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The Rise of Bioinspired Ionotronics

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Ionotronics is a concept which provides powerful tools and methods for narrowing the gap between conventional electronics and biological systems. Translational implementation of ionotronics gives birth to a wide range of novel and exciting paradigm breakers, which greatly innovates the development of the bio–technology interface. Herein, new trends and features of bioinspired ionotronics are presented, first introducing the basic mechanisms of ionotronic modulation and the essential building blocks. Despite its infancy, bioinspired ionotronics exhibit an unprecedented potential to advance a broad spectrum of applications such as robotics, neuromorphic engineering, and biosensing and readouts. Challenges and perspectives for the full exploitation of this concept for practical applications are also addressed. Leveraging bioinspired ionotronics shapes the interaction between human and machines in the future.

1. Introduction

Ionotronics is a concept that exploits the properties of both mobile ions and electrons, and is the source of the components as the ionic diode, ion-triggered field effect transitions or even complete ionic logical circuits, to name a few. In the development of metal–oxide–semiconductor (CMOS)-based digital systems, ions' influence has been generally associated with negative effects such as high interface state density,[1,2] high leakage current,[3,4] unstable electrochemical processes,[5,6] etc. However, ionotronic approaches could revise ion functions in the electronic devices, allowing the coupling of electrons (typically used in artificial devices) with ions (on which biological systems are typically based). Ionotronics have been proposed as a potential bridge between living matter and electronic devices to create bioelectronic interfaces[7–10] and bioinspired robotics,[11–14] as well as a powerful approach exploiting ionic dynamics to advance next-generation memory[15,16] and even artificial intelligence hardware.[17–19]

Ionotronic-based systems can greatly propel the development of emerging bioinspired systems (**Figure 1**), and can ultimately innovate the interaction between humans and man-made machines. As an example, next-generation bioelectronic systems with direct contact to the living matter can enable bidirectional interfacing while showing mechanical conformability[20,21] and controlled biological properties.[22,23] Ionotronics has facilitated the development of tissue engineering, regenerative medicine, and drug delivery through ion transport-based processes.[24] In the future, through their integration with other advanced functional modules, ionotronic-based systems offer the potential to become an integral part of the body for functional restoration[25,26] or reinforcing existing functions.[27,28] Furthermore, the similar ionic dynamics shared by a group of ionotronic devices with natural synapses has allowed the development of artificial synapses toward novel neuromorphic networks.[17–19,29,30] It is certain that the ionotronic-based neuromorphic engineering will furthermore stimulate a wider range of potential applications by endowing them with artificial intelligence or the capability for biocompatible processing.

In this article, an attempt has been made to understand and describe some of the new trends and features of such bioinspired ionotronics (Figure 1), which are focused on the development of fully integrated and intelligent systems. The essential primary components of ionotronic systems are introduced and discussed along with the proposed underlying mechanisms. The recent progress on the potential applications which could serve as benchmarks and inspirations for further exploration are briefly reviewed. Finally, open challenges for possible future advances in this emerging field are also addressed.

2. Fundamentals of Ionotronic Modulation

Generally, the ionotronic fields and responses are operated through three types of processes: 1) electrochemical reactions, 2) electric-double-layer (EDL) formation, and 3) ion diffusion, as sketched in **Figure 2**. These ionotronic modulations observed in a wide range of materials like conjugated polymers,[31] hydrogels,[32] nonmetal oxides,[33] correlated oxides,[34] ferroelectric materials,[35] piezoelectric materials,[36] and nanocomposites[37] have greatly advanced the development of electrochemistry, photochemistry, catalysis, energy storage, information technologies,

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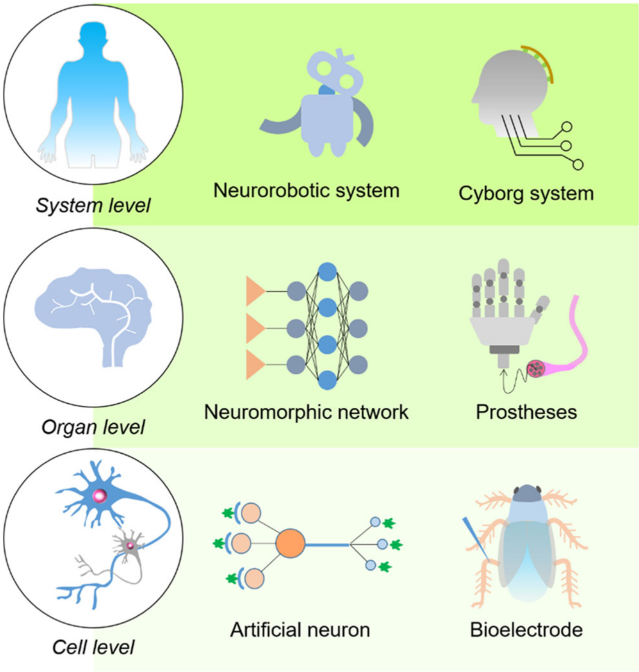

Figure 1. The concept of bioinspired ionotronics is sketched by examples with different complexities.

