistor and spatial orientations of gate electrodes (G_1 to G_6) defined by angles with respect to common gate electrode (G_0). (b) Spatiotemporal synaptic response in a multigate organic synaptic transistor at different pulse durations and distances between gate electrodes and OSC channel. (c) Diagram of EPSCs with input pulses from different spatial orientations. Panels a−c reproduced with permission from ref 23. Copyright 2017 AIP Publishing. (d) Schematic of homeostatic parameters. (e) Organic synaptic transistor array with global gate with a local input, I_i, a local output, O_i, and a global input, G. Panels d and e reproduced with permission from ref 32. Copyright 2018 Materials Research Society. (f) Modulation of output according to sine wave global input (frequency f_G = 1 mHz, G = ±600 mV) with constant local input (I_i = 100 mV, pulse width t_P = 50 ms, pulse period T_P = 5 s). Reproduced with permission from ref 25. Copyright 2017 Nature Publishing Group. (g) Triple-gated (electronic−ionotronic−photoactive) organic synaptic transistor. (h) Spike-timing-dependent plasticity (STDP) and reverse STDP with the electronic- and ionotronic-mode operations. (i) Potentiation and depression controlled by multimode stimulation. Panels g−i reproduced with permission from ref 33. Copyright 2018 Wiley.

sensory signals are integrated and muscle contractions are mediated.[8,9] This sensorimotor nervetronics might be used to support sensory and motor functions of living things (neuroprosthetics) by connecting to the living cells and to construct artificial sensory and motor systems of a bioinspired robot and exoskeleton to assist human beings.

Until now, researchers have focused on emulating chemical synapses, but interest in electrical synapses is certain to increase in the near future and will expand the functions and increase the efficiency of neuromorphic electronics.

3. ORGANIC SYNAPSE FOR BRAIN-INSPIRED COMPUTING

Organic synapses have been demonstrated with two-terminal (2T) and three-terminal (3T) geometries. In 2T geometry, the active layer is sandwiched between two electrodes; a presynaptic spike applied to one electrode (a presynapse) induces a change in conductivity (1/resistivity) of the active layer, and this change is read by the other electrode (a postsynapse). In 3T geometry, a presynaptic signal is generally applied to a gate electrode (a presynapse) and the change in conductivity of a semiconducting channel (a postsynapse) between the source and drain electrodes is measured. The geometry and working mechanism of organic synapses are much simpler and more straightforward than conventional silicon neuronal circuits.[12,16,17]

3.1. Operation Mechanisms and Synaptic Behaviors

Several mechanisms of organic synapses have been reported when an electrical pulse is applied to the presynaptic electrode; these include (i) charge trapping in which the charges are trapped at the conducting additives or organic semiconductor (OSC)/insulator interface (Figure 1b),[18,19] (ii) ion-diffusive conductive filament, in which metal ions diffuse into the organic layer and form a conducting filament pathway (Figure 1c),[20,21] (iii) ion migration (electric double layer, EDL) and electrochemical (ion penetration and redox) reaction of the ionic electrolytes and ion-permeable conjugated polymers, in which EDLs cause temporary accumulation of carriers in the semiconductor channels and electrochemical doping induces higher carrier concentration that is maintained longer than

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EDLs (Figure 1d),[8−11,16,26,27] (iv) floating-gate effect, in which carriers tunnel from a channel to a floating gate and become trapped there (Figure 1e),[28,29] and (v) dipole alignment of organic ferroelectric materials, in which gradually changed alignment of dipoles by applying voltage can control the height of a charge injection barrier in 2T structure and can control the gate-induced electric field in 3T structure (Figure 1f).[30,31] These mechanisms induce change in the conductivity of the organic active layer and result in postsynaptic responses.

