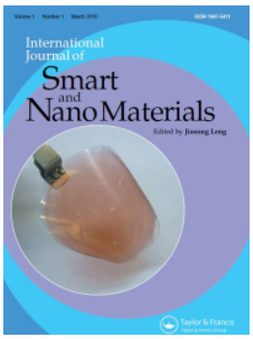

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Bioinspired ionic control for energy and information flow

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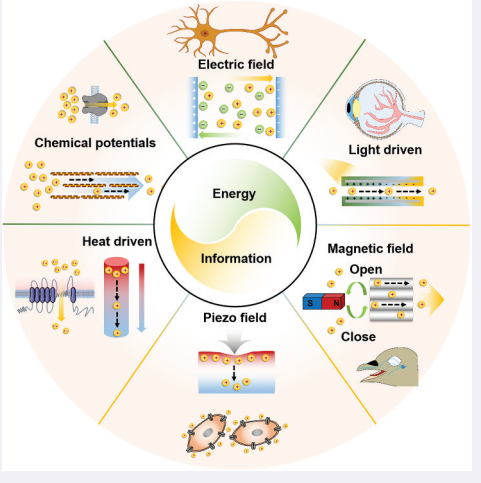
ABSTRACT

The control of ion transport by responding to stimulus is a necessary condition for the existence of life. Bioinspired iontronics could enable anomalous ion dynamics in the nano-confined spaces, creating many efficient energy systems and neuromorphic in-sensor computing networks. Unlike traditional electronics based on von Neumann computing architecture, the Boolean logic computing based on the iontronics could avoid complex wiring with higher energy efficiency and programmable neuromorphic logic. Here, a systematic summary on the state of art in bioinspired iontronics is presented and the stimulus from chemical potentials, electric fields, light, heat, piezo and magnetic fields on ion dynamics are reviewed. Challenges and perspectives are also addressed in the aspects of iontronic integrated systems. It is believed that comprehensive investigations in bioinspired ionic control will accelerate the development on more efficient energy and information flow for the futuristic human-machine interface.



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Iontronics; nanoconfined ion transport; ion dynamics; bioinspired iontronics; ionic-electronic coupling interface

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1. Introduction

Biological systems typically rely on ions or molecules for information transmission as well as energy exchange and storage, which fundamentally differ from the electronic signals in the information technology utilized today [1]. Iontronics as a potential bridge between electrons and biological ions has been rapidly developed recently [2–4]. It means a science and technology for coupling the electrons and ions to achieve specific functions. In detail, iontronic and electronic systems employ distinct charge carriers, namely ions and electrons, to fulfill their respective unique functions. While electrons possess efficient and reliable transmission/response speeds, they lack characteristics inherent to biological structures such as self-healing capabilities etc., and struggle to adapt to extreme working environments with the influence of magnetic fields, high temperatures and high humidity [4]. Furthermore, electronic-based integrated circuits are approaching the limit of Moore's law based on von Neumann's computing architecture [5]. In contrast, ions could simultaneously carry rich information as charge carriers, for instance, Ca^{2+} plays a pivotal role as a neurotransmitter release switch in neuronal conduction [6]. Iontronics provides opportunities to achieve more precise integrations of sensory, memory and computing, thereby overcoming the limitations of single-functionality in electronic sensors.

Over the past decade, inspired by natural biological nanoconfined systems, the integration of the ionic and electronic advantages has significantly propelled the development of iontronics [3,7–9]. Using various analog signals to stimulate ions and electrons coupling has created a range of multifunctional iontronic devices, including bio-interface device [10,11], iontronic power sources [12–14], ionic selective gating [15–18], ionic diodes/circuits [11,19,20], and neuromorphic computing components [11,21,22], etc. These iontronic devices exhibit remarkable adaptability, mechanical flexibility, and bio-like features such as safety and reparability, positioning them as potential bridges between advanced intelligent electronic devices and biology [23,24]. It is anticipated that they will play crucial roles in human-machine interfaces and bioinspired robots. They may also drive the development of next-generation in-sensory computing and energy technology through fine-tuning of ion dynamics [12,13,25,26]. In biological systems, super-bio-receptors, ranging from iconic super tactile receptors to olfactory recognition, could regulate various-sized molecules or ions and possess sensitivity far surpassing electronic devices with lower power consumption. Understanding the bioinspired ionic control and the ionic-electronic coupling interface in these bio-processes could help us develop iontronics with higher performance.

In this review, we will specifically focus on the bioinspired ionic control in the nanoconfined spaces for energy and information flow. Firstly, the ion dynamics including ion transport behaviors and ionic-electronic coupling interface were summarized, which is the basis for the iontronic functionalities. Secondly, bioinspired ion dynamics control for iontronic applications was briefly described from various stimulations, such as chemical potentials, electric fields, light, heat, piezo and magnetic fields. Last but not least, an outlook for future challenges and opportunities was provided to drive the development of bioinspired iontronics, such as finding new methods for precise ionic control and optimizations of ionic-electronic coupling interfaces, etc.

2. Ion dynamics

2.1. The transport behaviors of ions

Ion is a charged particle formed by the loss or gain of electrons from an atom or atomic group, which is also a fundamental particle, like molecules and atoms, that constitutes matter. Therefore, it also conforms to the basic thermodynamic relationship when transporting. The motion of ions from one place to another in a solution could be caused by the difference in electrochemical potential or chemical potential between two locations, or by diffusion in a certain solution. Therefore, ion transport occurs in the bulk system through three modes [27] as shown in Figure 1a-c: (1) the movement of a substance under the influence of a chemical potential gradient (i.e. salinity gradient), which is diffusion; (2) the movement of charged substances under the influence of an electric field/electrochemical potential gradient, known as migration; (3) the movement of a substance under the external forces (such as fluid density gradients, stirring, etc.), which is convection. However, anomalous transport behaviors of ions/molecules in the nanoconfined spaces have been observed [9,28,29]. Similar to efficient ionic motions in biological protein channels in Figure 1d, the ion dynamics in artificial nanoconfined spaces (about 2–100 nm) are significantly different from the bulk systems and present new mechanisms [30]. This may be attributed to the surficial/interfacial charge and chemical properties of nanoconfined channels that could amplify van der Waals and electrostatic forces on ions and molecules, resulting in enhanced ionic transport and interaction behaviors [31,32] as shown in Figure 1e. Even in a smaller sub-nanoconfined space under 2 nm, a range of anomalous ion dynamics will occur because of the short-range steric/hydration forces, such as the ionic coulomb blockade concerning single ion transport [29] (Figure 1f), the space-confined size of ions and desolvation [33], superionic states [34], drastic changes in diffusion coefficients [35], ion – ion correlation [32], and ultra-dense packing of ions [36–38], etc. Meanwhile, the classical mechanical and thermodynamic equations are challenged in such sub-nanoconfined system, for example, the Navier – Stokes equation is hard to describe the nanofluidic flow [30], Kelvin equation is inapplicable due to the nanoconfined size of water molecules [39], and Hertz – Knudsen equation is limited by the finite evaporation dynamics in the nanoconfined spaces [40].