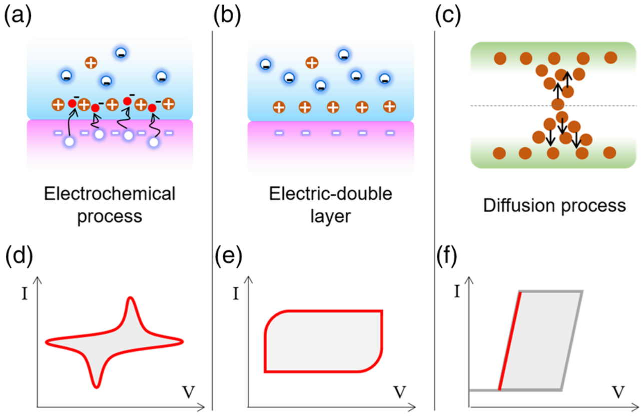

Figure 2. Fundamental ionotronic modulation processes: a) electrochemical process, (b) electric-double-layer (EDL) formation, and (c) diffusion process, respectively. Typical I–V characterizations of devices that involve in these processes: d) the faradaic processes in a pseudocapacitor, (e) the capacitive processes in double-layer capacitor, and (f) the diffusion processes in a diffusive memristor (the red curve indicates the spontaneous recover of the metallic conductive pathway), respectively.

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and the bioelectronics. Electrochemical processes are based on the transfer of electrons between two substances that usually induce a non-volatile change in the devices. Such processes generally occur under an external voltage or through the release of chemical energy, which involves electron transfer to or from a molecule/ion and changes the oxidation state of the molecule/ion. Contrarily, the formation of an EDL represents a transient and dynamic electrostatic process that leads to the formation of an ion- and electron-based field dipole at the interface between ionic and electronic conductors. Interestingly, EDLs exhibit often a high specific capacitance ($>1\ \mu\mathrm{F\ cm^{-2}}$), enabling strong capacitive coupling between the electrons and ions. Finally, the diffusion process refers to the ion movement that is generally observed when there is a concentration gradient and/or due to the minimization of interfacial energy, and such process always occurs after the removal of the external stimulation. Device function is always reflected in one more ionotronic modulation processes.

Electrochemical processes have been exploited in different research areas. For example, a battery-like artificial synapse was developed on a flexible plastic substrate.[38] In such a device, the protons triggered by the gate voltage can exchange between the three layers: gate, electrolyte (gate dielectric), and a channel. Subsequently, the protons are fixed in each layer inducing a stable channel conductance with a long retention time. Generally, in materials inherent with ionic and electronic conductivity, the migration of an ion can be involved in reduction and oxidation that electrochemically modified the electrical conductance. These have been widely exploited in several emerging non-volatile memory devices like the resistive random-access memory ((RRAM), often referred to as a memristor).[39,40] The memristor consists of two-terminal junctions that can be reversibly switched between a low- and a high resistive state by ion-triggered structural changes or redox processes. The memristor-based synapse can be created that relies on the electrochemical process or drift of ions to mimic the behaviors of biological synapse/neuron.[41–43] For instance, by exploiting mobile perchlorate ion-based electrochemical redox within the conjugated polymer (triphenylamine-containing polymer (BTPA-F)), history-dependent memristive behaviors were achieved to meet the fundamental requirements for mimicking the potentiation and depression processes of a biological synapse.[31] These are promising components for the development and construction of new hardware neural networks.[44–46] Finally, an electrochemical decision-maker based on hydrogen ion-conducting polymer (Nafion) has recently been reported with the ability to solve multiarmed bandit problems (MBPs), which clearly illustrates the trend toward integration into logical circuits and artificial intelligence.[47]

