Chinese Journal of
POLYMER SCIENCE

REVIEW

https://doi.org/10.1007/s10118-026-3651-4
Chinese J. Polym. Sci. **2026**, *44*, 1681–1694

Soft Polymer Iontronics for Neuromorphic Intelligence

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Abstract Ion-conducting soft polymers, encompassing hydrogels, ionogels, and organic mixed ionic-electronic conductors, have emerged as distinctive material platforms for neuromorphic electronics, fundamentally diverging from the rigid paradigms of silicon and metal oxides. By enabling volumetric ionic transport and intricate electrochemical interactions within mechanically compliant matrices, these materials offer a biologically inspired route for emulating synaptic plasticity, memory consolidation, and adaptive sensing. With artificial synapses serving as the cornerstone of neuromorphic architectures, this review highlights recent advances in four classes of polymer-based composites: ion-gated transistors, electrochemical random-access memory cells, nanofluidic memristors, and soft ionic synapses. We dissect the physical mechanisms underlying plasticity: electric double-layer gating, volumetric electrochemical doping, nanofluidic confinement, and distinct viscoionic relaxation effects stemming from the macromolecular nature of the host matrix. Emphasis is placed on the interplay between ionic dynamics and polymer structural evolution in shaping the device functionality, energy efficiency, and multimodal responsiveness. We identify the critical challenges in switching speed, hydrolytic stability, and large-scale integration while highlighting emerging opportunities at the interface of biology, soft robotics, and in-sensor computing. Ion-conducting polymers, by uniquely combining brain-like ionic signaling with soft-matter versatility, are expected to play a central role in the next generation of intelligent, bio-integrated systems.

Keywords Ionotronics; Ion-conducting polymers; Neuromorphic functions; Soft ionic synapses; Electrochemical memory

Citation: Liu, S.; Zhan, H.; Zhang, L. W.; Wang, Z. Z.; Lei, Z. Y. Soft polymer iontronics for neuromorphic intelligence. *Chinese J. Polym. Sci.* **2026**, *44*, 1681–1694.

INTRODUCTION

Neuromorphic electronics aims to emulate the computational architecture of the brain by utilizing physical devices that mimic the analog, parallel, and adaptive nature of neurons and synapses.[1–3] While traditional solid-state electronics have achieved remarkable success in computational density, they fundamentally rely on electronic charge carriers within rigid inorganic lattices.[4] In stark contrast, biological systems process information through the transport of ions within soft, aqueous environments, enabling energy-efficient signaling and seamless integration of sensing and processing.[5] In recent years, ion-conducting soft polymers have emerged as pivotal materials for bridging the dichotomy between conventional hard electronics and biological soft matter.[6–9] These polymers, which include organic mixed ionic-electronic conductors, polyelectrolyte hydrogels, and ionogels, possess the unique capability to transport ionic charges through their bulk volume while often supporting electronic transport along the polymer backbone.[10–14] This mixed conduction capability allows for the fabrication of devices that do not merely switch structurally but also respond and adapt *via* coupled ionic-electronic fluxes, providing a direct physical analog to the electrochemical dynamics of biological synapses.[15]

The fundamental advantage of utilizing soft polymers for neuromorphic functions is their molecular structure and bulk properties. Unlike inorganic semiconductors, in which ionic interactions are typically confined to the surface or grain boundaries, conducting polymers behave as volumetric sponges for ions.[16–18] This volumetric capacitance allows high-density charge storage and efficient signal modulation at low operating voltages, typically below one volt, which matches the electrochemical window of biological fluids.[19] Furthermore, the macromolecular matrix of these polymers is not a static scaffold, but a dynamic environment. The motion of ions within the polymer is intimately coupled with segmental motion and conformational changes of the polymer chains. This coupling introduces rich, time-dependent behaviors, such as viscoelastic relaxation and swelling, which naturally emulate the complex temporal dynamics of synaptic plasticity, including the transition from short-term to long-term memory.[20] Consequently, ion-conducting polymers offer a material platform where the physics of the device is intrinsically neuromorphic, reducing the need for complex external circuitry to simulate neural behavior.[21]

This review provides a comprehensive analysis of the four

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Special Topic: Functional Gels
Received December 26, 2025; Accepted March 10, 2026; Published online May 12, 2026

major classes of ion-conducting polymer devices that have been defined over the past decade. We first explore ion-gated transistors (IGTs), focusing on the mechanisms of channel conductivity modulation and simulation of synaptic weight updates *via* reversible electrochemical doping. We then examine electrochemical random-access memory cells (ECRAMs), which leverage the stability of ion insertion into polymer lattices to achieve nonvolatile, linear weight states essential for in-memory computing. Subsequently, we investigated nanofluidic memristors, where ion transport is regulated by the spatial confinement effects of the nanochannels, enabling the modulation and storage of conductance. Finally, we discuss soft ionic synapses, a class of devices that eschew electronic currents entirely in favor of manipulating ionic fluxes within hydrogel networks, representing the ultimate convergence towards bio-identical signaling. Through this analysis, we emphasize the physical mechanisms unique to soft matter, such as electric double-layer gating, bulk ion insertion, and viscoionic relaxation, and discuss how the material engineering of the polymer structure determines the trajectory of this rapidly evolving field.

OPERATING PRINCIPLES AND ARCHITECTURES

There are distinct differences between inorganic neuromorphic devices and soft-polymer ionic neuromorphic devices. The former primarily relies on the transport and modulation of electrons or holes within solid inorganic media (charge trapping, ferroelectric polarization, and phase change). Their dynamics are typically dominated by electronic processes, which are fast and stable but present an intrinsic disconnect from biological ionic processes. The latter is established by coupled ionic and electronic mechanisms. The migration, accumulation, and interfacial rearrangement of ions within the electrolyte and polymer networks were directly mapped to the weight modulation and temporal behaviors. Consequently, these devices more readily achieve multiscale temporal plasticity and soft interface compatibility, similar to biological synapses. Table 1 summarizes the comparison of the key characteristics of the two device categories.

Viscoionic relaxation is a fundamental physical feature that distinguishes soft ionic neuromorphic devices from traditional solid electronic devices. This phenomenon originates from the mutual coupling among ion migration, viscoelastic dynamics of the polymer matrix, free volume fluctuations, and local coordination or ion-pair formation. Ion movement perturbs the polymer chain configuration and local free volume. The recovery of these structural elements modulates the effective mobility and diffusion coefficient of the ions, thereby introducing significant nonequilibrium hysteresis and slow relaxation.[22–24] Although this is a complex process, it can be approximately described by the Stokes-Einstein equation, $D = \frac{K_B T}{6\pi\eta r}$, where D is the ionic diffusion coefficient, k_B is the Boltzmann constant, and T is the temperature. Variable r represents the effective radius of the ion, including its solvation or coordination shell, while η denotes the time-dependent effective viscosity jointly determined by polymer chain relaxation, ion-polymer interactions, and free volume evolution. Therefore, η is typically not constant in soft polymer systems. When an external field is applied, local structures are driven to rearrange, open, or block the channels. After the external field is removed, the structure slowly recovers according to its viscoelastic timescale, causing D to exhibit a slow decline and history dependence. This is the fundamental reason why viscoionic relaxation endows the devices with memory characteristics. In terms of timescales, viscoionic relaxation typically encompasses both rapid ion migration processes on the order of milliseconds to seconds and slower polymer relaxation and structural reconfiguration processes lasting from seconds to hours or even longer. The former dominates the transient response and short-term conductance modulation of the device, whereas the latter dictates the ion retention, interfacial rearrangement, and bulk structural stability, serving as a major source of hysteresis, non-volatility, and history-dependent behavior. This coexistence of multiple kinetic timescales provides a physical foundation for achieving both fast responsiveness and slow forgetting within a single-material system. At the functional level of neuromorphic engineering, viscoionic relaxation offers mechanistic support for the natural transition from short-term plasticity (STP) to long-term plasticity (LTP). Under weak stimulation, ions primarily undergo reversible migration, and the device exhibits rapidly decaying short-term memory. Under repetitive or strong stimulation conditions, ions form more stable bound states within the polymer matrix, accompanied by chain relaxation and local structural evolution, thereby realizing long-term plasticity with high retention characteristics. It is worth emphasizing that this process does not generally require the introduction of explicit physical phase transitions or irreversible chemical reactions. Instead, it originates from the unique viscoelastic dynamics of soft-matter systems. Consequently, it possesses universal applicability across IGTs, ECRAMs, nanofluidic memristors, and soft ionic synapses, and can be custom-tailored through molecular design and structural regulation.