2.2. The ionic-electronic coupling interface

Ionic diffusion, migration, convection, and transport in the nanoconfined spaces are the basis for building the iontronics. The moving ions would ultimately maintain thermodynamic equilibrium within the system or undergo charge transfers at the ionic-electronic coupling interface [41]. The ionic-electronic coupling usually occurs between the liquid and solid interface [42]. Generally, the coupling process can be categorized into three fundamental types as shown in Figure 2a-c: (1) the electric double-layer (EDL) capacitance process, (2) the electrochemical redox reaction process, and (3) the pseudocapacitance process. By employing electrochemical methods such as cyclic voltammetry (CV) and electrochemical impedance spectroscopy (EIS), the dynamic coupling processes at the ionic-electronic interface can be rapidly characterized through key descriptors from CV characteristic curves or 3D-Bode plots derived from EIS [43,44]. For instance, the CV curve

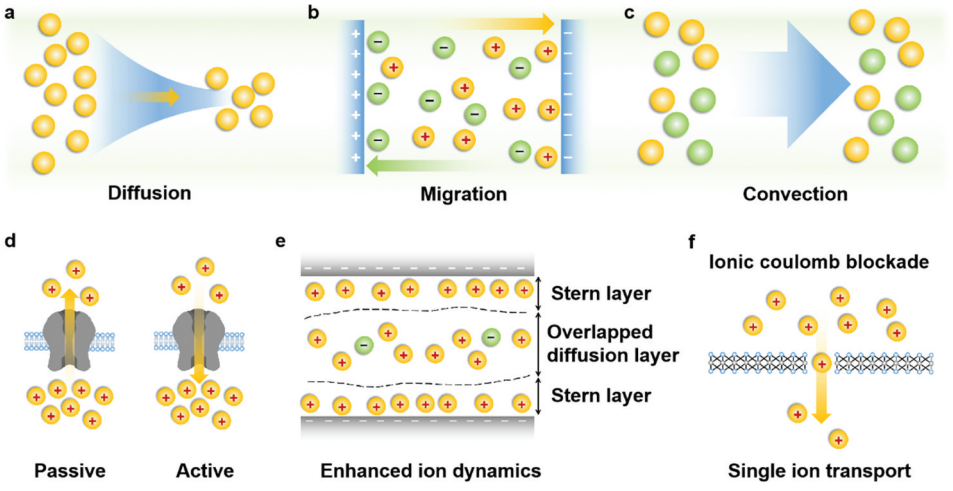

Figure 1. The transport behavior of ions in bulk system of (a) Diffusion; (b) Migration; (c) convection. (d) Ion transport in biological system, with passive diffusion and active transport. (e) The overlapped diffusion layer will result in a selective nanoconfined channel, with co-ions being excluded and counter-ions being able to pass through the channel. (f) Ionic coulomb blockage due to single ion transport in sub-nanoconfined space.

of the EDL process could be demonstrated as an approximate rectangle, where the current remains relatively stable with varying voltage. The redox reaction process would exhibit redox peaks in the curve, representing the Faradaic process of the electrochemical reaction, and the position as well as shape of such peaks are influenced by the rate of the electrochemical reactions. Under pseudocapacitive control, the rising current curve is almost linearly related to the voltage, which indicates that the ions diffusion and the redox reactions at the interface are simultaneous and it may cause the long-distance electron and ion transport coupling.

In detail, the EDL capacitance describes a rapid dynamic absorption of ions and electrons, resulting in the formation of ionic-electronic field dipoles at the interface as shown in Figure 2d. This process exhibits remarkable high specific capacitance (greater than 1 $\mu F \cdot cm^{-2}$), implying that the EDL can achieve strong capacitance coupling between electrons and ions [45]. Similar EDL structures also exist on the surface of biological neural cell membranes, which can establish an ionic potential difference between the intracellular and extracellular sides. Once a neural impulse is sensed, it triggers the ions flow across the membrane, facilitating the neural signal transmission through EDLs [46]. Inspired from such feature, an integrated ultra-thin layer microstructure iontronics could be employed for pressure sensing and realize a skin-like super tactile neural sensor [47]. Its linear range could reach 1700 kPa and the sensitivity was demonstrated as 33.7 Pa^{-1}. It could also achieve an exceptional pressure resolution of 0.36% under 2000 kPa pressure, showing outstanding pressure detection capabilities. The EDL capacitance interface without redox reactions also allows iontronic devices to be seamlessly connected to neurons, enabling effective neural signal conduction. For instance, an ionic junction fiber diode made by polymeric electrolyte has been developed to mimic synaptic characteristics and neural signal transmission [11]. When connected to the distal end of

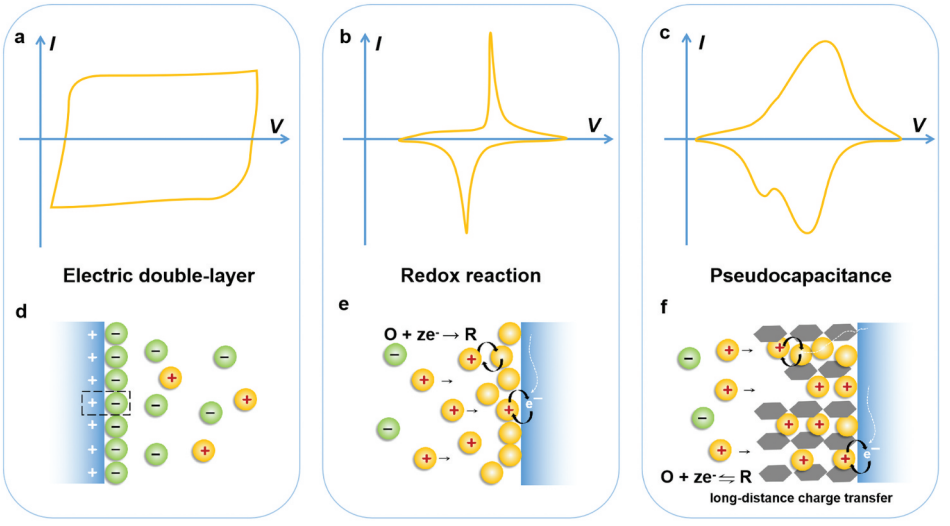

Figure 2. Typical I-V characterizations of the fundamental ionic-electronic coupling interface processes of (a) electric double-layer; (b) Electrochemical redox reaction; (c) Pseudocapacitance; and the corresponding schematic illustration of (d) electric double-layer; (e) Electrochemical redox reaction; (f) Pseudocapacitance.

the mouse sciatic nerve, synaptic characteristics and neural signal transmission of ions were achieved based on the EDL driven ion and electron interaction mechanism. In addition, in the field of energy storage, the high specific capacitance property of the EDL can be utilized to design efficient iontronic capacitors [48]. These capacitors enable rapid energy storage and release, providing robust support for applications like electric vehicles and wearable electronics [49].

The electrochemical redox reaction represents a significant process involving the transfer of charges between electrodes and electroactive materials [50] as shown in Figure 2e. In comparison to the EDL process, it avoids charge accumulation, leading to a more complete charge transfer at the ionic-electronic interface, which is ideal for efficient osmotic energy collection, and it could provide additional chemical energy [3,35]. In addition, similar to the irreversibility of neurotransmitter release processes, electrochemical reactions are normally irreversible and could result in nonvolatile changes in the device [51]. Therefore, electrochemical processes are also widely used in applications like artificial synapses and information storage. For instance, a memristor made by ions triggered structure changes or redox reactions has been developed on a flexible plastic substrate, which can be used in artificial synaptic-like memory and neuromorphic computing [52]. Furthermore, electrochemical processes play essential roles in iontronic energy storage as well as bioinspired neurologic circuits [53].