To emulate learning and memory functions of brain, artificial synapses generally demonstrate several characteristics of a brain's plasticity. A single short presynaptic voltage spike induces a sharp increase in current (excitatory postsynaptic current, EPSC) to a level, A_1, which then decays rapidly back to its initial value; this trend is STP (Figure 2a).[10] When the next voltage pulse is applied within a short interval before the decay of EPSC is complete, the current, A_2, is higher than A_1; this phenomenon is PPF, and its strength is calculated as A_2/A_1 (Figure 2b).[10] This neural facilitation depends on the interval between the pulses and is involved in simple learning, synaptic filtering, and sound-source localization.[10] Long-term potentiation (LTP) is a response to stimulation by a large number of presynaptic action potentials at high rate (Figure 2c).[10] As the number of presynaptic spikes applied to the organic synapse increases, the behavior changes from STP to LTP (Figure 2d).[22] The synaptic weight is increased, then decays slowly to a certain state, at which it remains for a long time (Figure 2e).[22] During this time, signals can be reinforced (retrained) with a smaller number of stimuli than was originally necessary (the brain's learning−forgetting−relearning process) (Figure 2f).[20−22,28] For example, application of 40 consecutive spikes (learning) induced long-term memory, but the synaptic weight decayed spontaneously over time, like a brain forgetting (Figure 2f).[22] After the first stage, 9 (at the second learning) and 4 (at the third learning) pulses are sufficient to restore the same synaptic weight (Figure 2f),[22] so artificial synapses can mimic the "learning-experience" behavior of human beings.

Potentiation and depression of the postsynaptic response are involved in learning and forgetting functions of the artificial brain, and multilevel memory states can be demonstrated; for multibit memory and computation devices, the storage steps should be linearly controlled. Depending on the polarity of the applied voltage, hundred-step potentiation and depression of the synaptic weights could be controlled almost linearly (Figure 2g).[26] Also, several kinds of spike-dependent plasticity such as spike-rate-dependent plasticity, spike-voltage-dependent plasticity, spike-duration-dependent plasticity, and spike-timing-dependent plasticity (STDP) (Figure 2h)[31] have been demonstrated; these are related to the learning rules of the brain. All these characteristics have been the main synaptic responses of organic synapses that emulate the brain's plasticity, and more characteristics will probably be realized as knowledge expands in this field.

3.2. Expanding Device Functionalities

Artificial synapses have the expandability of functionalities. A biological neuron has synaptic junctions with several preneurons; it receives many signals with various spatial and temporal features and integrates them. Synaptic transistors composed of multigate electrodes can take synaptic signals from multiple preneurons and process spatiotemporally correlated synaptic signals (Figure 3a−c).[10,16,23] Spatiotemporal dendritic integration has been demonstrated using multiple gate electrodes with different special orientations and distances between gate electrodes and OSC channel (Figure 3b,c).[23] These functions will be important in constructing an artificial neural network and in processing neural information.[23,24]

The overall activity of large neural networks in the brain is regulated by global parameters such as temperature and hormonal/ionic concentration, in a process called homeostatic regulation (Figure 3d).[25,32] A synaptic transistor array with one global gate electrode and multiple channels can demonstrate this homeoplasticity function by using the electrolytes as global parameters to control the whole set of channels (Figure 3e).[25,32] By controlling the global input, the overall synaptic weight of neural networks is adjusted (Figure 3f).[25,32] This global regulation would provide simplified neural network architectures without requiring complex interconnections between individual neurons or additional functions to manipulate synaptic weights.[25,32]

Moreover, a recently reported triple-gated (electronic−ionotronic−photoactive) artificial synapse shows expandability, although it is not based on an organic active layer (Figure 3g).[33] Three operating modes provide additive/subtractive modulations of synaptic weight, with various forms of STDP, and a learning rule that depends on temporal correlations between the pre- and postsynaptic spikes through positive or negative feedback loops to each other (Figure 3h,i).[33] This multifunctional approach can also be applied to organic synapses; it would facilitate dynamic tuning of synaptic weights with higher modulability and homeostatic stability than are currently available, and may enable high-density device integration.[33]

Research on organic synapses that emulate the computational and memory functions of the brain has been active but to date has focused on development of unit devices based on new materials and on development of structures to demonstrate the synaptic responses. For practical applications, a system-level implementation is required, and mechanical flexibility and stretchability will improve biocompatibility and enable the development of wearable and bioimplantable devices. Furthermore, by integrating with external components, it can be applied to demonstrate nervetronics in conjunction with sensory and motor systems.