Ion-gated transistors employ an electrolyte or ionic dielectric to control the conductance of a semiconductor channel, effectively coupling ionic fluxes to electronic currents with an exceptional efficiency.[25] Structurally analogous to conventional field-effect transistors, IGTs consist of an electrolyte, a channel, and three terminals (gate, source, and drain) (Fig. 1a); however, their operating principles diverge fundamentally. When a gate voltage is applied, cations or anions in the electrolyte migrate to the electrolyte-channel interface, inducing modulation in the channel conductivity. Depending

Table 1 Comparison of inorganic devices and polymer devices.

	Transport carrier	Young's modulus	Biointerface impedance	Environmental sensitivity	Carrier mobility ($cm^2 \cdot V^{-1} \cdot s^{-1}$)	Energy efficiency	Wearable integration
Inorganic device	Electron/hole	High	High	Insensitive	High (10^3–10^5)	Low	Inappropriate
Polymer device	Ion and electron	Low	Low	Sensitive (Humidity, temperature, and pH)	Low (10^{-4}–10^2)	High	Appropriate

https://doi.org/10.1007/s10118-026-3651-4

on the ionic permeability of the channel material, IGTs are classified as electric double-layer transistors (EDLTs) with impermeable channels or organic electrochemical transistors (OECTs) with permeable channels (Fig. 1b).[26–29]

In EDLTs, ions are redistributed at the interface to induce carrier accumulation in the channel, creating an electric double layer (EDL) that exerts an electrostatic gating effect. In contrast, OECTs involve ions penetrating the volume of a redox-active polymer channel *via* a reversible electrochemical process coupled with a charge-balancing electron injection from the source. This process generates a volumetric capacitance that is often several orders of magnitude higher than the surface capacitance. This phenomenon, which is central to the operation of organic electrochemical transistors, allows for high transconductance and signal amplification, enabling a single device to integrate weak presynaptic inputs with a gain analogous to that of a biological neuron.

All-polymer ECRAMs and OECTs share significant similarities in device architecture and ion-electron coupling mechanisms. Both typically employ a three-terminal configuration and rely on ion injection from the electrolyte to modulate the conductance of the polymer channel, thereby enabling continuous and reversible analog weight updates. This volumetric doping mechanism provides both devices with common advantages including low-voltage operation, high linearity, and excellent analog plasticity. However, the underlying mechanisms and functional objectives of all-polymer ECRAMs and OECTs differ fundamentally. OECTs primarily emphasize transient-field-enhanced electrochemical modulation. Their conductive states generally require the maintenance of an applied gate voltage. Once the bias is removed, the ion distribution tends to relax, resulting in volatile or weakly non-

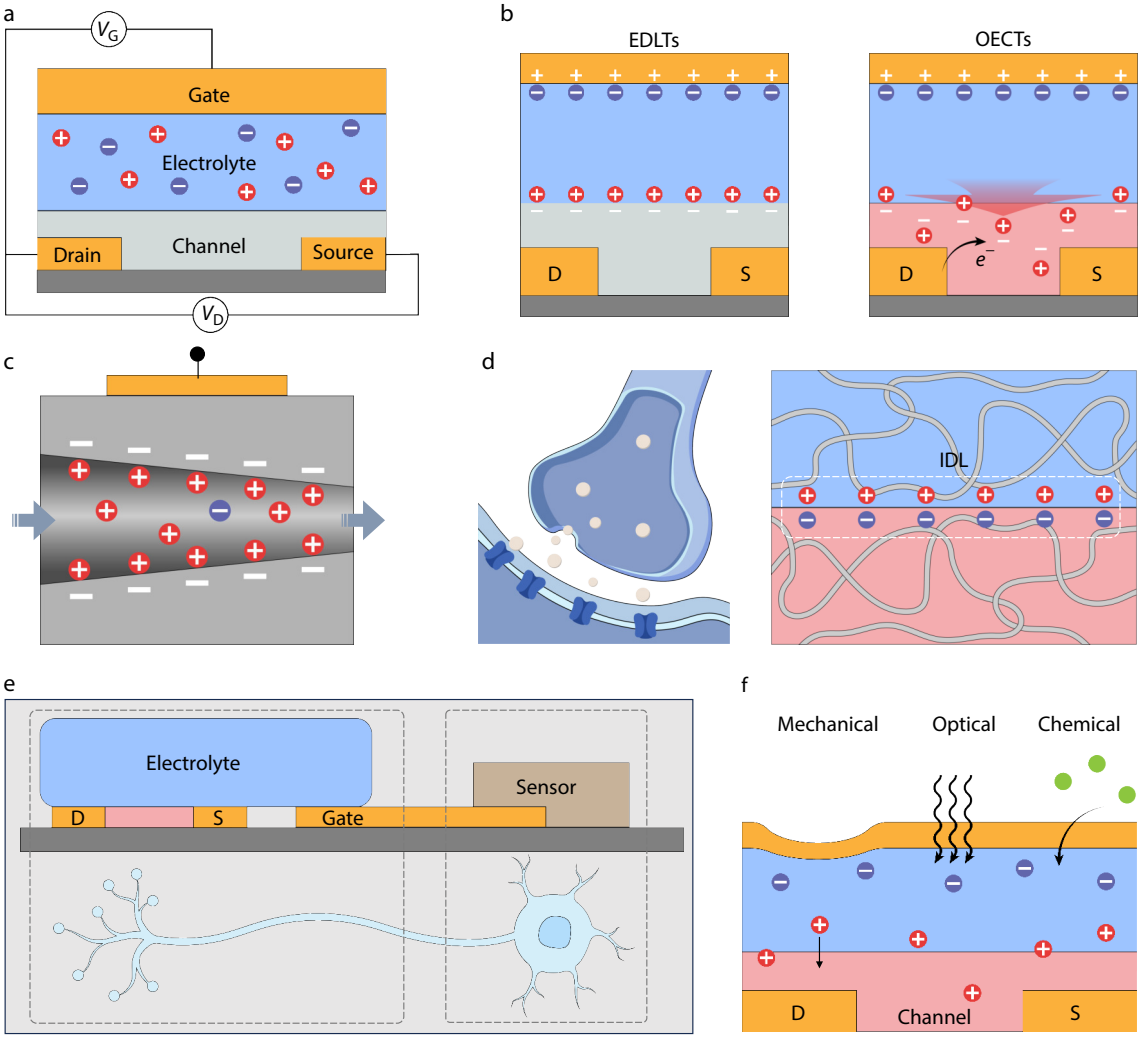

Fig. 1 Mechanisms and architectures of soft polymer iontronics for neuromorphic intelligence. (a) Typical three-terminal configuration of an IGT, comprising a gate, source, drain, and electrolyte; (b) Distinct operating modes: the EDLTs (left) rely on interfacial electrostatic gating, while the OECTs (right) involve volumetric electrochemical doping; (c) A nanofluidic memristor where ionic transport is modulated by surface charge interactions within a nanoconfined channel; (d) A soft ionic synapse (right) that mimics the structure and neurotransmitter diffusion of a biological synapse (left); (e) A bio-inspired neuromorphic system achieved through the integration of a sensory element with a synaptic transistor; (f) monolithic in-sensor computing architecture that intrinsically integrates sensing (*e.g.*, light, chemicals), memory storage, and signal processing into a unified structure.