Mobile ions and electrons form EDLs at the interface between ionic and electronic conductors (e.g., at the hydrogel/metal interface). As for sufficiently small voltages (i.e., ≤ 1 V), the interfaces of electronic and ionic conductors can be considered as non-Faradaic (no matter or charge crossing the interface), EDLs function as nanogap capacitors with a huge specific capacitance. Additionally, in dynamic space, they couple the ionic current and the electronic current on both sides in two different media, allowing to realize an ionic–electronic transducer with an extraordinary vast range of functionality and applications. In field-effect devices, such electrostatic coupling can be used to enable electrical control over insulator–metal transitions, magnetic ordering, and even superconductivity.[48] As an example, electrolyte-gated transistors are based on the EDL effect and have been proposed as a three-terminal synaptic device for mimicking the synapses' behavior. Interestingly, multiple and spatial isolated inputs in such EDL transistors have been demonstrated to trigger a joint output, due to the strong interfacial capacitive coupling, clearly showing their potential for parallel computing like in dendritic integration.[33]

Based on EDL coupling, an ionotronic artificial axon (the so-called ionic cable) was designed, in which the electrical stimuli transmission relies on ions.[49,50] This ionotronic artificial axon is composed of two hydrogel layers separated by a dielectric layer of elastomer. Although one end of the artificial axon serves as the input port and is connected to an external power source, the other terminal serves as the output port and is connected to an impedance load. It has been demonstrated that the obtained artificial axon can transmit alternating signals from the input port to the output port over a long distance.

The coupling of ions and electronic conductors has been also used in the detection of electrophysiological signals, which are essentially based on the ions. In research areas such as human–machine interfaces and biohybrid robotics, such signals are coupled to electronic conductors and, thereby, either measured or triggered. As an example, hydrogel-based soft micropillar electrode arrays have been proposed for electrophysiological recording *in vitro*.[51] In these set-ups, it is important to note that the interfacial impedance between the electrode and cells is greatly reduced that, in turn, enhances the electrophysiological recording capability. Additionally, the developed hydrogel-based microelectrode arrays (MEAs) serve as a soft interface material for cells, providing a microenvironment with tissue-like mechanical properties.

Both electrochemical and EDL processes have been exploited in many diffusive devices. For example, in the EDL-based synaptic transistors, ion diffusion yields a time-dependent stretched-exponential function, which exhibits synapse-like behaviors, such as short-term plasticity.[52,53] Diffusive memristors are other typical examples of diffusive ionotronic devices, in which the electrochemical and diffusive dynamics of the active metals enable the temporally evolution of the conductance.[54] In these devices, the resistance spontaneously recovers after an external bias, because the metallic conductive pathway is physically dissolved,[54–56] contrarily to the situation found in nonvolatile electrochemical metallization (ECM) cells (e.g., RRAM). Due to the varying time scale of the relaxation processes, diffusive memristor can be considered for novel applications such as selector devices for addressing and synaptic devices for neuromorphic computing. Furthermore, a diffusive memristor was proposed for developing signal generator applications such as random number generator[57] and neural spiking generator,[30] representing the versatility of such devices.

3. Building Blocks of the Bioinspired Ionotronic System

Despite the first availability of a number of ionotronic devices, system of bioinspired ionotronics are far from being fully developed. In **Figure 3**, the required basic elements that can construct a bioinspired ionotronic system (BIS): 1) sensing/signaling, 2) wiring/interfacing, 3) memory/computing, and 4) decision/actuation components are summarized.

The first point represents the input of the system, which is responsible for the detection of the information from the external environment and can also include a signal conversion and stimulation part. Although electronic sensors have been developed to high precision and speed, they lack features as adaptability, fault-tolerance, and biotexture-like properties (e.g., stretchability, softness, or self-healing capabilities). In biological systems, numerous types of receptors can be found, ranging from iconic over haptic receptors to chemical recognition. Furthermore, biological sensory systems rely on ions as the signal carrier and transmit signal using the spiking signals, which is power-efficient and robust to noise. The combination of photoelectric signal conversion and transduction into an ionic signal was recently described in the development of an artificial light-driven ion pump.[58] Indeed, such devices can be deemed as an ionotronic optical receptor as well.[59] In the field of haptic receptors, the recent enormous development in e-skin devices that resemble natural haptic sensors can be regarded as a salutary lesson for the development of bioinspired receptors with various sense modalities. As a clock generator that is responsible for synchronization and timing sequences is crucial for digital systems, a primary signal