4. ORGANIC SENSORY SYNAPSES

The sensory nervous system connects sensory organs and the CNS, detects external stimuli from sensory receptors, and transmits sensed signals to the brain. A sensory system composed of artificial sensory organs and artificial synapse recognizes stimuli, then generates a postsynaptic response in the same way as biological sensory receptors and synapses. Then synaptic response would be transferred to the brain or the operative organ, so that learning and cognitive functions and the sensory system of the future electronic device can be developed. Sensory synapses can be configured in several ways, including (1) direct stimulation of stimuli-sensitive artificial synapses and (2) connection of an external sensor and an electronic component to an artificial synapse. In later configurations, the generated presynaptic electrical signals from the stimulated sensor are transmitted to the artificial synapse and induce postsynaptic responses.

4.1. Light-Sensory Synapses

Light-sensory synapses have been demonstrated with organic synapses.[34−36] A single-unit organic phototransistor composed

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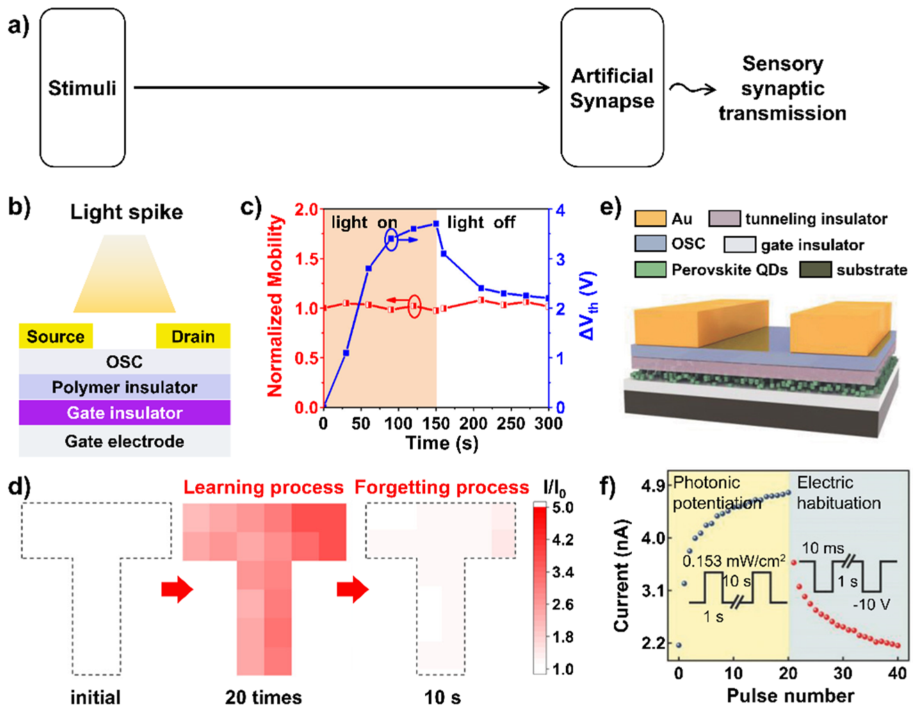

Figure 4. Examples of light-sensory organic synapses. (a) Overall configuration of direct stimulation of a stimuli-sensitive artificial synapse. (b) Light-sensory organic synaptic transistor with interface charge trapping. (c) Threshold voltage changes, ΔV_{th}, and normalized carrier mobility of light-sensory organic synaptic transistor with different UV light exposure times. (d) Dynamic learning and forgetting process of light-sensory organic synaptic transistors array. Panels b−d reproduced with permission from ref 34. Copyright 2018 American Chemical Society. (e) Light-sensory organic synaptic transistor with floating gate. (f) Potentiation and depression controlled by photonic pulses and electrical pulses, respectively. Panels e and f reproduced with permission from ref 35. Copyright 2018 Wiley.

of an OSC that absorbs light and forms light-induced electron−hole pairs and a charge trapping polymer dielectric material that has strong electron-withdrawing groups showed light-triggered synaptic responses as a result of charge trapping at the OSC/dielectric interface where photoinduced carriers of the OSC are trapped (Figure 4a,b).[34] Illumination causes gradual filling of the charge-trapping states at the OSC/dielectric interface; and the threshold voltage, V_{th}, of the transistor gradually shifts positively (Figure 4c).[34] When the light is turned off, the holes are retrapped and the V_{th} returns to its initial state.[34] Thus, a short-term EPSC occurs; irradiation increases the current, which then gradually decreases to the original value in darkness.[34] Moreover, a T-shaped light-sensory synaptic transistor array was fabricated; it showed the functions of dynamic learning and forgetting (Figure 4d).[34] This device proves the feasibility of using light-sensitive synapse arrays for image-sensing and pattern-learning functions in a light-sensory system of artificial brain.