https://doi.org/10.1007/s10118-026-3651-4

volatile behavior. In contrast, all-polymer ECRAMs achieve nonvolatile modulation of channel conductance through highly stable redox reactions. Their programmed states can be retained without a gate bias, functioning more like true electrochemical random-access memory cells. Furthermore, because ECRAMs are specifically designed for high-precision analog storage, their evaluation metrics such as write symmetry, state retention, and cycling endurance differ significantly from those of OECTs, which are primarily targeted at signal amplification and sensing applications. Given these differences, although all-polymer ECRAMs overlap with OECTs in material systems and device formats, their distinct operating principles, intrinsic non-volatility, and specific roles in neuromorphic storage warrant a dedicated, systematic discussion rather than simply grouping them under the OECTs framework.[30,31]

Nanofluidic memristors adopt diverse structural forms but fundamentally rely on regulating the electrostatic interactions between surface charges and ionic species within nanoconfined channels.[32–35] This regulation modulates ionic transport to achieve history-dependent conductance (Fig. 1c). Although these devices exhibit memristive behavior through ion transport, their electrochemical operation essentially depends on the solid-state, electron-based nanochannel materials, and applied electric fields. Additionally, the requirement for highly precise and rigid nanochannels, along with bulk fluid reservoirs as ion storage media, poses significant barriers to their miniaturization and large-scale integration.

Recently, soft ionic synapses based on heterogeneous hydrogel-ionogel interfaces have emerged. By completely eliminating the electronic currents, these devices can be viewed as a functional progression from nanofluidic memristors. They advanced confined ion transport beyond simple conductance modulation towards system-level information encoding and spatiotemporal processing. This shift reduces the reliance on finely fabricated rigid nanostructures, placing greater emphasis on coupling among ions, polymer networks, and temporal dynamics within soft matter systems. Therefore, while both classes share similar electron-free memristive principles, nanofluidic memristors act more as fundamental, physics-driven ionic memory units, whereas soft ionic synapses represent an extension of these mechanisms into a bio-inspired information processing paradigm. Their core mechanism involves the formation of an ionic double layer (IDL) *via* anion-cation alignment at the junction.[36,37] By tailoring the polymer network architecture and ionic species, they regulate the ionic flux to emulate biological action potentials and neurotransmitter diffusion (Fig. 1d).[38]

Integrating these artificial synapses with sensory components establishes ionic neuromorphic circuits (soft iontronics). In this architecture, sensors transduce external stimuli into signals that are stored and processed by synaptic devices, mirroring the biological dendrite-axon-synapse pathway (Fig. 1e).[20,39] Current integration strategies for neuromorphic systems can be broadly categorized into three approaches: planar circuit integration, vertical stacking integration, and monolithic integration. Planar integration typically connects the output of independent sensors directly to the input of the synaptic devices. This approach offers a clear structural design and functional decoupling, facilitating independent optimization of device performance and circuit debugging. However, it often occupies a large system footprint, introduces additional signal transmission delays and energy consumption, and limits further scaling of the system. In comparison, the vertical-stacking strategy layers the sensing and synaptic components within the same spatial area. This increases the integration density and shortens the signal pathways, thereby improving response speed and energy efficiency. Nevertheless, this method imposes strict requirements on material compatibility, interlayer interface stability, and fabrication complexity. Monolithic integration further merges sensing and synaptic functions into a single device or material system, showing significant advantages in reducing system complexity and minimizing redundant signal conversion. However, this strategy usually relies on coupled multiphysics ionic dynamics, which present challenges in signal decoupling. Realizing fully soft neuromorphic systems depends on developing soft ionic materials that simultaneously possess sensing, memory, and computational capabilities. Building upon these materials, researchers must achieve synergistic optimization of high mechanical compliance, multiple stimulus decoupling, low power consumption, and system-level stability (Fig. 1f).[40–42] By introducing modular soft device designs, stretchable interconnects, and energy harvesting strategies, neuromorphic circuits are expected to evolve from discrete component integration into highly biomimetic and continuously deformable intelligent systems. This evolution lays the foundation for applications in wearable electronics, soft robotics, and biological interfaces.

IGTs: Electrolyte Gating and Synaptic Response

The plasticity in IGTs, which is the ability to change the conductance strength over time, is governed by the specific interaction between the distinct ionic species and the polymer matrix. The EDLTs predominantly exhibited STP.[43] This characteristic mimics the rapid strengthening or weakening of synapses, known as paired-pulse facilitation or depression, which typically arise from transient ionic dynamics. When a gate pulse is applied, ions rapidly accumulate at the polymer interface or shallowly dope the surface of the film. Upon removal of the bias, these ions diffused back into the electrolyte driven by concentration gradients, causing the channel conductance to decay to its initial state. The time constant of this decay is dictated by the diffusion coefficient of ions within the polymer network and the geometry of the device. By tuning the hydrophilicity and mesh size of the polymer, researchers can engineer these timescales to match the biological short-term memory, ranging from tens of milliseconds to seconds.

Leveraging these dynamics, researchers have successfully constructed artificial afferent nerves by the planar integration of IGTs with organic ring oscillators and pressure sensors. These systems can integrate multiple voltage input signals, effectively emulating the signal processing of the sensory neurons (Figs. 2a and 2b).[44] However, such lateral integration approaches often suffer from large spatial footprint. To address this, monolithic vertical architectures have been developed that integrate graphene oxide channels, ion-gel gate dielectrics, and elastomeric sensing layers into a single stack.

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These devices achieve intrinsic synaptic functionality, successfully fusing slowly adapting (SA) and rapidly adapting (RA) postsynaptic responses within a synaptic array (Fig. 2c).[45]

Transitioning from short-to long-term plasticity involves deeper and more stable interactions between the ions and polymer chains. In OECTs, applying a sustained or high-magnitude gate voltage drives ions deep into the bulk of the polymer channel to compensate for electronic charge carriers. This reversible electrochemical doping or de-doping process is typically accompanied by the conformational reorganization of the polymer backbone, leading to effective structural swelling or contraction.

Recent studies have highlighted the role of viscoionic relaxation in stabilizing these states. As ions penetrate the dense polymer matrix, they can become trapped owing to steric hindrance or specific chemical interactions with the polymer side chains. The viscoelastic polymer chains relax slowly around the inserted ions, creating an energy barrier for their exit. This coupled relaxation results in hysteresis and nonvolatile retention of the conductance state, effectively emulating the long-term potentiation. The degree of nonvolatility can be controlled by molecular design, such as introducing side chains that sequester ions or using electrolytes with bulky ions that have low mobility within the polymer phase. For instance, an OECT-based artificial chemosensory