Pseudocapacitance represents a more intricate process of the coupling interface [42] as shown in Figure 2f. It often involves long-distance charge exchange between electrons and ions due to the interfacial redox reactions are related to the ionic diffusion beyond the interface. At the same time, pseudocapacitive process typically provides higher energy density with decent power density. In iontronic devices, the pseudocapacitance effect is often viewed as an enhancement to improve the performance, compared to the

EDL and electrochemical reaction process. For example, a MXene-based pseudocapacitive iontronic pressure sensor has achieved a maximum sensitivity exceeding 45,000 kPa^{-1} and minimum sensitivity exceeding 200 kPa^{-1}, demonstrating outstanding performance over a wide range beyond 1.4 MPa. It also has a low detection limit of 20 Pa, and could exhibit stable long-term operational performance for monitoring bodily activities and tactile feedback for flexible robots [54]. Pseudocapacitance also plays a crucial role in bioinspired iontronics, and the pseudocapacitance effect may be employed to mimic the charge storage mechanism of biological synapses, thereby achieving signal transmission and processing functions akin to biological synapses.

3. Bioinspired ionic control for iontronic energy and information flow

Ion regulation is one of the important processes to maintain intracellular homeostasis and perform many biological functions. The most efficient transport of ions in living organisms occurs at the cellular or subcellular level in nanoconfined spaces, such as ion channels [55]. These nanoconfined spaces can accommodate particular biomolecules for specific biological functions. In addition, super-bio-receptors play a crucial role in regulating ions. They can perceive various stimulus from the internal and external environments, such as chemical potential, electric field, light, heat, piezo and magnetic fields, directly influencing the state of ion movement, or prompting specific molecules/proteins to react or undergo structural changes to transport ions [56]. These processes can generate energy and transmit information, realizing various life activities within the organism, such as exocytosis, fertilization, muscle contraction, pH balance, nerve conduction, cell volume regulation, and modulating adenosine triphosphate (ATP) hydrolysis and synthesis [57–60]. Thus, by learning from creatures, we can utilize specific material structures and physical or chemical simulated signals to modulate the ions by artificial 'stimulation methods.'

3.1. Chemical potential driven ion dynamics

Chemical potential is the tendency of a higher potential state transform to a lower potential state by chemical reactions or chemical environment changes, such as the salty gradient. It is the primary way to drive ion dynamics for regulating life activities in biological systems. For example, electric eel cells can utilize the chemical potential of salinity gradients to generate high voltages and currents [14,61] as shown in Figure 3a. Each cell has highly selective nanoconfined ion channels (radio of ~ 0.6–0.8 nm) that can select small ions (Na^+ and K^+) for passage, thereby generating a transmembrane potential. Among these, the Na^+ and K^+ gradients could act with potentials of 65 and 85 mV, respectively. It could produce a total transcellular potential of about 150 mV when added in series. Thousands of electrocytes arranged in series and parallel can generate potentials and currents as high as 600 V and 1 A, respectively. Osmotic energy, using salty gradients to modulate ions, holds broad prospects for applications in energy conversion as well as intelligent sensors, and its ion selectivity and energy conversion efficiency depend on the performance of the ion channels [7]. To achieve more efficient ion selection and transport structures for rapid osmotic energy harvesting, various techniques such as ion or electron beam lithography, etching, surface modification, and photolithography have been employed to

create bioinspired ionic nanoconfined channels and pumps [9,63–66]. Typically, they are designed as asymmetric structures (e.g. conical, bullet-shaped, cigar-shaped, and funnel-shaped), or with a substantial surface EDL charge on the inner wall. Upon exposure to an electrolyte solution or environmental humidity, capillary action and Coulomb adsorption from the surface EDL charge would lead to the rapid transport of opposite charged ions, and hindering ions with the same charge. Specific enhancement of ion dynamics is more pronounced in nanoconfined structures of 2–100 nm or smaller, as the overlapping of the EDL on the inner wall of the channels would intensify the electrostatic force that acting on the ions. For, example, the osmotic energy based on a single MoS_2 pore (pore size of 2–25 nm,) can reach up to 10^6 W·m^{-2} theoretically [62] (Figure 3b), and a single double-walled carbon nanotube (with an inner radius of 2.3 nm) could achieve significant power densities up to 22.5 kW·m^{-2} [67]. Furthermore, a tunable iontronic amplifier can respond to sub-nanoampere currents with a gate voltage of less than 1 V, resulting in an approximately 300-fold amplification of ionic current output based on the ion transport in nanochannels [22].

In addition, life activities such as metabolism, nerve signaling, and enzyme catalysis also involve biochemical reactions. Inspired from those processes, chemical reactions can also be used to drive ion dynamics and lead to higher performance of iontronics. In biological systems, the chemiosmotic coupling mechanism for adenosine

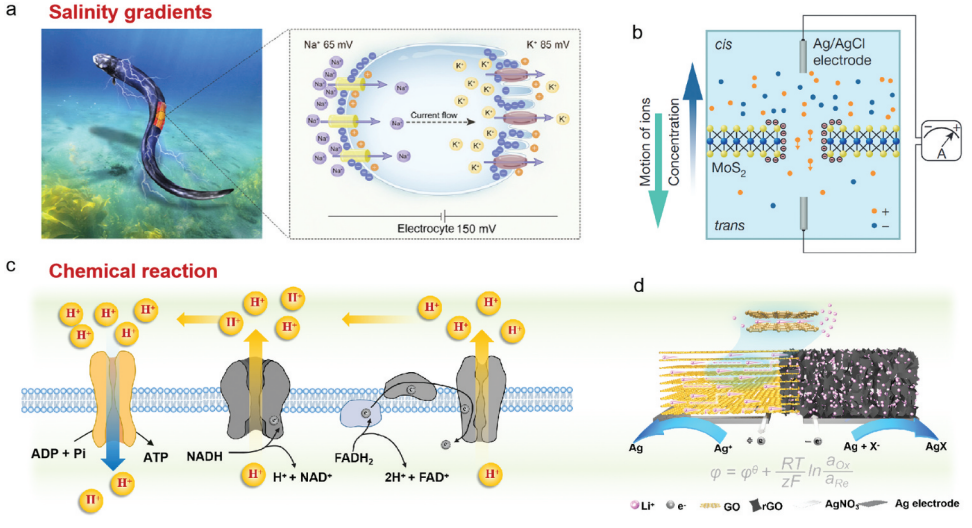

Figure 3. (a) Schematic illustration of structures of the electric eel's electrocytes and each electrocyte of the electric eel can generate 150 mV when stimulated via ion transportation of K^+ and Na^+ through highly selective nanoconfined ion channels. Reproduced with permission [14]. Copyright 2021, National Academy of Sciences. (b) Schematic illustration of harvesting osmotic power by separating salinity gradients using a MoS_2 nanoconfined pore membrane with a thickness of 0.65 nm, which has been reported to achieve10^6 W m^{-2} power density theoretically. Reproduced with permission [62]. Copyright 2016, Springer nature. (c) Schematic illustration of the chemiosmotic coupling in mitochondria, the electronic energy generated by the chemical reaction will construct a proton gradient, and the proton osmotic energy will ultimately be involved in ATP synthesis. (d) Schematic illustration of the GO-based iontronic power source, which realized the coupling of chemical and osmotic energy. Reproduced with permission [12]. Copyright 2023, John Wiley and sons.