Floating gate organic transistors also can be used to realize light-sensitive synapses by using a photosensitive material as the floating gate. A photoactive layer composed of perovskite quantum dots (QDs) sandwiched between two dielectric layers (tunneling insulator and gate insulator) absorbs light and generates photoinduced charges, which increase hole accumulation in an OSC on the tunneling insulator (Figure 4e).[35] Illumination of the perovskite generates hole−electron pairs, which separate due to a bending of the energy band of OSC/tunneling insulator/perovskite film; the holes move to the OSC, whereas the electrons remain trapped in the perovskite.[35] The trapped electrons form additional internal fields, so hole transport in the OSC is further facilitated, and the V_{th} shifts positively.[35] The captured charge carriers remain in the potential well for a long time after the light is turned off; the result is long-term memory retention.[35] Then when gate voltage, V_G, is applied, the holes are retrapped, so V_{th} returns to its original state and the memory is erased.[35] The device with photoprogrammable and electrical-erasable characteristics shows synaptic plasticity as a result of partial trapping and detrapping of carriers under pulsed irradiation with various wavelengths, intensities, and durations, and demonstrated learning and forgetting functions (Figure 4f).[35]

When light-sensory synaptic devices are used for image learning and pattern learning, they need learning and memory functions that emulate the brain's plasticity including long-term memory retention. To mimic the function of a light-sensory receptor, the synaptic device must have fast response and fine temporal resolution, so short-term plasticity and fast decay at the biological-synapse level would be important. Also, light-sensory synapses would be able to distinguish light wavelength (i.e., color) and yield a light-controlled actuator that mimics a biological retina. Artificial synaptic retinas could also detect infrared and ultraviolet ranges. By matching synaptic signals of a multipixel artificial synaptic retina with signals of degenerative biological retinal nerve cells, nerves, or the visual cortex, such a device might be used to restore impaired vision. Light-sensing synapses will certainly have a wide range of applications.

4.2. Pressure-Sensory Synapses

An artificial tactile nervous system (including pressure-sensory synapse) that combines an artificial tactile sensor with an artificial synapse that simulates tactile nerves of the skin can be used to realize an e-skin system that achieves bionic touch perception (Figure 5a).[37]

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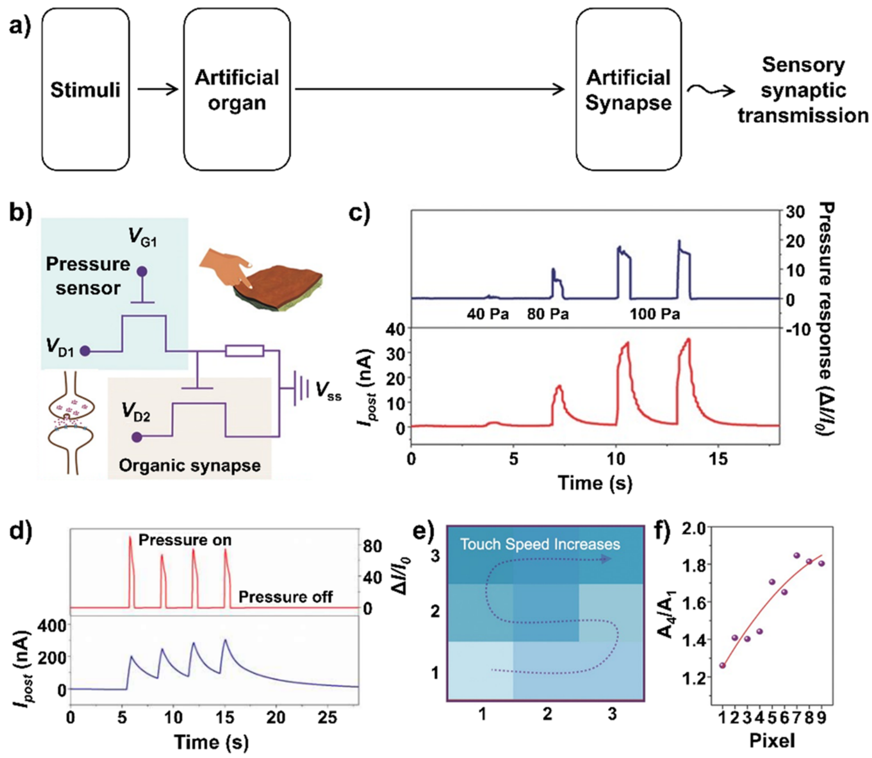