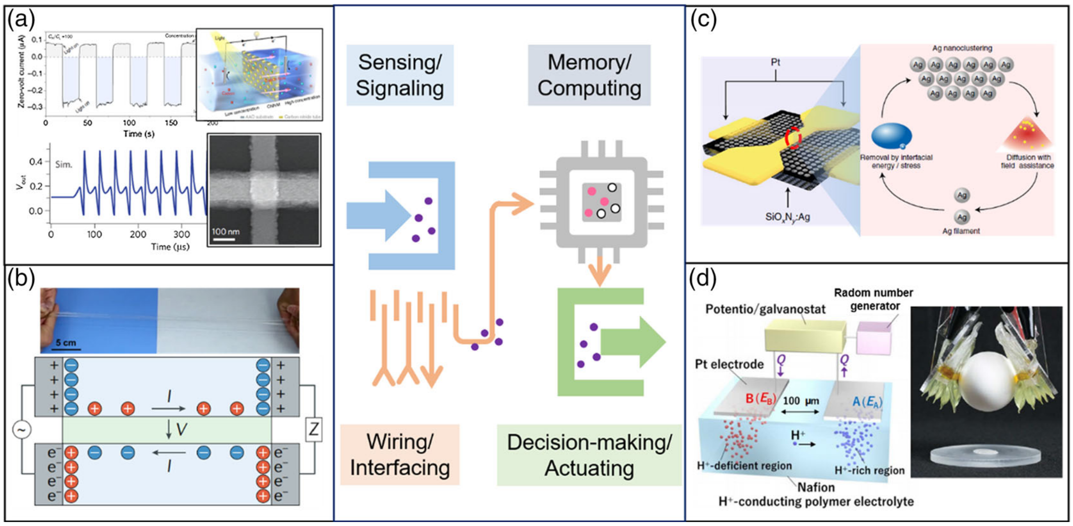

Figure 3. The key components for constructing a bioinspired ionotronic system include (a) sensing/signaling, (b) wiring/interfacing, (c) memory/computing, and (d) decision-making/actuating components. The top panel in (a) is the response and schematic diagram of the artificial light-driven ion pump. Reproduced with permission.[58] Copyright 2019, Springer Nature. The bottom panel in (a) is the response and scanning electron micrograph of the artificial Hodgkin–Huxley axon. Reproduced with permission.[59] Copyright 2012, Springer Nature. The top panel in (b) is the image of an ionic cable. Reproduced with permission.[49] Copyright 2015, Elsevier Ltd. The bottom panel in (b) is the schematic diagram of the ionic cable. Reproduced with permission.[32] Copyright 2018, Springer Nature. The diffusive memristor in (c). Reproduced with permission.[60] Copyright 2019, Springer Nature. The left panel in (d) is the ionic decision-maker. Reproduced with permission.[47] Copyright 2018, Springer Nature. The right panel in (d) is the hydrogel-based artificial muscle. Reproduced with permission.[61] Copyright 2018, Springer Nature.

generation component should also be considered for constructing BIS. Especially, the spiking neural network (SNN) is deemed as the next-generation model for neuromorphic computing. Such a model closely mimics natural neuronal networks in which the computation starts from the generation of action potential in axons. An artificial Hodgkin–Huxley axon was achieved by a Mott memristor (NbO_2)-based module in 2013,[60] and diffusive memristor-based[30,63] and electrolyte-gated synaptic transistor[64]-based artificial axons were reported later, serving as a pioneering piece of inspiration.

The second class of components describes the wiring or interfacing system that allows the interconnection of all the functional units. The aforementioned hydrogel-based ionic cable stands out as a strong candidate for wiring purposes. In addition to its EDL-mediated capability in coupling electronic and ionic signals, ionic cables importantly show bio-like properties such as high stretchability that enables good mechanical robustness and biocompatibility that allows its use in implantable systems.[65] Ionic conductors can enable high voltages and frequencies applications as well to trigger dielectric elastomer-based actuators,[50] illuminate phosphors,[66] and drive liquid–crystal devices.[67]

The third group includes devices/circuits responsible for processing, that mainly stands for memory and computing functions. Although memory and computing units are generally separated in conventional digital computer, our brain however organizes the neural network with in-memory computing architectures. Therefore, future ionotronic systems can favorably exploit more such bioinspired neuromorphic strategies, for example, using the memristor-based synaptic devices as a basic in-memory computing unit. Furthermore, endeavors on ionotronic processing components can focus on the construction of neuromorphic chips for the realization of fully analog, in-memory-computing, which may potentially accelerate the computing speed significantly while reducing power consumption. Additionally, ionotronic devices can simplify or extend the capability of conventional computational schemes. For example, a cross-point array of resistive memory devices can directly solve a system of linear equations, or find the matrix eigenvectors.[68]