triphosphate (ATP) synthesis is an important example of utilizing interfacial redox reaction and efficient ion translocation. As shown in Figure 3c, to provide energy, mitochondrial nanostructured ionic pumps (about 9–12 nm size) would capture the electron energy generated from the decomposition of nicotinamide adenine dinucleotide (NADH) and pump protons into the intermembrane space [58,68]. The conversion of adenosine diphosphate (ADP) to ATP, driven by proton motive force, is effectively coupled to the ionic flow of protons back across the membrane, and generating proton transfer redox chains. Redox reactions and proton gradient induced chemical osmotic coupling are crucial for ATP synthesis, and they provide us with an idea to control ion dynamics by coupling ions and reactions [12,13]. For example, it is common to employ an EDL process or electrochemical redox process in the ionic-electronic interface of the iontronic power source to convert ionic current to electronic current [69–72]. A neuromorphic power generator was developed by utilizing uniformly nanoconfined laminar channels and Ag/AgCl interface redox reactions. It can selectively deliver cations and exhibit 95.8% ion selectivity, 41.4% energy conversion efficiency, and a power density of 5.26 W m^{-2} [35]. Recent studies have shown that rapid ion dynamics could be realized by efficient translocation of Li^+ within 2D nanofluidic channels of graphene oxide (GO) under salinity gradients, along with the fine-tuned interface redox reactions [12] (Figure 3d). This results in exceptionally high osmotic power (or iontronic power) with the volume power density and energy density reaching 438.02 mW·cm^{-2} and 30.02 mWh··cm^{-2}, respectively. The areal power density was reported to be 1095.05 mW cm^{-2}, surpassing most flexible batteries.

On the other hand, specific ionic reactions and transport play a crucial role in the biological information flow, such as the releasing of neurotransmitters after catching the action potentials in biological neurotransmission, which means ions can also act as a switch to reactions. Neural impulse conduction is achieved by sub-10 nm synapse ion channels, where the action potentials could open specific Ca^{2+} channels and lead to the Ca^{2+} influx. This would trigger the synaptic vesicles to rapid release neurotransmitters into the synaptic cleft, enabling the transmission of information between neurons [6,73]. This has inspired the development of ion-gated and ionic memristors. For example, a cone-shaped polyimide nanochannel functionalized with alkali metal cation-responsive molecules has been devised as the Na^+-activated and K^+-activated cation gates [74]. Additionally, a neural structure based ionic memristor process involves the oxidation, migration, and reduction of ions, resulting in the formation of local conductive filaments and leading to nonvolatile changes in conductivity. For instance, an Ag based filament growth kinetics has been demonstrated. A single Ag atom could transfer through a series of ionization, ionic migration and reduction processes to implement memristor for nanoscale solid-state physical evolutionary networks [75]. It shows attractive electrical properties, including a high on/off ratio (>10^2), low current (≈nA), and long retention period (>10^4 s). Making it a promising candidate for large-scale hardware implementation. Moreover, by leveraging proton transport and the electrochemical reaction interface transfer process, an electrochemical neuromorphic device has been fabricated [52]. It is capable of exhibiting over 500 distinct nonvolatile conductance states with extremely low energy consumption (<10 pJ per 10^3 μm^2) and achieving high spatiotemporal precision in neural network simulation. Bioinspired memristor devices can faithfully replicate the

dynamics of synapses and neurons in biological systems, allowing for the development of complex switching behaviors through network evolution, holding promise as model systems for testing hypotheses in neuroscience [76].

3.2. Electric field driven ion dynamics

Like the membrane potential generated by the ion flow across the nanoconfined ion channels on the electric eel cell membrane, the ion channels can also undergo conformational transform when the membrane potential changes. One possible mechanism for voltage-dependent ion channel conformational changes is shown in Figure 4a. The link connecting the voltage sensor to the ion gate is rigid, and membrane potential changes would cause charge transfer through the polarized proteins and activate the voltage sensor (usually protein aggregates structural changing), resulting in a voltage-gated ionic current [77]. Namely, many voltage-dependent nanoconfined channels also exist in organisms, such as 31 kDa membrane proteins on the mitochondrial outer membrane as well as the Shaker K and KcSa proton channels, which can sense changes in the electric field across the cell membrane and open rapidly when the voltage reaches a certain threshold [78]. Such channels play an important function in regulating biological ionic gating, osmosis, selection, and neural conduction [15,16,77]. This feature has also inspired researchers to use electric fields to drive ion dynamics for ionic-electronic gating, efficient ion selection etc. In fact, the utilization of an external electric field stands as one of the earliest methods to control the movement of ions. Its origins could be traced back to Luigi Galvani's discovery of the biological muscular-electrical effect in the 18^{th} century [3]. The pivotal aspect in controlling ions movement via an external electric field lies in the design of nanoconfined channels and electrodes with specific features. For instance, a voltage-gated nanofluidic system inspired by cellular voltage-gated ion channels was designed. Integrating with 2D lamellar MXene membranes with a stable layer spacing of 0.7 nm, it could exhibit a surface charge-controlled ionic transport and gating effect [18] (Figure 4b). Its ionic conductivity measured under the positive voltage is an order higher than that measured under the negative voltage. In addition, by mimicking the selectivity of biological ion channels, a bias controlled ion-selective nanoconfined pore was developed in graphene flakes to discriminate the very similar ions, K^+ and Na^+ [17]. Pore size and edge surface functionality can be tailored with bias voltage, which can be used to select specific ions to pass through.

3.3. Light-driven ion dynamics

In photosynthesis, Chlorophyll molecules could release electrons when sensing light, and the electrons would activate the nanoconfined proton channels (~0.6–0.8 nm) in chloroplast thylakoid membranes to transport protons and form the gradients. The energy of this proton gradients could ultimately participate in the Calvin cycle by osmosis, facilitating the synthesis of glucose and ATP, thereby providing energy and a carbon source for plants [79,80] as shown in Figure 5a. To mimic this process, previous studies have attempted to separate charges in intra- or inter-molecular artificial photoactive centers i.e. generating chemical products (such as H^+ or O_2) through receptor chromophores to

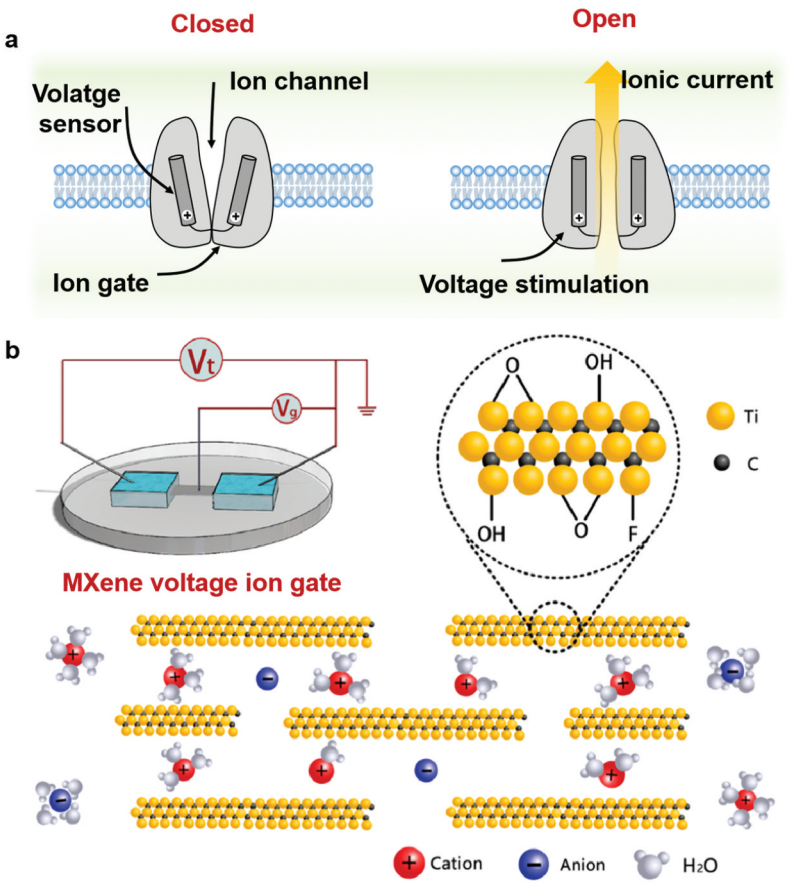

Figure 4. (a) Schematic illustration of the parts forming a voltage-dependent nanoconfined ion channel. (b) Schematic illustration of the MXene voltage ion gate between two hollow cells and the hydrated ions could pass through the nanofluidic sub-nanoconfined channels of the MXene. The enlarged schematic of the membrane shows a single-layer MXene structure and its functional groups. Reproduced with permission [18]. Copyright 2019, American Chemical Society.