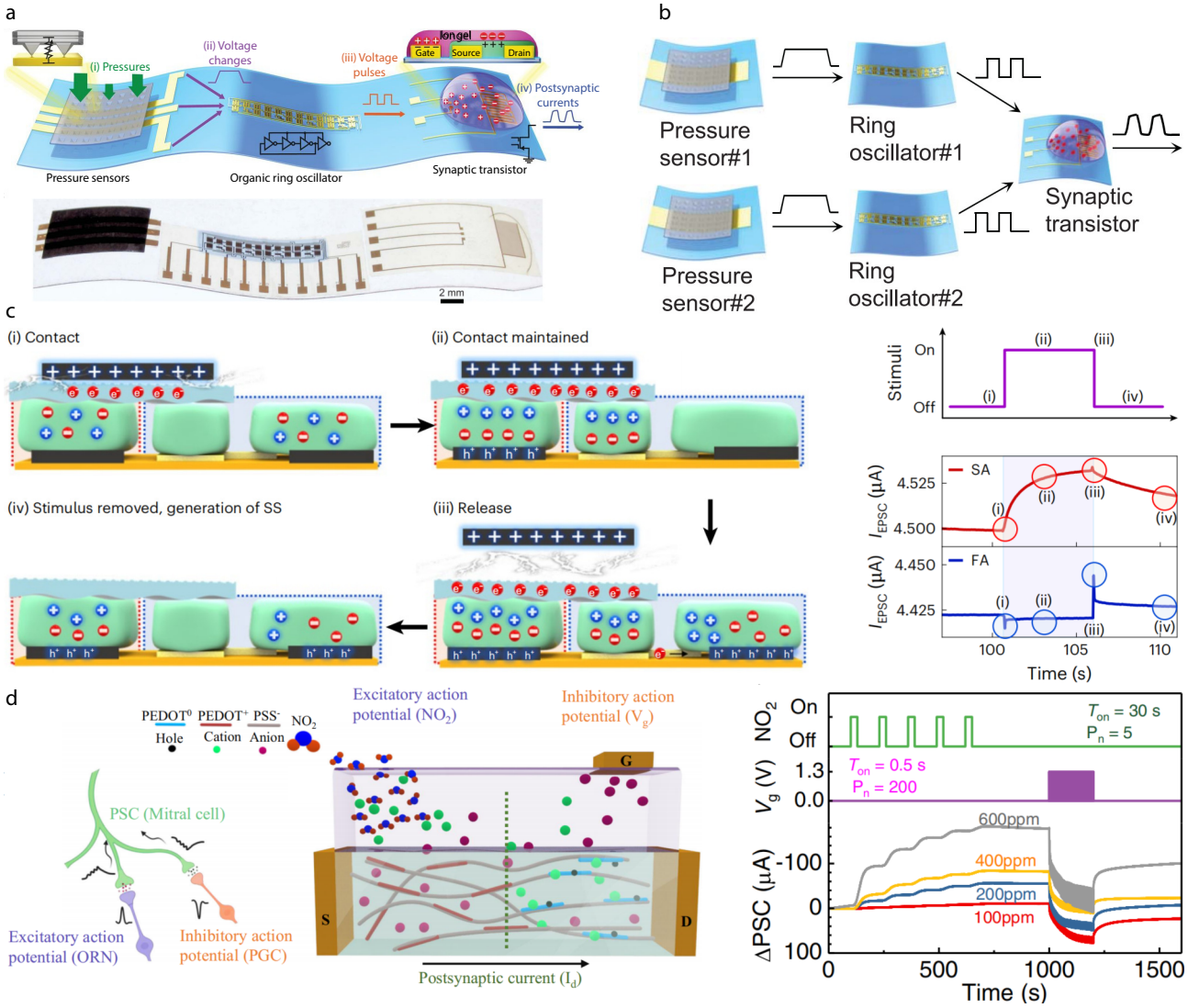

Fig. 2 IGT for integrated neuromorphic systems. (a) Structure of an artificial nerve comprising pressure sensors, organic ring oscillators for signal conversion, and ion-gated synaptic transistors; (b) Schematic showing the integration of multiple ring oscillators, each driven by a cluster of pressure sensors, to a common synaptic transistor (a, b: reproduced with permission from Ref. [44]; Copyright (2018), The American Association for the Advancement of Science); (c) Illustration of ionic migration in a monolithic artificial synapse under mechanical contact (left), enabling the generation of distinct slowly adapting (SA) and rapidly adapting (RA) postsynaptic signals (right) (Reproduced with permission from Ref. [45]; Copyright (2025), The Author(s), under exclusive licence to Springer Nature Limited); (d) An OECTs-based artificial synapse designed for multimodal processing, demonstrating dual responsiveness to electrical gating and chemical stimulation (*e.g.*, NO_2 gas) (Reproduced with permission from Ref. [46]; Copyright (2023), The Author(s)).

neuronal synapse has been demonstrated to mimic the release of neurotransmitters (ions) from a presynaptic terminal (ion gel) in response to electrical or chemical stimuli, triggering excitation or inhibition of the postsynaptic membrane (conductive polymer channel). Driven by ion penetration, the high volumetric capacitance enables OECTs to operate at low voltages, making them ideal for bio-plausible synaptic emulation (Fig. 2d).[46]

Beyond individual synaptic emulation, the dynamic properties of ion-gated transistors enable the implementation of complex neuronal functions such as spatiotemporal integration and filtering. The inherent delay associated with ionic migration through the polymer film acts as a physical memory trace, allowing the device to summate the inputs over time. This property has been exploited to create dendritic processing units and orientation-selective circuits that rely on a specific temporal overlap of signals. Furthermore, the compatibility of these soft transistors with aqueous environments allows their direct integration into biological tissues. Vertical organic electrochemical transistor architectures and fully printed stretchable arrays have demonstrated the ability to operate reliably under mechanical deformation, paving the way for neuromorphic skins to process tactile information locally before transmission.[47] The versatility of the ion-gated transistor platform lies in its ability to tune synaptic dynamics through material synthesis, transforming the polymer channel from a passive conductor into an active history-dependent computational element.

Polymer-based ECRAMs as Artificial Synapses

ECRAMs represent a specialized evolution of the electrochemical transistor, optimized specifically for nonvolatile analog weight storage in artificial neural network accelerators. In ECRAMs devices, the intrinsic physicochemical properties of the channel and electrolyte materials directly determine key performance metrics. First, ionic mobility and diffusion anisotropy dictate the switching speed and energy consumption: high ionic mobility, alongside continuous, low-barrier diffusion pathways, facilitates sub-microsecond or even nanosecond weight updates. Second, the stability of the reversible redox reactions and the electrochemical window directly govern state retention and cycle life. Intercalation or doping processes characterized by smooth potential variations, which avoid drastic phase transitions or structural collapse, are highly beneficial for achieving long-term stable modulation of multilevel conductance states. Finally, the mechanical compliance and tolerance for volume expansion cannot be overlooked. Maintaining the structural integrity during repeated ion insertion and extraction is a prerequisite for ensuring device reliability and consistency. Overall, an ideal ECRAMs requires the synergistic optimization of high ionic mobility, excellent redox reversibility, controllable electronic structure responses, and mechanical stability, thereby striking a balance between switching speed, retention, and operational endurance. These requirements have driven a material shift from inorganic intercalation hosts to all-polymer systems; for example, employing a PEDOT:PSS film partially reduced by poly(ethylenimine) (PEI) as the channel (Fig. 3a).[31] Such devices operate at low voltages and energy consumption, exhibiting over 500 distinct non-volatile conductance states within an approximately 1 V range while achieving high classification accuracy in neural network simulations (Fig. 3b).