Figure 5. Examples of pressure-sensory organic synapses. (a) Overall configuration of connection of an external sensor and an artificial synapse. (b) Pressure-sensory organic synaptic transistor. Current change in pressure sensor and postsynaptic current, I_{post}, of synaptic transistor under (c) different pressures and (d) different numbers of stimuli. (e) Schematic of a 3 × 3 pixel synaptic transistor array and (f) EPSC gain, A_4/A_1 for each pixel with increasing repeated touch speed from pixel 1 to 9. Reproduced with permission from ref 37. Copyright 2017 Wiley.

The function of tactile perception systems has been emulated by integrating a pressure-sensitive organic transistor and an organic synaptic transistor (Figure 5b).[37] In this device, the capacitance of the insulator in the pressure-sensitive organic transistor is changed by applied pressure, so the output current of the synaptic transistor changes.[37] This response matched well with the signal sensing, transduction, and processing functions of touch perception by human beings. As the strength of the tactile stimulus (the intensity, touch duration, and number of the pressure events) was increased, the synaptic weight was increased (Figure 5c,d).[37] Also, a 3 × 3 pixel synaptic transistor array was fabricated to mimic the tactile perception of position-resolved dynamic mechanical contact (Figure 5e).[37] As the frequency of repeated touches increased, EPSC gain (A_4/A_1) increased as a result of synaptic potentiation (Figure 5f). The tactile-perception system would enable use of pressure to encode and sense information.

Development of artificial synapses that can detect stimuli that biological sensory organs cannot detect (e.g., infrasound or ultrasound, ionizing radiation, chemical signatures of gases or liquids) would enable development of transhuman sensing functions that would have practical applications in numerous fields, including military and aerospace.

5. ORGANIC SYNAPSES FOR SENSORIMOTOR NERVETRONICS

Sensory and motor nervous systems respond to stimuli from sensory nerves and transmit signals from the CNS to the somatic motor system, which is composed of voluntary skeletal muscles. Sensorimotor nervetronics is the conjunction of artificial synapse and artificial or biological sensory and motor systems to produce movement reactions (reflex arc and spike-dependent muscle contraction of twitch summation and tetanus) that are similar to those of a living body[8,9] and that will facilitate production of soft robots, neuroprosthetics, and exoskeletons that are capable of natural movement. Little research has considered sensorimotor nervetronics, despite its obvious usefulness for future electronic and robotic devices.

5.1. Light Sensorimotor Nervetronics

A light-sensitive artificial synapse based on the charge-trapping effect is advantageous in that it can be implemented as a single unit with a common organic transistor structure, but large driving voltages have been required in some cases. For example, a phototransistor composed of polymer dielectric−OSC with interfacial trapping effect required applied voltages of V_G = −40 V and drain voltage V_D = −60 V,[36] and a device with a floating gate of perovskite QDs used V_G = −20 V to erase memory states.[35]

The operation voltages of sensory synapses can be reduced by adopting a configuration that uses a self-powered sensor that detects light signals then produces output that stimulates an organic synapse. A self-powered photodetector is used as an artificial light receptor, and the output voltage generated from the photodetector by light stimulation is applied to the ion gel-gated artificial synapse as a presynaptic spike (Figure 6a,b).[8] As the number and frequency of light pulses increased, the EPSCs were amplified by increased ion migration from the ion-gel gate dielectric toward the OSC and thus induced hole accumulation inside the channel in the ion gel-gated transistor (Figure 6c,d).[8] These synaptic behaviors are analogous to activation responses in biological skeletal muscle.[8,38] The biological skeletal muscle contracts according to the action potentials received from the motor neuron through a chemical