transfer charge carriers (electrons or holes) and converting light energy into chemical potential energy [82]. For example, the introduction of F_0F_1-ATP synthase into an artificial photosynthetic membrane containing a proton-pumping component has been developed to catalyze light-driven ATP production [83]. Additionally, an artificial light harvesting system utilizing a reversible transformation of photosensitive spiropyran on membrane could drive protons by illuminating UV light and visible light on opposite sides [84]. It could achieve an approximate 0.12% current efficiency and an open-circuit voltage of about 210 mV. Another approach involves combining photodiodes/dye-sensitized materials with the membrane to construct hetero-structured photovoltaic effect induced ionic pumps. Aiming to control ion migration via a photo-generated electric field in the membrane. For instance, as shown in Figure 5b. a bioinspired light-driven ionic pump of carbon nitride nanotube can reliably produce a sustained open-circuit voltage of 550 mV and an ionic current density of 2.4 $\mu A \cdot cm^{-2}$ [81]. This light-driven ionic pump can separate electrons and holes, generating a transmembrane potential,

which is considered as the fundamental to the pumping phenomenon. Recent research has also indicated that the sensitized bipolar ion exchange membrane with photo-acidic dye could convert the visible light into ionic energy [85]. It could yield an open-circuit photovoltage exceeding 120 mV and holds promise for low-cost, rapid desalination of seawater by using solar energy. Moreover, in a novel light-controllable iontronic direct current triboelectric nanogenerator (TENG), the photo-generated electric field can remotely manipulate the dissipation of ion gradients between graphene oxide and reduced graphene oxide, enabling continuous ionic current generation and counteract the opposite electronic current of the TENG for rectification [86].

On the other hand, biological vision is a sophisticated and marvelous perceptual ability in the natural world, enabling organisms to perceive light, resolve images, and thereby comprehend their surroundings [87]. Within this complex and intricate system, the eyes retina plays a crucial role. When light passes through the lens and reaches the retina, it could induce a conformational change in rhodopsin within the cone and rod cells. The rhodopsin would convert to inverted retinaldehyde and active opsin after separating from opsin. Then, the coupled G-proteins would initiate a series of reactions to trigger receptor potentials and form visual ionic nerve impulses [19,88]. Bioinspired iontronic eyes hold the promise of providing machine vision systems with enhanced perceptual capabilities through efficient processing and transmission of light signals, and may even offer new hope for the blind in the future [89]. For example, an artificial visual system known as the Electrochemical Eye (EC-eye) is constructed using a high-density perovskite nanowire array [19] as shown in Figure 5c,d. Here, perovskite serves as the component for photon capture, while an ionic liquid acts as the common contact of the nanowires or ionic-

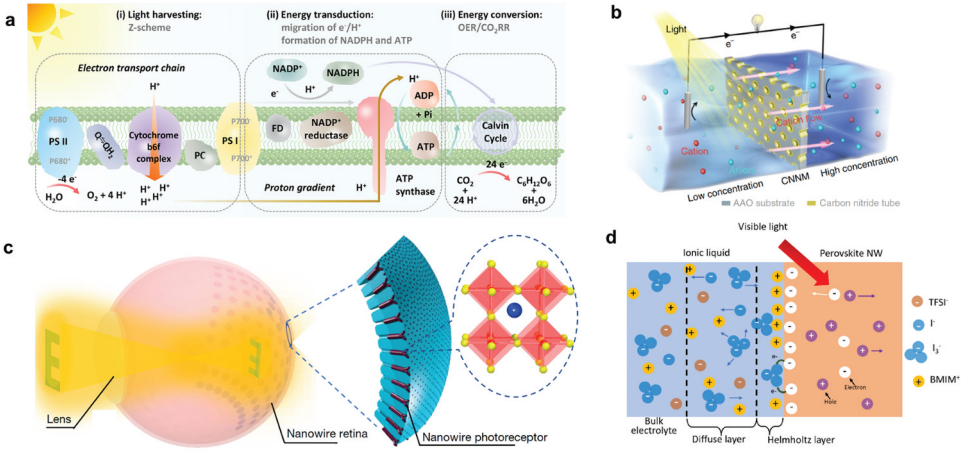

Figure 5. (a) Schematic illustration for the three key photoelectrochemical processes and ion transport in nanoconfined channels of natural photosynthesis. Reproduced with permission [79]. Copyright 2023, John Wiley and sons. (b) Schematic illustration of the light-induced ion pump, which can pump ions transport against a concentration gradient. Reproduced with permission [81]. Copyright 2019, Springer nature. (c) Schematic illustration of the electrochemical eye with the perovskite nanowires in the nanoconfined porous aluminum oxide membrane. Reproduced with permission [19]. Copyright 2020, Springer nature. (d) Schematic illustration of the detailed ionic-electronic coupling interface and working mechanism of the EC-eye. Reproduced with permission [19]. Copyright 2020, Springer nature.

electronic charge transfer based on the electrochemical reaction mechanism. In detail, the perovskite nanowires will generate separated electron-hole pairs under photovoltaic effect when the light stimulus applies. The light-generated electrons will move to the electric double layer interface between the perovskite and ionic liquid, coupling and reducing I_3^- in the ionic liquid to I^-. The excessive I^- will diffuse toward the counter electrode as ionic current. This bioinspired electrochemical eye emulates the photoreceptor of the human retina, demonstrating high responsiveness, reasonable response speed, low detection limits, and a wide field of view, achieving precise optical imaging. Additionally, a hemisphere-shaped neuromorphic retina that incorporating a tunable liquid crystal optical lens and filter-less color vision system has been developed [90]. It could use the wavelength-dependent bidirectional synaptic light responses to construct full-color images, that constructed by the hybrid structure of metal oxide nanotubes and perovskite nanowires. Bioinspired iontronic eyes represent a novel simulation and extension of the biological vision system, bringing new prospects and opportunities to medicine, machine vision, and artificial intelligence.