The critical advantage of all-polymer ECRAMs over their inorganic counterparts is the mechanical compliance of the host lattice. Inorganic intercalation materials often suffer from phase transitions or lattice strain during ion insertion, which leads to nonlinear switching and structural degradation. In contrast, soft polymers can expand and contract to accommodate ions with minimal energetic penalties, resulting in highly linear and symmetric potentiation and depression. Another limitation of inorganic ECRAMs is their rigid, planar, and inherently 2D structure, which is not suitable for biological integration, and where direct stacking often impedes ionic transport. In contrast, the adoption of semiconducting hydrogels not only enables thickness tunability at the millimeter scale, but also provides tissue-level softness and biocompatibility.[48]

Material selection for polymer-based ECRAMs focuses on organic mixed ionic-electronic conductors that offer both high ionic mobility and stable redox states. Conjugated polymers, such as PEDOT:PSS, and more recently developed *n*-type polymers, such as poly(benzimidazobenzophenanthroline) (BBL) and glycolated polythiophenes, serve as ideal channel materials. The open, amorphous microstructure of these polymers facilitates the rapid diffusion of ions, enabling switching speeds that are competitive with traditional non-volatile memories while operating at significantly lower energy levels.[49] The switching energy is minimized because the process is governed by the Nernstian potential of the redox reaction, which typically requires only fractions of a volt to drive the state change. Furthermore, the amorphous nature of the polymer prevents the formation of grain boundaries that typically act as scattering sites or trap states in crystalline materials, thereby reducing the device-to-device variability and random telegraph noise.[50]

Although these intrinsic properties provide a foundation, the transition from single devices to large-scale integrated arrays imposes other requirements, particularly regarding the dynamic range between the highest and lowest conductance states. A wide dynamic range is critical for two reasons: on the one hand, it enhances tolerance to device-to-device variability. In large arrays, insufficient overlap of conductance ranges among individual devices, which is caused by fabrication variance, can severely degrade the accuracy of the neural computations. A broader dynamic range ensures a sufficiently common working window for all devices. On the other hand, an expanded conductance window effectively mitigates the impact of write noise and improves state precision. Optimizing the channel material is essential for addressing these challenges. For instance, advanced glycolated polythiophenes, specifically poly(2-(3,3-bis(2-(2-(2-methoxyethoxy)ethoxy)-ethoxy)-[2,2-bithiophen]-5-yl)thieno[3,2-b]thiophene) (p(g2T-TT)), have emerged as alternatives to the traditional PEDOT:PSS channels (Fig. 3c).[51] Comparative studies revealed that ECRAMs utilizing p(g2T-TT) achieved a significantly higher dynamic range, even under shorter write pulse durations (Fig. 3d). Beyond the switching magnitude, the use of p(g2T-TT) in conjunction with anhydrous electrolytes confers exceptional thermal stability, maintaining reliable read/write operations at elevated temperatures that are compatible with

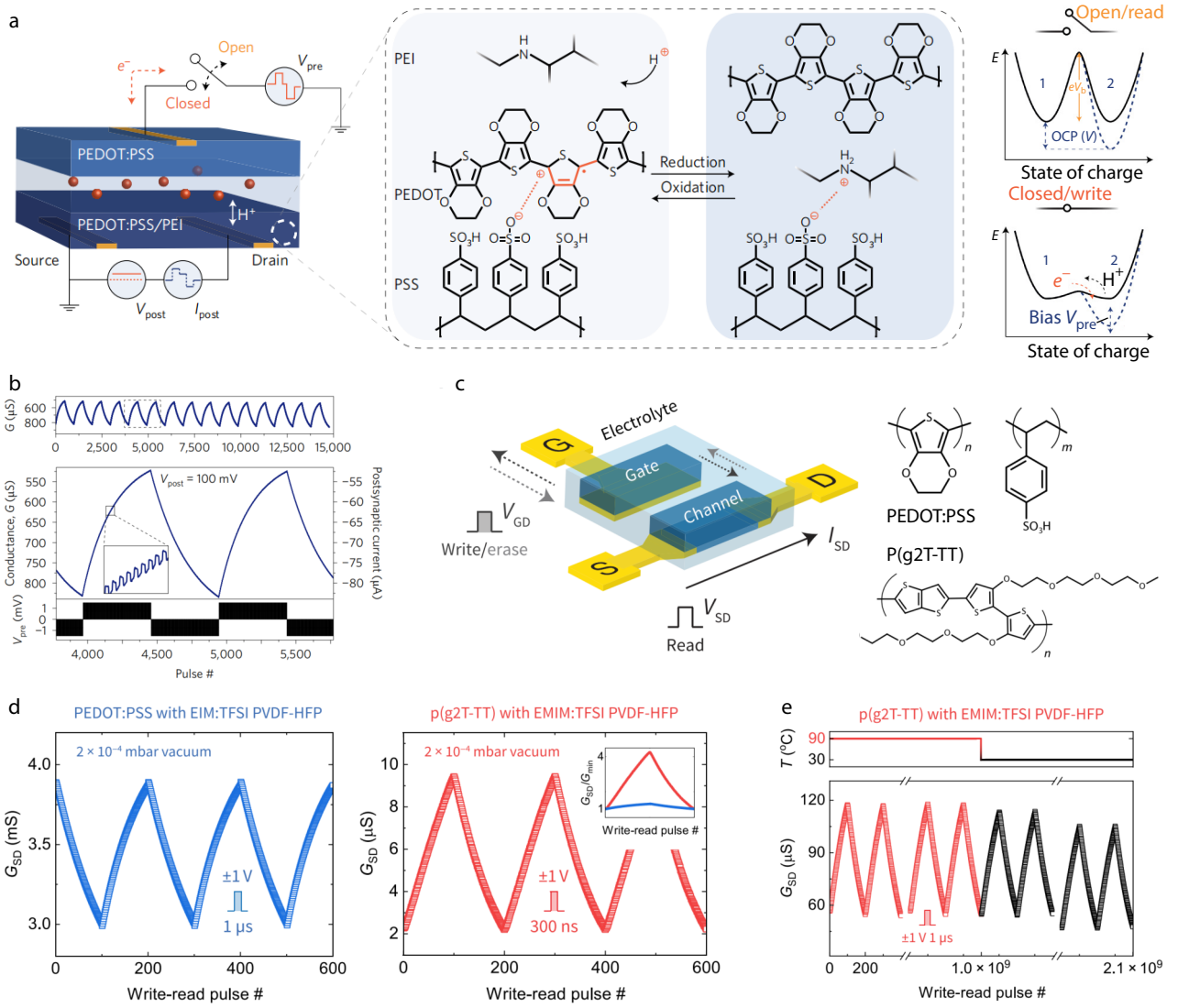

Fig. 3 ECRAM cells for non-volatile analog computing. (a) Structural design of an all-polymer ECRAM. The schematic (right) illustrates how the energy barrier between different redox states of the PEI-reduced PEDOT channel enables the effective decoupling of read and write operations; (b) Analog switching characteristics show the device capable of achieving 500 distinct conductance states with high precision and linearity (a, b: reproduced with permission from Ref. [31]; Copyright (2017), Springer Nature Limited); (c) The material evolution from standard PEDOT:PSS to the advanced glycolated polymer p(g2T-TT) as the channel material; (d) Comparative switching performance reveals that p(g2T-TT) maintains a superior dynamic range even under significantly shorter write pulse durations (300 ns) compared to PEDOT:PSS; (e) Long-term reliability testing under thermal stress, showing that p(g2T-TT)-based ECRAMs exhibit an endurance of >10^9 cycles at 90 °C without degradation in switching linearity (c–e: reproduced with permission from Ref. [51]; Copyright (2020), The American Association for the Advancement of Science).

integrated circuit environments (Fig. 3e).

A critical engineering challenge in polymer-based ECRAMs is balancing the trade-off between the switching speed and state retention. To achieve long retention times, ionic mobility must be low when the device is in the memory (read) state, but high during the programming (write) pulse. This can be addressed through interface engineering using solid polymer electrolytes or ionogels to confine ions and prevent spontaneous self-discharge. Recent innovations include the use of block copolymers, where one phase allows ion conduction and the other provides mechanical rigidity, and the design of polymers with specific ion-binding sites that stabilize the inserted species.[51] These molecular engineering strategies ensure that the synaptic weight remains fixed until a specific electrochemical driving force is applied, granting the device the necessary nonvolatility for offline learning tasks.[52]

Scaling polymer-based ECRAMs into high-density crossbar arrays poses challenges that are being addressed through advanced lithography and material processing. Because the memory element relies on the bulk volume of the polymer, reducing the device footprint requires maintaining a sufficient volume-to-surface ratio to preserve the dynamic range of the conductance modulation. Vertical stacking and three-dimensional patterning of polymer channels are emerging as viable solutions for increasing density. Moreover, the problem of ionic crosstalk, in which the operation of one device influences its neighbors through the electrolyte, is mitigated by patterning the electrolyte into discrete islands or using ion-

blocking encapsulation layers.[49] Despite these engineering hurdles, polymer-based ECRAMs are considered promising candidates for the hardware implementation of backpropagation algorithms, driven by their fundamental attributes of linearity, low switching energy, and extreme cycling endurance exceeding millions of cycles without degradation.