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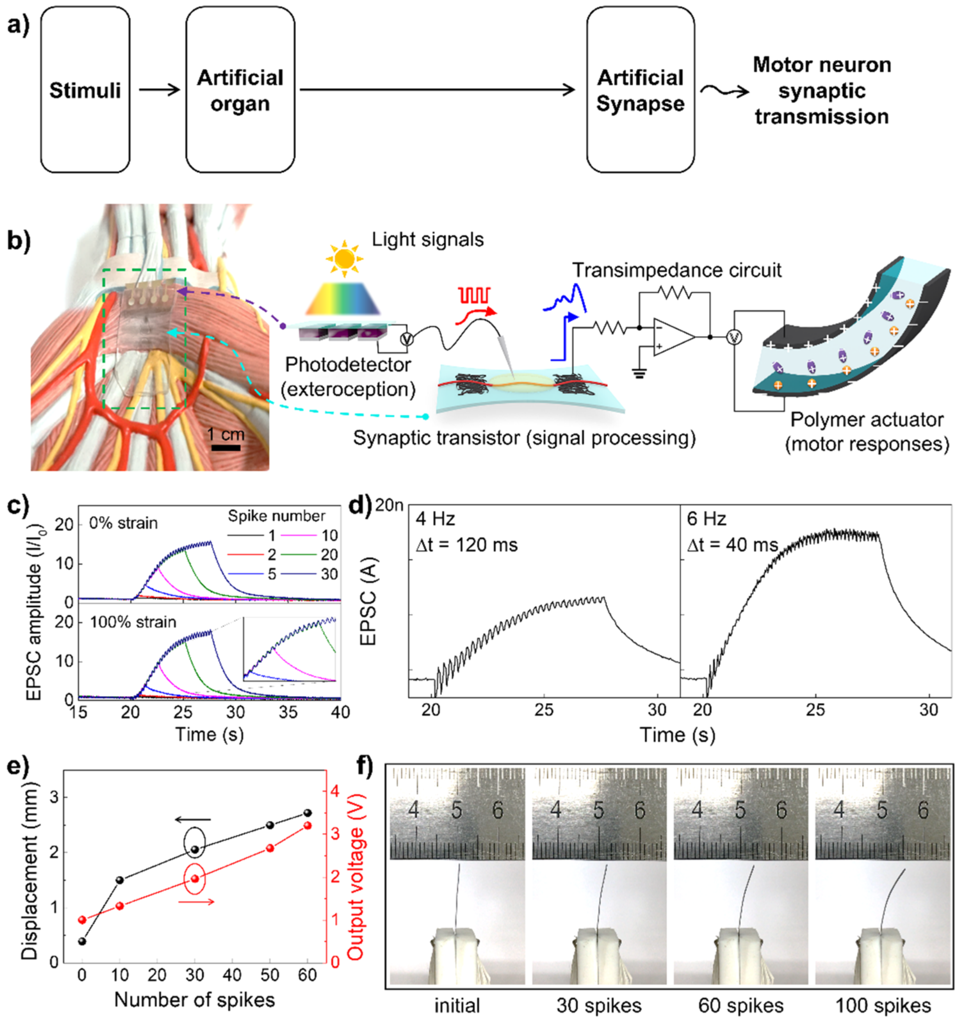

Figure 6. Organic synapses for light-sensitive sensorimotor nervetronics. (a) Overall configuration of connection of an external sensor, an artificial synapse, and a motor unit. (b) Schematics of light-sensitive sensorimotor nervetronics. (c) Spike-number-dependent EPSC amplitude and (d) spike frequency-dependent EPSC of light-sensitive sensorimotor nervetronics. (e) Output voltage from synaptic transistor and displacement of an artificial muscle fiber (a polymer actuator) according to number of presynaptic spikes. (f) A photograph of a polymer actuator according to the presynaptic spikes. Reproduced with permission from ref 8. Copyright 2018 American Association for the Advancement of Science.

synapse that is called a neuromuscular junction.[8,38] The intensity of a muscle contraction increases as the frequency of action potentials increases.[8,38] Motor nervetronics composed of artificial neuromuscular junctions of organic synapse and an artificial muscle can mimic this contraction response of biological muscles to activate the artificial muscle by receiving presynaptic action potentials from a light-sensory device and transferring postsynaptic signals to an artificial muscle fiber (a polymer actuator) (Figure 6b).[8] Postsynaptic output voltage and actuation of artificial muscle fiber were controlled by the presynaptic action potentials and showed contraction responses that were similar to those of real skeletal muscle (Figure 6e,f).[8]