3.4. Heat driven ion dynamics

Thermal radiation constitutes a crucial support condition for sustaining most biological activities, involving the life regulation and sensing by heat driven ion-gated protein within biological systems when the environmental temperature changes [91,92]. For instance, cyclic nucleotide-gated ion channels (CNGCs) are nonselective ligand-gated cation channels present in eukaryotes [93]. They can be activated with an increase in temperature, leading to the influx of Ca^{2+} as shown in Figure 6a. This process subsequently activates or inhibits a range of gene expressions, thereby regulating plant growth etc. Some extremophiles also exhibit thermophily, generating energy through catalyzing heat driven redox reactions and releasing ions under high-temperature conditions. Both heat driven processes inspired us the fact that heat can be used to control ion dynamics. A thermal ionic battery has been developed for the recovery of high-entropy thermal energy [94] as shown in Figure 6b. Ions would diffuse from the hot side to the cold side under the driven force of a temperature gradient according to the Soret effect. Asymmetric ionic gradients and electric fields would be induced directly to convert thermal energy into ionic-electric energy. Although ionic thermal batteries exhibit higher ionic thermoelectric (i-TE) efficiency compared to thermoelectric devices based on the Seebeck effect, they often display strong capacitive behavior at the ionic-electronic conversion interface, limiting their continuous operation over extended periods. To address this limitation, a novel strategy employs the drift of interface electrons/holes under the influence of the ionic thermal field [94]. It can effectively convert ionic current into a utilized electronic current in the external circuit. Furthermore, using electrochemical redox reactions to convert thermal-generated ionic current has also been developed. Recent research has reported a gelatin-based ionic thermal material that utilizes ion providers like KCl, NaCl, and KNO_3 to modulate thermal diffusion effects, and regulate the thermal ionic current through the redox pair $[Fe(CN)_6^{4-}/Fe(CN)_6^{3-}]$ (Figure 6c), resulting in an enhancement of thermal ion-electric power to 320.7 mW m^{-2}, with the capability of continuous operation for 2 hours [95]. Additionally, an iongel electrolyte with an interpenetrating polymer network structure was reported [96]. It utilized a carbon fabric loaded with polymer redox polyaniline as

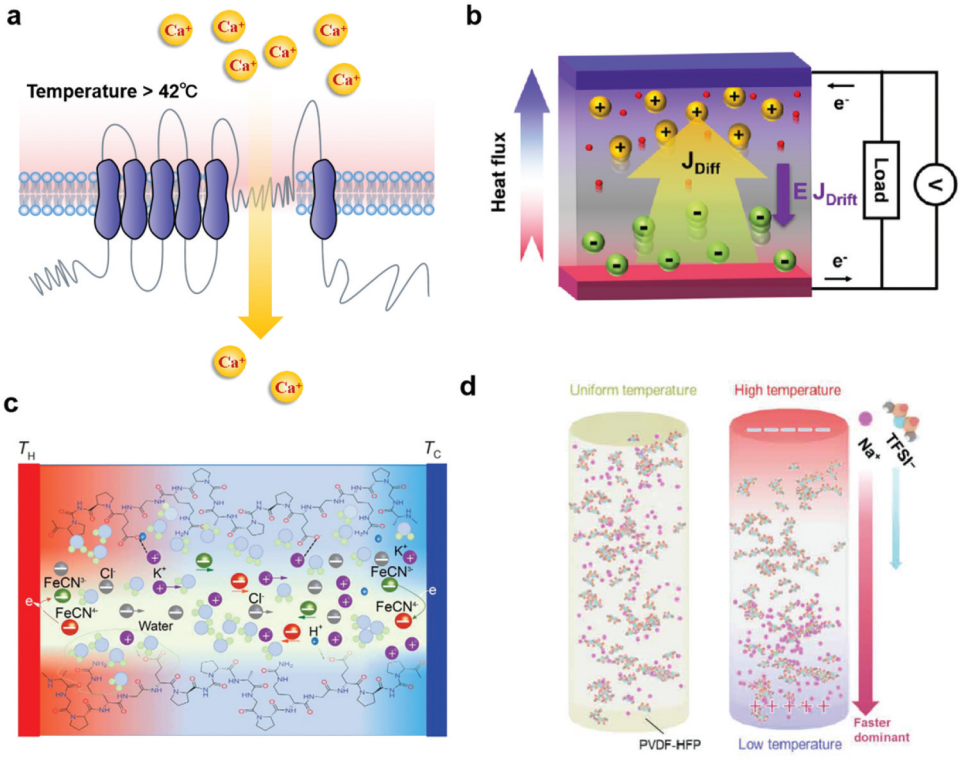

Figure 6. (a) Schematic illustration of the predicted protein structures for CNGC, as the temperature increases, it will stimulate the biological nanoconfined ion channels to open up and allow ions to pass through. (b) Schematic illustration of the working process under the condition of ion – electron/hole thermoelectric synergy. Reproduced with permission [94]. Copyright 2023, Springer Singapore. (c) Schematic illustration of the diffusion, redox reaction, and interaction of the ions in the as-fabricated i-TE materials under the temperature gradient. Reproduced with permission [95]. Copyright 2020, AAAS. (d) Nanoconfined polyvinylidene fluoride – hexafluoropropylene (PVDF-HFP) i-TE materials in porous aluminum oxide provide the channel for ion transport under temperature gradients. Reproduced with permission [20]. Copyright 2022, AAAS.

an electrode to convert ionic thermal current, achieving a continuous output of one hour and a maximum normalized output power of 11.31 mW m^{-2} K^{-1}.

On the other hand, biological thermos-sensors represent an intricate mechanism developed by living organisms. It can help creatures to adapt to the environment, seek food and avoid dangers. Bioinspired iontronic thermosensitive devices utilize the ions motion and the ionic thermoelectric effect to control the opening and closing state of ion channels. It can respond to heat changes sensitively within specific temperature ranges. For instance, a high Seebeck coefficient (~10000 μV K^{-1}) ionic thermoelectric gate-controlled transistor could convert the temperature response to a drain current, which broadens its application in the simple logic resistor-load inverter circuits [97]. Furthermore, a curved infrared thermal imaging device based on ion thermoelectric materials has also been designed [20] as shown in Figure 6d. It consists of a high-density i-TE polymer nanowire array and demonstrates an ultra-fast thermal ion response of 0.69s under subtle temperature differences ($\triangle T < 5K$), with a recovery time of 0.83s. This

device can be effectively used for gazing imaging of human gestures and heating symbols at approximately 60°C. Additionally, inspired by shark noses, a composite material of fluoroelastomer and a specially designed hydrophobic ionic liquid has been designed for an elastic, self-healing, and extremely sensitive heat sensor [98]. It can discern temperature differences as low as 0.01 K, with a resolution of 0.001 K. The sensor can reliably operate in seawater or under pressures of up to 110 MPa, without any encapsulation. In summary, bioinspired thermal iontronics devices represent an innovative and promising technology by the ionic thermoelectric effect, enabling highly efficient energy conversion and temperature recognition. This opens up new possibilities for the development of energy technology, artificial intelligence, and biomedical technology.