The components in the fourth group stand for an action output (or feedback) of the system, which deals with decision-making and/or actuating. These modules are of utmost importance as a complete bioinspired system not only detects external stimuli, but also, equally importantly, elaborates and accordingly reacts to the stimuli via actions to the outside environment. Thus, the aforementioned ionic decision-maker[32] and ionotronic artificial muscle fall into this category.[62] Interestingly, the latter is made up of a dielectric elastomer sandwiched between two hydrogel layers, in which the electrostatic force is generated from the two hydrogel–elastomer interfaces.

4. Constructing the Bioinspired Ionotronic System

Despite many breathtaking accomplishments, the current progress is still far from the realization of completely autonomous BISs. However, remarkable attempts have been reported in the last years. Some examples are reported in **Figure 4** and further discussed the following paragraphs.

An artificial afferent nerve (Figure 4a) was developed for collecting sensory data, converting the obtained information into action potentials, and integrating the action potentials for pattern recognition and muscle control.[69] Such an ionotronic

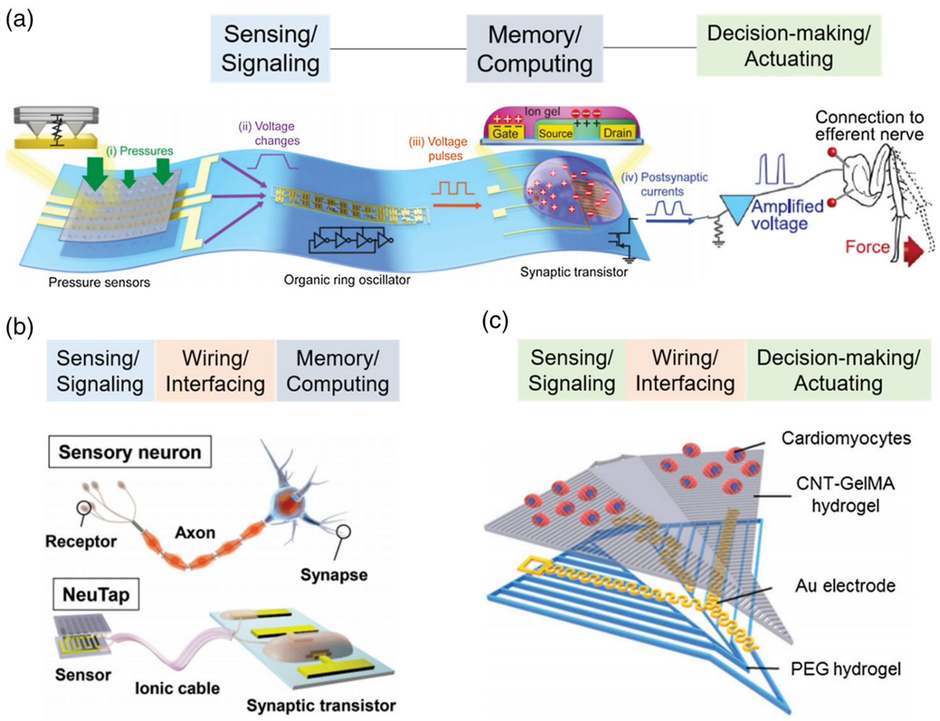

Figure 4. Examples of BIS: a) an artificial afferent nerve; Reproduced with permission.[69] Copyright 2018, AAAS. b) an artificial sensory neuron; Reproduced with permission.[70] Copyright 2018, WILEY-VCH. c) a biohybrid batoid ray. Reproduced with permission.[71] Copyright 2018, WILEY-VCH.

system consists of three fundamental components: 1) resistive pressure sensors, 2) organic ring oscillators, and 3) a synaptic transistor. Although the first two components belong to the sensing/signaling category, the latter belongs to memory/computing category. In brief, the experimental realization of this device relies on an ion gel-gated synaptic transistor that can combine input signals coming from multiple ring oscillators (simulating parallel tactile inputs) and elaborate a single "cumulative" output via EDL-mediated electrostatic coupling. As this system perfectly emulates the functions of biological afferent nerves, it has indeed potential applications in neurorobotics and neuroprosthetics.