Nanofluidic Memristors: Ion Transport under Spatial Confinement

Nanofluidic memristors distinguish themselves by utilizing ion transport within nanoconfined spaces as the physical carrier for memory, where the conductance state depends not only on the instantaneous applied field, but also on the history of ionic distribution within the channel. Early investigations into single conical nanopores revealed characteristic memristive and memcapacitive behaviors, demonstrating that within charged nanopores, the interplay between finite ion mobility and the surface-charge-induced EDL creates a significant history dependence of the ionic current on the applied field (Fig. 4a).[53] These findings, supported by theoretical frameworks elucidating the intrinsic memristive properties of electrolytes under high spatial confinement (Fig. 4b), establish the physical foundation for this class of devices.[54]

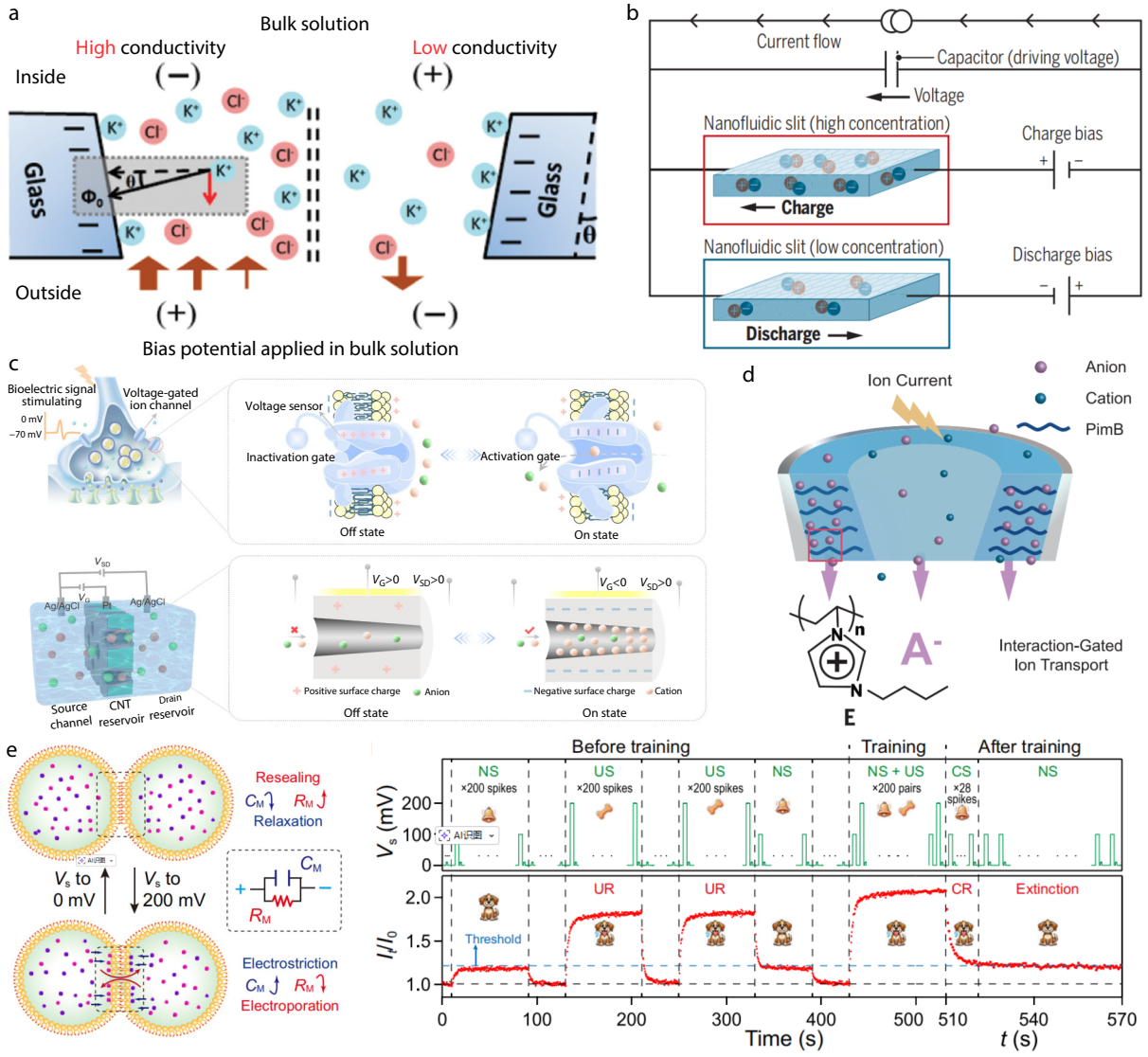

Fig. 4 Mechanisms and applications of nanofluidic memristors. (a) Schematic illustrating ion transport processes confined within a conical nanochannel, governed by the interactions between ionic flux and surface charges (Reproduced with permission from Ref. [53]; Copyright (2012), American Chemical Society); (b) Theoretical modeling of a neuronal circuit derived from the intrinsic memristive properties of electrolytes to high spatial confinement (Reproduced with permission from Ref. [54]; Copyright (2021), The American Association for the Advancement of Science); (c) Design of a bio-inspired nanofluidic memristor (bottom) mimicking biological voltage-gated ion channels (top), showing the transition between off and on states (Reproduced with permission from Ref. [55]; Copyright (2024), The American Association for the Advancement of Science); (d) A polyelectrolyte-confined fluidic memristor (PFM) where ion transport is regulated by interaction-gating within a channel lined with polyimidazolium brushes (PimB) (Reproduced with permission from Ref. [56]; Copyright (2023), The American Association for the Advancement of Science); (e) A bilayer droplet demonstrating synaptic plasticity and the realization of Pavlovian associative learning (classical conditioning) (Reproduced with permission from Ref. [57]; Copyright (2025), The American Association for the Advancement of Science).

Building on these physical foundations, researchers have engineered various channel architectures to emulate the synaptic functions. Initial designs leveraged rigid scaffolds, such as carbon nanotube-based nanofluidic ion-gated transistors, which demonstrated high on/off ratios of 10^4 at gate voltages as low as 1 V (Fig. 4c).[55] These devices have been successfully integrated into neuromorphic circuits to construct fundamental logic gates. Moving beyond simple electrostatic gating, complex polyelectrolyte-confined fluidic memristors (PFMs) have been introduced to enhance plasticity mechanisms. By employing *in situ* grown polyimidazolium brushes with a high charge density, these devices confined the fluid and introduced a mechanism based on the slow enrichment and release of anions within the brush layer (Fig. 4d).[56] This process generates stable, tunable hysteretic ion transport, enabling short-term synaptic plasticity and coupled electrochemical signaling in aqueous environments.

However, the aforementioned architectures rely fundamentally on rigid channels, which restrict flexible integration. Recent breakthroughs have addressed this limitation by realizing artificial synapses based entirely on soft-matter interfaces, eliminating the need for solid-state channels (Fig. 4e).[57] By achieving coupled memristive-memcapacitive characteristics, the design demonstrated fundamental neuromorphic behaviors, such as associative learning under classical conditioning, highlighting the potential of pure ionic dynamics for computing without rigid confinement. Despite representing a unique technological route for neuromorphic intelligence that is capable of information storage and logic construction, significant challenges remain. The electrochemical performance of these devices is highly sensitive to the precision of the channel geometry, which leads to difficulties in device-to-device consistency. Furthermore, the reliance on a fluid medium as an ion storage reservoir currently presents a substantial barrier to miniaturization and large-scale integration compared to their solid-state counterparts.