3.5. Piezo driven ion dynamics

The human skin encompasses various micro mechanoreceptor cells, such as Merkel cells (~10–50 μm), Meissner's corpuscles (~30–100 μm), and Pacinian corpuscles (~200–500 μm), enabling the highly sensitive transmission and perception of a wide range of pressures [99], as shown in Figure 7a. Under resting conditions, there is an internally maintained polarized potential in the membrane (typically ranging from −40 mV to −80 mV relative to the extracellular side), owing to salinity gradients of ions (such as Na^+/K^+) on both sides of the membrane [100,103]. Concurrently, ion channels would remain closed and there is no ions flow across the membrane. When external mechanical stimulus induces physical deformation of the mechanoreceptor cell membrane, ion channels open, and allowing Na^+ ions to pass through the cell membrane, thereby generating an action potential. Inspired by this process, piezo driven ion dynamics could be used in energy harvesting and sensing devices. For instance, as shown in Figure 7b, the nitrogen-doped carbon nanofibers were coherently embedded with SnO_2 /$BaTiO_3$ heterostructure as a sodium-ion battery anode [101]. It could utilize the local potential generated by the piezoelectric effects of $BaTiO_3$ to facilitate the diffusion dynamics of Na^+ and enhance the rate performance of the SnO_2 anode. This configuration demonstrates high-capacity retention (82% over 2000 cycles at 2 A g^{-1}) and remarkable long-term cyclability (183.4 mAh g^{-1} after 10,000 cycles at 5 A g^{-1}). Similarly, self-charging supercapacitors have been fabricated by using a silicone oxide membrane as the electrode, and a silicone oxide-based polymer piezoelectric fiber membrane fixed with an ionic gel as the electrolyte [104]. It was demonstrated that it has the self-charging capability under varying levels of compressive force, with a maximum voltage of up to 207 mV. Furthermore, to enhance the electrochemical performance of Li^+ based batteries or supercapacitors, a multifunctional Li^+ pump based on ferroelectric materials has been developed to improve the Li^+ migration by generating an internal electric field [105]. Not only could it effectively promote Li^+ dynamics, but also it enhances the interface stability of lithium metal/electrolyte by Li^+ uniform deposition on the surface. Full battery with Li^+ pump exhibits excellent performance, with a capacity over 1000 mAh g^{-1} and remaining stable even after 100 cycles.

In the field of piezo-iontronics, ionic tactile systems composed of deformable ion-conductive materials represent a novel adaptable sensing platform. It is designed to emulate the tactile sensing characteristics of human skin for potential applications in artificial skin technologies. Various ionic conductors, such as ionic liquids, ionic gels, and

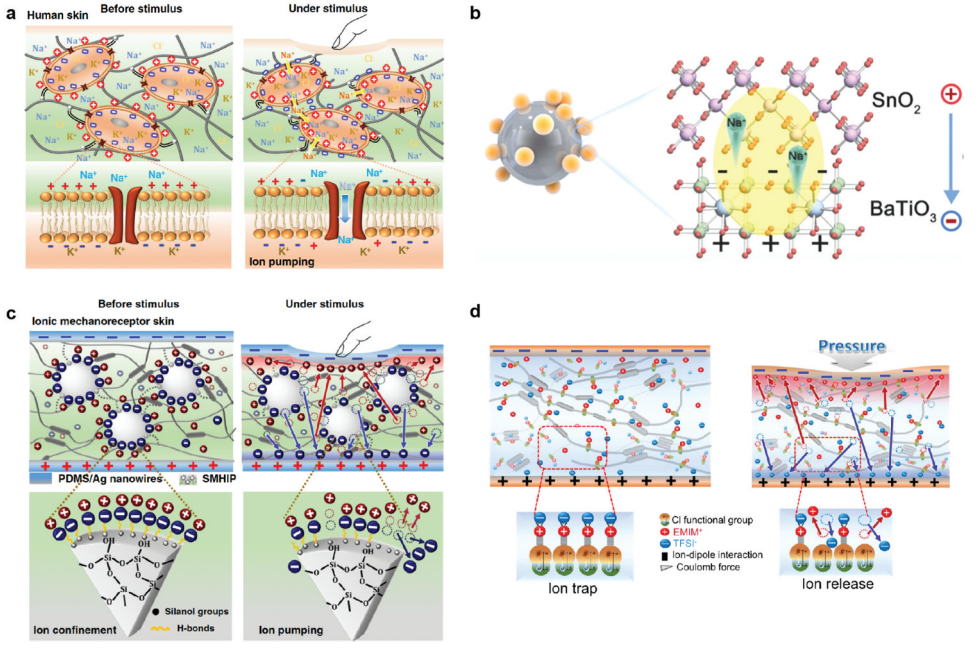

Figure 7. (a) Schematic illustration of human skin as a multicellular organ consisting of various mechanoreceptor cells. Once stimulated, Na^+ ions are pumped across through the ion nanoconfined channels, leading to depolarization of the cell membrane and the establishment of an action potential. Reproduced with permission [100]. Copyright 2019, Springer nature. (b) Schematic illustration of the local potential produced by piezoelectric and ferroelectric effect of $BaTiO_3$. The local potential may boost Na^+ diffusion kinetics across the crystal and promote rate performance of SnO_2 anode. Reproduced with permission [101]. Copyright 2021, Elsevier BV. (c) Schematic illustration of the piezocapacitive ionic skin that composed of the synthetic multicellular hybrid ion pump (SMHIP) film sandwiched between silver nanowires/polydimethylsiloxane flexible electrodes. Once induced pressure, the ions could be driven from the surface of the silica microstructure due to the breaking of H-bonds and establishment of EDL at SMHIP/electrode interfaces. Reproduced with permission [100]. Copyright 2019, Springer nature. (d) Schematic illustration of the piezocapacitive device consisted of the Cl-functionalized iontronic pressure sensitive material (CLiPS) film. It could drive ions owing to pressure-impelled breaking of ion – dipole interactions under deformation and EDL formation at the CLiPS/electrode interface. Reproduced with permission [102]. Copyright 2022, Springer nature.

hydrogels, have been employed to realize ionic tactile systems with human skin-like perception attributes [106,107]. They are also adept at effectively perceiving pressure, strain, shear, torsion, and another external stimulus. For instance, as shown in Figure 7c. a multi-cell mixed ionic pump, utilizing a confined ionic liquid composition on silicon dioxide microstructures, has been developed for highly sensitive ionic tactile skin [100]. It could exhibit sensitivities within a broad pressure range from 0–135 kPa and a sensitivity of 48.1–5.7 kPa^{-1}. Here, the silicon dioxide microstructures are constructing an effective ionic constraining matrix that could dynamically confine the ionic fluid under external stimulus, which is the pivotal for this ultra-sensitive artificial mechanoreceptor skin. Additionally, a Cl-functionalized ionic-electronic piezoresistive material has been reported [102] as shown in Figure 7d. By introducing Cl-functionalized moieties into a polyurethane matrix, it could achieve ultra-fast and autonomous self-repair rates (4.3 μm min^{-1}) with

high efficiency. This strategy also promotes effective ion dynamics control, as the Cl groups could capture ions by the ion-dipole interaction system and yield exceptional pressure sensitivity (7.36 kPa^{-1}). In the future, such iontronic mechanoreceptors can be embedded in medical devices for monitoring patients' physiological parameters, as well as in robotic hands for tactile perception, and facilitating the enhanced interactions with the environment.

3.6. Magnetic field driven ion dynamics

Magnetic fields exert a remarkable influence on the migration, positioning, and navigation of creatures [108], especially in the birds. Within the beaks of pigeons, magnetic clusters are directly linked to mechanosensitive ion channels in the terminals of the trigeminal nerve [109]. Once these magnetic clusters sense changes in the Earth's magnetic field, the mechanosensitive nanoconfined ion channels would be stimulated to open, and allowing cations (such as Na^+ and Ca^{2+}) to flow into the cell to elicit a neural action potential [109,110], as shown in Figure 8a. In artificial systems, there are some studies involving the magnetic fields driven ion dynamics, achieving stable and efficient smart liquid gating. For instance, a magnetic-gated nanofluidic system, that integrated with super-hydrophilic nanochannels and reconfigurable ferrofluids, has been developed [111] as shown in Figure 8b. By adjusting the spatial configuration of the ferrofluid, it can control the opening and closing state of the nanochannels, achieving an ultrahigh selectivity ratio of up to 10,000 and outstanding stability over up to 130 cycles without performance degradation. Additionally, magnetic fields can be employed to control the transport of ions in confined spaces, obtaining ionic currents. For example, a tartaric acid modified chiral magnetic iron oxide (Fe_3O_4) film with nanoconfined channels was prepared [112] as shown in Figure 8c. By tuning the interaction between the magnetic and electronic dipoles and combining it with the nanoconfined ion transporting, a stable ionic current of 6.8 nA was successfully obtained, and it can be repetitively used in 30 magnetic switching cycles. Moreover, the preparation of bioinspired nanoconfined channels also requires the coordinated action of magnetic and electric fields to control ion bombardment. These technologies play a pivotal role in the research and technological development of integrated circuits, nuclear physics experiments, and nanotechnology.