5.2. Pressure Sensorimotor Nervetronics

A synaptic response in an artificial sensory synapse can be induced by controlling the number and frequency of stimuli, but this approach does not exactly emulate the working principle of a biological sensory nerve.[1] In response to a stimulus, a biological neuron fires action potentials at frequencies that depend on the intensity of the stimulus; that is, they transduce stimuli to action potentials; this ability is desirable in an artificial neuron.[1] Pyramidal pressure sensors, organic ring oscillators, and flexible artificial synapses in pressure-sensory nervetronics are analogous to mechanoreceptors, sensory neurons, and synapses in a biological afferent nerve, respectively (Figure 7a,b).[9]

An artificial mechanoreceptor that used pyramidal piezoresistive pressure sensors showed similar pressure-detecting range (1−80 kPa) to that of biological mechanoreceptors (Figure 7c).[9] Output signals from the pressure sensor were applied to an organic ring oscillator (artificial neurons) to generate oscillating signals of artificial action potential at frequencies similar to those of biological neurons (0−100 Hz) (Figure 7c).[9] Increase in pressure applied to the sensor elicits increase in the frequency of artificial action potentials that the artificial neuron emits; this response is similar to the biological system (Figure 7c).[9] This similarity of stimulus-detecting range, action potential firing range, and signal transmission mechanism is quite important for development of neural prosthetics that integrate artificial nerves with a living body. When the generated presynaptic action potential pulses are transferred to an artificial organic synapse, they induce postsynaptic EPSC responses.[9] Compared to conventional

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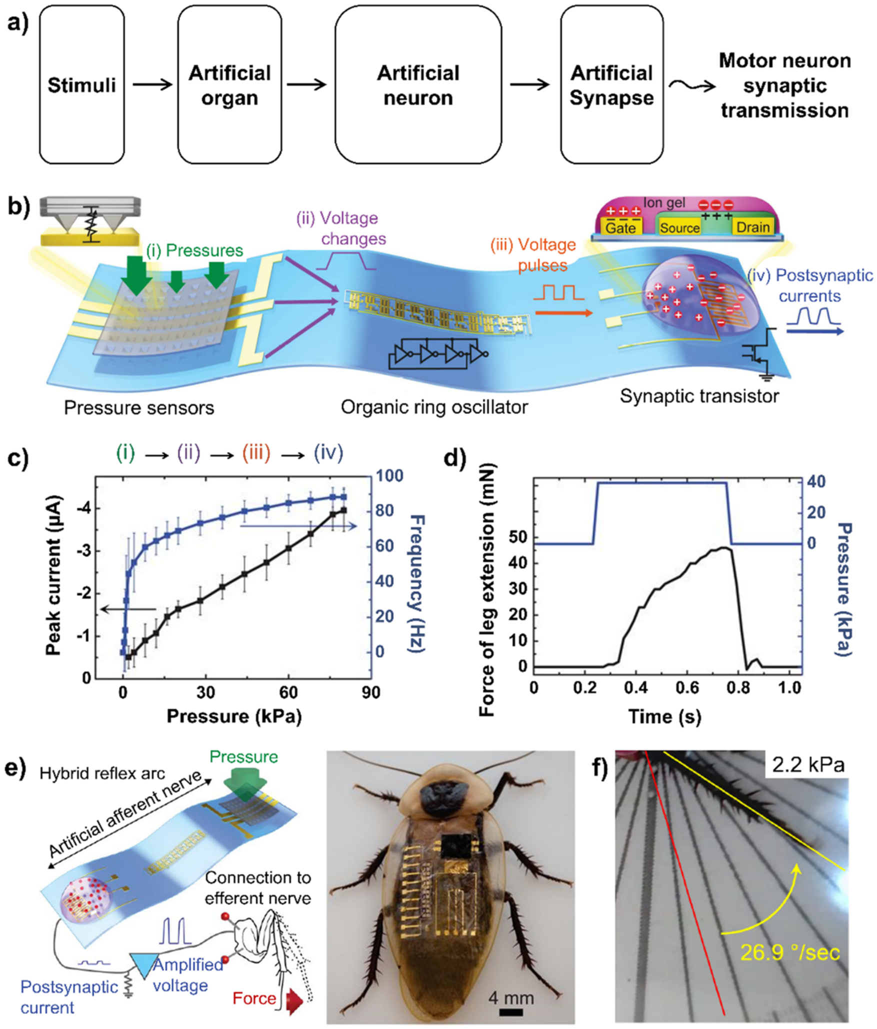