Soft Ionic Synapses: Bio-mimetic Signaling *via* Polyelectrolyte Networks

The final class of neuromorphic systems, probably the most faithful to biological principles, comprises of soft ionic synapses. These systems depart from the reliance on electronic currents and instead utilize the flow of ions to transmit and process information, mirroring the action potentials and neurotransmitter diffusion found in the nervous system.[58] The primary materials enabling this technology are hydrogels and ionogels, which consist of cross-linked polyelectrolyte chains swollen with liquids containing mobile ions. These materials provide a continuum that mimics the extracellular matrix, allowing the diffusion and migration of hydrated ions. These materials are soft and stretchable with tunable mechanical properties and excellent biocompatibility.[59–61] By synthesizing hydrogels with fixed positive or negative charges on the polymer backbone.

Early investigations demonstrated that joining a cation-selective hydrogel with an anion-selective hydrogel establishes an ionic junction that rectifies the ionic current and effectively functions as an ionic diode.[62] However, these materials have limitations beyond solvent evaporation. They primarily rely on Faradaic electrochemical processes to convert ionic currents into electrical signals at electrodes. This reliance not only restricts signal response speeds due to slow reaction kinetics, but also introduces issues such as electrode dissolution, gas evolution, and chemical compositional drift.

To address these challenges, oppositely charged nonaqueous ionoelastomers have been designed to construct robust heterostructures.[63] At the interface, this structure forms an IDL analogous to the depletion region in a p-n semiconductor, while excess fixed polyanionic and polycationic charges remain in the bulk. Crucially, by integrating these junctions with high-surface-area carbon nanotube electrodes, they achieved efficient rectification through a fully non-Faradaic capacitive process. This approach eliminates electrochemical degradation and enables rapid signal transduction, thereby laying the groundwork for the development of stable and stretchable ionic circuits (Fig. 5a).

Building on these fundamental rectifying junctions, subsequent research has focused on manipulating the ion transport to achieve neuromorphic intelligence. One major avenue is the translation of these transport mechanisms into emulation of temporal synaptic dynamics. Utilizing asymmetric hydrogels with multiple heterojunction interfaces, researchers have demonstrated that tunable synaptic dynamics can be realized through ionic diffusion and interfacial barriers, thereby eliminating the need for solid-state semiconductors (Fig. 5b).[64] Parallel to these functional demonstrations, fundamental advances have also been made in refining gating mechanisms. For instance, cascade-heterogeneous biphasic gels have been developed to modulate ionic free energy barriers and hydration states.[65] These devices achieve selective transport and gating of multiion signals, providing a physical basis for the complex information encoding required in such ionic systems. To expand sensory modalities, hydrogel fiber-based heterojunctions have been employed to construct artificial axons.[66] These fiber structures not only couple mechanical stimuli, but also induce ionic hysteresis at the fiber-gel interface, endowing the device with temporal signal integration capabilities. The transmission characteristics of such fibers can be tuned to exhibit signal attenuation or amplification, simulate the passive cable properties of neuronal dendrites, and successfully modulate sciatic nerve stimulation.

Although structurally regulated materials exhibit synaptic characteristics, a gap remains in mimicking the complex behaviors associated with memory and learning, particularly the trade-off between responsiveness and retention. To address this issue, recent advancements have systematically resolved the conflict between fast response and slow memory in soft ionic materials by exploiting ionic relaxation dynamics. By introducing cation-π interactions and adaptive IDL interfaces within the bilayer ionogels, a synergistic mechanism was achieved. A rapid ionic response enables immediate sensing and learning, whereas slow ionic relaxation supports long-term memory (Fig. 5c). Compared with one-time stimulation, repeated stimulation significantly improved memory strength, demonstrating the biomimetic memory characteristics of this device (Fig. 5d). This approach has realized diverse biomimetic functions, including sensitization, habituation, classical conditioning, and multimodal memory, thereby significantly elevating the intelligence level of ionic synapses.[67] The latest breakthroughs have pushed ionic synapses from

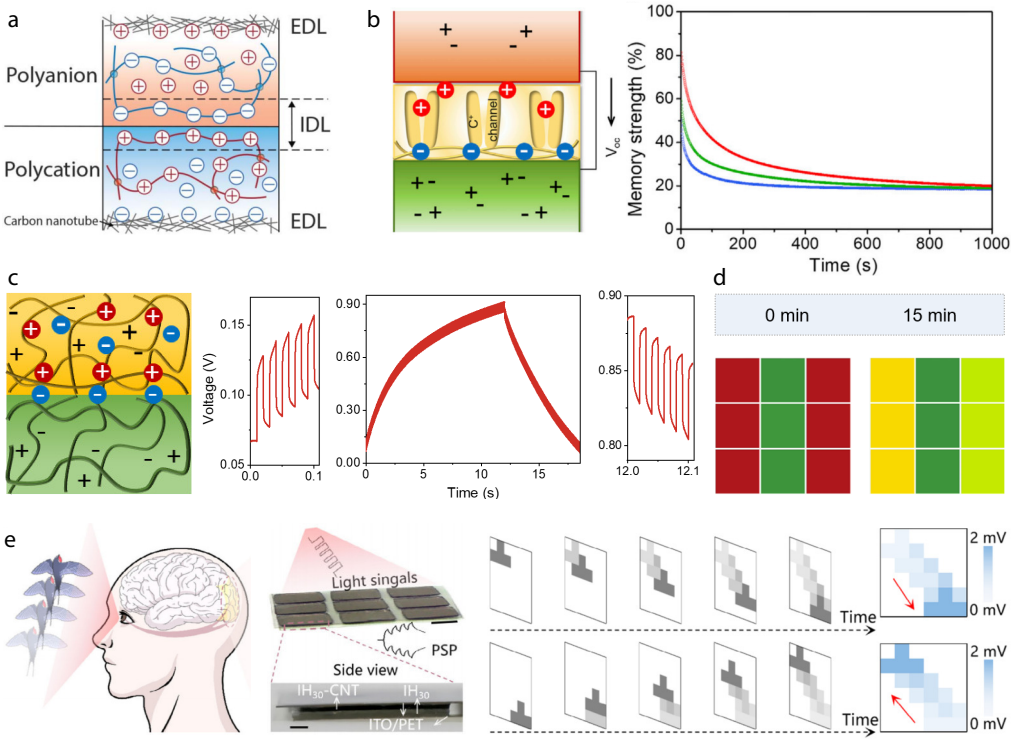

Fig. 5 Mechanisms and integration of soft ionic synapses. (a) The architecture of an ionoelastomer heterojunction forms a stable IDL at the interface analogous to a semiconductor depletion region (Reproduced with permission from Ref. [63]; Copyright (2020), The American Association for the Advancement of Science); (b) Synaptic dynamics and memory retention realized by the diffusion barriers in asymmetric hydrogels (Reproduced with permission from Ref. [64]; Copyright (2022), Elsevier Inc); (c) Schematic illustrating the regulation of ion transport in ionogels by self-adaptable IDL (left). When the bilayer ionogel is stimulated by 100 pulses, it can exhibit 100 different conduction states, thereby enabling the writing of information. Reverse stimulation can erase this information (right). (d) The comparison of memory strength after inputting five sets of 100-s stimulus training (left column) and a single 500-s stimulus training (right column). The transition from red to yellow to green represents different levels of memory strength. (c, d: reproduced with permission from Ref. [67]; Copyright (2025), The Author(s).) (e) A digital photograph of a bionic retinal array that processes light signals by mimicking the human visual system (left). The array enables the recognition of motion direction (right). (Reproduced with permission from Ref. [68]; Copyright (2025), Wiley-VCH GmbH).