4. Conclusion and prospective

A universal feature of creatures is their ability to respond to multiple stimulus, which will cause ion flow at the cellular and subcellular levels in biological systems. The bioinspired ionic control in the nanoconfined spaces enables iontronics and has demonstrated tremendous potential in energy and information flow. In this review, we have comprehensively presented various bioinspired methods for ionic control to realize multifunctional iontronics, encompassing chemical potential, electric field, light, heat, piezo, and magnetic field etc. In the realm of energy, iontronics could offer a versatile platform for efficiently harvesting natural high-entropy energies like osmotic, photonic, and thermal energies. Especially by manipulating the shape and surface chemistry of the nanoconfined channels and by coupling the ionic-electronic interfaces, a substantial enhancement in energy harvesting efficiency could be achieved [7,9,55]. This breakthrough opens up

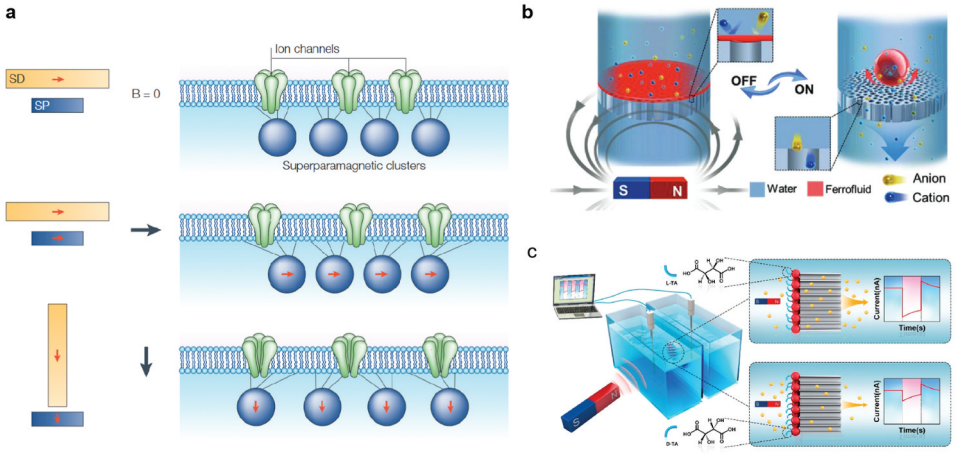

Figure 8. (A) a hypothetical transduction mechanism based on interacting clusters of superparamagnetic crystals located in the membranes of neurons. Depending on the orientation of the external field, the clusters will either attract or repel each other, deforming the membrane and possibly opening or closing ion channels. Reproduced with permission [108]. Copyright 2005, Springer nature. (b) Schematic illustration of a magnetic gated nanofluidic reversibly opened or closed by removal or application of a permanent magnet. Application of the magnet will cause the ferrofluid to cover the surface of the nanopores and blocks ion or molecular transport. Reproduced with permission [111]. Copyright 2019, John Wiley and sons. (c) Schematic illustration of the chiromagnetic Fe_3O_4 nanoconfined ion channel, showing the L-tartaric acid (TA)-modified channels could obtain 7.91 times more magnetron ion current than the D-TA-modified channels. Reproduced with permission [112]. Copyright 2022, American Chemical Society.

new avenues for the development of energy scavenging technologies. In the domain of sensing and computation, inspired by the neural behavior in biological systems, bioinspired iontronics could provide a unique opportunity to construct ionic memristor and synaptic neural networks for neuromorphic computing architectures [21,52,113]. Unlike traditional electronics based on von Neumann computing architecture, the Boolean logic computing architecture based on the neuromorphic transistor, ionic gate, ionic diode and circuits could avoid complex wiring and have higher energy efficiency in programmable logic analog. Additionally, considering factors involved in other biological processes, such as sound wave frequencies, amplitudes, phases and even magnetic field to control ions, it may offer an avenue for expanding the application scope of bioinspired iontronics in the future [114–116]. The combination of different stimulation methods may enable the realization of more complex, multifunctional bioinspired iontronics, capable of simultaneously performing functions like sensing, energy storage, and data processing.

Up until now, there have been several challenges in the bioinspired iontronics. The first challenge lies in our limited understanding of biological processes. For instance, the underlying mechanisms of how the nervous system achieves functions like learning, memory, and temporal ordering require further in-depth investigation. Achieving more complex computations, including machine emotions, consciousness, constitutes a developmental direction for simplified models like Spiking Neural Networks (SNN) and Convolutional Neural Networks (CNN) at present [117,118]. The second challenge lies in the need to further explore the potential of ions as charge carries per se. For instance, ions

inherently carry information, and ion circuits developed based on ion-based memristors and transistors have the capability to simulate neural networks, perform neuromorphic computations, and engage in pattern recognition tasks [119]. However, the majority of current research has only achieved basic ion logic gates, with few applications combining logic computation and power supply. Hence, advancing toward the realization of more complex ion logic circuits represents a paradigm for the comprehensive control and utilization of ions. The third challenge lies in the integration of the iontronics systems [30,120]. For example, it is still difficult to realize the efficient commercial energy management system and the in-sensor ionic logic circuits based on neural computing networks. The optimization of nanofluidic materials (length, surface charge, ionic conductance, etc.), low-cost preparation technologies [121,122] (printing, drop casting and laser engraving, etc.) and characterization methods may provide feasible ways to solve this problem. The fourth challenge is the need for continued exploration and optimization of the ionic-electronic coupling interface in bioinspired iontronics. Through precise design and control of the structure and properties of the ionic-electronic interface, it is possible to achieve accurate regulation of ion transport rates, selectivity, and stability, thereby enhancing the overall performance of the device while reducing energy consumption. Furthermore, the development of noninvasive processes for the ionic-electronic interface and biological interfaces holds the potential to realize more effective bio-electronic hybrid systems, such as authentic ionic eyes, skin, and prosthetics. This will offer new possibilities and sustained advancement for bioinspired iontronics in healthcare and biosensing applications. Last but not least, the long service life of iontronics is an important challenge, facing problems such as dehydration, mechanical durability, and repeatability [123]. In the future, the development of all-solid iontronics, or packaging and surface treatment, is necessary to protect the ionic system. Selection of the proper bioinspired ion control approach should be determined based on their unique scenarios. Factors, such as efficiency, stability, cost, and scalability etc. should also be considered when using iontronics.

In summary, the application of bioinspired ion control methods in the research of energy and sensing holds immense potential. With a deeper understanding of bioinspired ionic control and the continuous development on multifunctional iontronics and iontronics integrated systems, we are confident in pushing forward future computation architecture and paradigm with higher energy efficiency.

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