Figure 7. Organic synapses for pressure-sensory sensorimotor nervetronics. (a) Overall configuration of connection of an external sensor, an artificial neuron, an artificial synapse, and a motor unit. (b) Schematics of an artificial mechanosensory afferent nerve system. (c) Oscillating frequencies of ring oscillator depending on pressures and resultant postsynaptic peak current of synaptic transistor. (d) Contraction force of leg extension in response to pressure (39.8 kPa and 0.5 s) on the artificial afferent nerve. (e) Schematics and a photograph of hybrid reflex arc made of an artificial afferent nerve and a biological efferent nerve. (f) A photograph of movement of the cockroach leg after pressures onto the artificial afferent nerve. Reproduced with permission from ref 9. Copyright 2018 American Association for the Advancement of Science.

complex silicon circuits, the organic synapses simply mimic the decay time of the postsynapse similar to the biological synapse.[12] In this way, organic nervetronics can process tactile information by using a mechanism similar to that used by biological tactile neurons.[9] The postsynaptic current was affected by the magnitude and the duration of the pressure (Figure 7c).[9]

An artificial reflex arc has been demonstrated using sensory and motor nervetronics based on organic synapses (Figure 7e). The postsynaptic electrical signal from pressure-sensory nervetronics was used to stimulate the biological motor nerves in a cockroach leg to make it move (Figure 7f).[9] The angular movement and leg extension force were changed according to the intensity and duration of the pressure (Figure 7d); that is, this work hybridized an artificial sensory neuron and a biological motor organ.[9]

These sensorimotor nervetronics will enable development of life-like natural movement of future soft robots, exoskeletons, and neural prosthetics. It may also be applicable to biomedical implantable devices that assist in the movement of voluntary or involuntary muscles.

6. SUMMARY AND OUTLOOK

In this Account, we have reviewed recent trends in research on organic neuromorphic systems based on organic synapses. In addition to bioinspired sensors and motor systems, an organic neuromorphic system may also be a critical component of bioinspired robotics and electronics. Organic synapses with simple device structures provide an alternative to conventional von Neumann computing architecture and silicon neuron circuitry. Organic synapses also have the advantages of low fabrication cost and low energy consumption. They have demonstrated a variety of learning and memory functions that mimic the brain's plasticity, and thereby prove the feasibility of development of artificial brains based on organic synapses.

Sensory synapses based on organic synapses recognize various external stimuli and form and transmit neural signals by a sensing mechanism similar to that of biological sensory nerves. Sensorimotor nervetronics based on organic synapses transmit sensory neural signals to living or artificial motor nerves and elicit motor reactions in muscles in the same way that biological sensory and motor nervous systems react.

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Studies of nervetronics based on various organic materials and device structures, and studies of system-level integration will continue. Various biomimetic electronic devices including neural prostheses, exoskeletons, and soft robots will be developed based on organic synapses connected with a diversity of living and artificial organs. Furthermore, spike neural network (SNN)-based nervetronics must be considered when designing biomimetic robotic and electronic applications to enable interaction by signal transmission among neural networks (i.e., information processing and signal integration) and sensory and motor parts that are driven by neural spikes. Future bionic neural prosthetics based on organic neuromorphic electronics will require precise matching of signals between artificial and biological nervous systems. Comprehensive understanding of biological organs and nervous system including key cells and detailed patterns of neural activity (e.g., amplitude, firing rate, and plasticity) is essential. Although many neuromorphic electronics and neural prosthetics based on conventional software algorithms and silicon neuronal circuits composed of several transistors and capacitors have been developed, organic artificial synapses are promising because even a single artificial synapse with simple device configuration can generate every form of synaptic plasticity with high energy efficiency and is compatible with biological tissues, organs, skin, and in the future, even the brain. The mechanical stability, self-healing ability, and biocompatibility of organic materials will give organic synapses applications in electronic and robotic skins and biomedical devices and will therefore contribute to the development of humanoid robots, bioimplantable neural prosthetics, and cybernetic electronics.

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