The artificial sensory neuron (Figure 4b) consists of sensing, wiring, and memory/computing components integrated together to functionally and structurally replicate a tactile sensory neuron.[70] An ionic cable was introduced to wire the sensing and memory/computing components, transmit sensory information, and also confer mechanical durability to the system. The information flow/processing is based on the electronic/ionic coupling and proceeds as follows: the pressure sensor converts the pressure stimuli into electrical signals, the ionic cable transmits these signals to the synaptic transistor, which finally induces specific decay properties depending on the stimuli patterns.

Finally, a biohybrid soft robot (Figure 4c) was developed combining signaling, interfacing, and actuating components.[71] Herein, an electrical signal is applied via Au electrodes to allow the electrically controllable motion of adhered cardiomyocytes. In the reported system, cell-to-cell coupling was enhanced by the introduction of an ionic/electronic hybrid conductor, composed of carbon nanotubes (CNTs)–gelatin methacryloyl (GelMA) hydrogel. Interestingly, further development of such BIS can lead to cardiac- or muscle-based integrated electrical stimulators, which have potential in such applications as in vitro muscle models for drug testing.

5. Challenges and Prospects

The final goal of BIS is the development of next-generation bionic and biohybrid systems for computing, robotics, prosthetics, and many more applications. Helping achieve this goal, ionotronics provide powerful tools and methods for narrowing the gap between conventional electronics and the biological world on the other side, potentially giving birth to novel and exciting paradigms with comparable or even superior capabilities compared with their electronic predecessors.

In the field of computing systems' development, ionotronics devices can provide a unique opportunity for constructing brain-like computing architectures at the neuron and/or synapse level. Conventional computing devices are based on Von Neumann architecture which suffers from low-level of parallelism and physical separation between processing and memory. Although the newly developed neuromorphic hardware architectures have greatly extended the capabilities of current computing systems with extremely increased parallelism, it is still difficult or impossible to achieve human-like sensorimotor abilities by them (known as Moravec's paradox). A possible explanation for this still unsolved challenge can be the aforementioned physical separation between sensing and processing/memory in these hardware architectures, which is absent in biological systems, where detection, identification, and interpretation of multisensory information are fully integrated.

Future intelligent robots will also benefit from the development of ionotronic computing systems with remarkable perceptual capabilities. On one hand, the incorporation of artificial intelligence and perceptual capability can endow robotic artifacts with awareness and machine cognition, which possibly greatly boost the interaction with humans. On the other hand, ionotronic devices/systems can allow the automatic readout of individuals' sensations and emotions because of the already demonstrated capability in the accurate monitoring of electrophysiological signals. This also propels the creation of intelligent prostheses and advanced medical-care instruments that can perceive human feelings and can respond accordingly. Experiments on social–physical human–robot interaction have proven that humans can obtain support from the humanoid robots.[72] Therefore, the development in this area would offer unprecedented opportunities to empower current digital systems with emotional, psychological, and social attributes, making closer work with robots possible.

Yet, several scientific and engineering challenges have to be overcome to fully exploit the BISs in practical applications. A first challenge can be identified by our sometimes-limited understanding of the biology that can hinder and even mislead the development of ionotronics-based reverse engineering. As an example, as the operating mechanism of the neural network is still unclear, only simplified models such as the SNN[30] and the convolution neural network (CNN)[29] have been practically used, and they are struggling with achieving higher sophistication. In addition, it is necessary to better understand the interaction between ions and electrons to build a theoretical model of complex interactive systems based on them. As research on BIS is a new and multidisciplinary field, it is expected to dovetail with other fields, such as micro-electromechanical systems (MEMS), microfluidics, and integrated circuits. In that case, interfacing components and technologies to incorporate all these units are to be definitely addressed. Secondly, long-term durability remains an open important challenge for the practical use of BIS. Ionic cables, for example, suffer from dehydration, which, however, can be avoided by proper insulation from the environment.[73–75] Additionally, higher mechanical durability and conformability could be beneficial for improved performance at the electric–biological interfaces.[7,8,76] Here, technologies like encapsulation, surface treatment, and molecular design are necessary to protect the devices from harsh environmental conditions. Finally, repeatability and device-to-device variation have to be improved for the success of neuromorphic computing.[77]

In summary, the transition from electronic-based to the ionotronic-based bioinspired system is still in its infancy, and several challenges have to be addressed before full translational implementations can be achieved. Nevertheless, optimistically, synergies between electronics and ionics can accelerate and expand the renovation in many areas included robotics, bioelectronics, biomedicine, and many more.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

bioelectronic interfaces, bioinspired systems, ionotronics, neuromorphic computing, neurorobotics

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