single devices to system-level emulation. Through precise regulation of hydrogel structure and interfacial transport, both excitatory and inhibitory artificial synapses can be obtained within the same material system. Based on the synergy of these synapses, artificial retinal structures have been constructed to perform visual preprocessing functions, such as edge enhancement and motion detection, marking the transition of ionic artificial synapses towards functional integration and neural circuit-level processing (Fig. 5e).[68]

The energy efficiency and biocompatibility of soft ionic synapses are unparalleled; however, they face fundamental speed limitations. The ionic mobility in water is orders of magnitude lower than the electron mobility in semiconductors, restricting the operation frequencies of these devices to the sub-kilohertz range. However, this slowness is not necessarily a disadvantage for neuromorphic applications that interface with biological events, which typically occur on a millisecond to second timescale. The intrinsic time constants of ionic diffusion in hydrogels can be leveraged for temporal pattern recognition and reservoir computing, in which the material's fading memory of past inputs is a computational resource. The future of soft ionic synapses lies in harnessing these specific dynamics and prioritizing applications where low speed is an acceptable trade-off for high bio-integration, such as in implantable health monitors or biodegradable environmental sensors.

OUTLOOK

The field of ion-conducting polymer neuromorphic devices is rapidly transitioning from isolated-component demonstrations to integrated system-level validation. The trajectory of this research is increasingly defined by the co-design of materials and algorithms, in which the intrinsic physics of the polymer is tailored to specific computational models. A significant frontier lies in precise molecular engineering of polymer dynamics. Future synthetic efforts will likely focus on polymers with programmable relaxation spectra, where the side-chain architecture and crosslinking density are tuned to control the free volume and, consequently, the retention time of the synaptic weight. This would allow for the creation of hardware with a hierarchy of memory timescales, ranging from the immediate sensory buffer to consolidated long-term storage, all within a single-material platform.

Stability remains a significant challenge for the practical deployment of soft neuromorphic electronics. Organic materials, particularly when subjected to continuous electrochemical cycling in aqueous environments, are prone to degrada-

tion mechanisms, such as parasitic side reactions, hydrolysis, and delamination. The development of robust encapsulation strategies that can retain the necessary hydration for ionic function, while excluding reactive oxygen species, is critical. Furthermore, the synthesis of self-healing polymers that can repair the structural damage induced by volumetric expansion or mechanical wear offers a promising route to extend the operational lifetime of these devices. Research into hydrophobic-encapsulated mixed conductors and ionogels that operate with nonvolatile ionic liquids is also expanding the environmental range of these devices beyond strictly aqueous conditions.

Scaling and integration are the final major hurdles. Although individual polymer devices have shown exceptional performance, organizing them into high-density arrays requires overcoming issues related to patterning resolution and signal crosstalk. The concept of 3D integration, which utilizes the vertical dimension to stack polymer layers, is gaining traction as a method to increase the synaptic density. Additionally, hybrid architectures that combine the high-speed processing capabilities of silicon electronics with the efficient bio-interfaced sensing and learning capabilities of soft polymers represent a pragmatic path forward. In such systems, ion-conducting polymers would serve as the "front-end" interface, processing noisy, analog biological signals before handing off condensed information to a digital back-end. For instance, a recent study reported a 5×5 array of photoresponsive ionoelastomer synaptic devices that were designed to construct an artificial retina.[21] Upon illumination, the surface tension of the exposed units increased. However, owing to the relaxation effect, the stimulated units exhibited lower surface tension values than those stimulated more recently. By computing these dynamically modulated data in real time, the system forms images and successfully captures the motion trajectories of the light source.

CONCLUSIONS

In conclusion, ion-conducting soft polymers have established themselves as a transformative material class for neuromorphic hardware, uniquely blending the operational principles of solid-state electronics with the wet ionic nature of biology. By moving away from surface-limited field effects to volume-active ionics, these materials enable high-gain signal processing, linear synaptic plasticity, and biomimetic dynamics, which rigid inorganic materials cannot replicate. Whether through the viscoionic memory of organic electrochemical transistors, the precise linear switching of all-polymer ECRAMs, or the diffusive logic of soft ionic synapses, the underlying power of this technology lies in the molecular versatility of the polymer matrix. As researchers continue to master the synthesis and processing of these complex fluids and soft solids, we move closer to realizing intelligent systems that do not merely simulate the brain but also function with the same fundamental physics, thinking in ions, adapting through plasticity, and seamlessly integrating with the living world.

BIOGRAPHY

Zhou-Yue Lei received her B.S. degree from Sichuan University in 2014 and her Ph.D. degree from Fudan University (Supervisor: Professor Peiyi Wu) in 2019. She did postdoctoral research at Harvard University with Prof. David A. Weitz. In 2023, she returned to China and was selected as a recipient of the National High-Level Young Talent Program. From 2020 to 2025, she has been consecutively listed among the World's Top 2% Scientists by Stanford University. Her research focuses on ionotronic polymers, including conductive elastomers, ionic skins, and intelligent soft materials integrating sensing, memory, and computation.

Conflict of Interests

The authors declare no interest conflict.

ACKNOWLEDGMENTS

This work was financially supported by the National Natural Science Foundation of China (Nos. 52573019 and 22305033) and the Fundamental Research Funds for the Central Universities (No. 2232024A-05).

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Graphical Abstract

Soft Polymer Iontronics for Neuromorphic Intelligence

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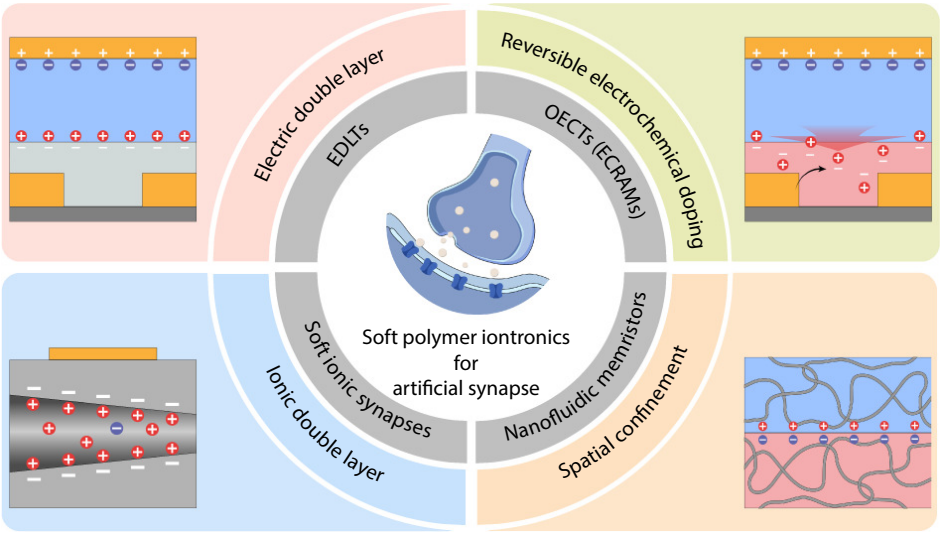
Donghua University; Southeast University

This review covers four ion-conductive soft polymer devices-ion-gated transistors (IGTs), electrochemical random-access memory cells (ECRAMs), nanofluidic memristors, and soft ion synapses-highlighting their mechanisms, challenges, and integration potential for biomimetic synaptic plasticity.



Chinese J. Polym. Sci. 2026, 44, 1681–1694 *https://doi.org/10.1007/s10118-026-3651-4*

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https://doi.org/10.1007/s10118-026-3651-4