

Green electronics based on biopolymer memristors toward sustainable neuromorphic devices

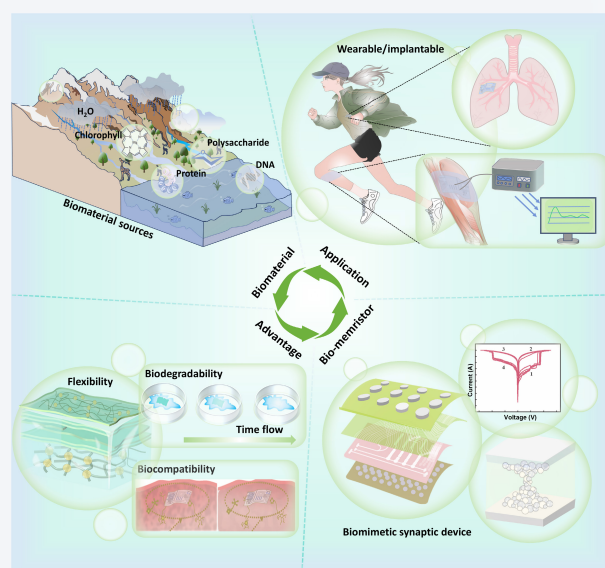
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ABSTRACT: The artificial intelligence-driven data deluge presents formidable challenges to conventional computing architectures, which are constrained by the von Neumann bottleneck and complementary metal-oxide-semiconductor (CMOS) scaling limits. Neuromorphic computing demonstrates breakthrough potential through its in-memory computing paradigm. Memristors, recognized as the most promising core devices in this field, excel at emulating synaptic plasticity while exhibiting nonlinear dynamic responses and conductance modulation capabilities. However, conventional inorganic memristors face critical limitations, including insufficient mechanical flexibility, biotoxicity, and non-degradability, which hinder their applications in wearable and implantable neuromorphic devices. In contrast, biomaterials and biological tissues emerge as viable platforms for overcoming these bottlenecks and realizing next-generation sustainable neuromorphic systems, owing to their exceptional biocompatibility, environmental benignity, and ultra-thin lightweight characteristics. This review focuses on cutting-edge developments in renewable biopolymer-based memristors (e.g., natural biomolecules like proteins and DNA, alongside bioengineered polymers such as poly(lactic acid) (PLA)). We elucidate how their molecular-level multifunctional groups and hierarchical structures drive high-performance memristive behaviors and bio-inspired synaptic functions. Special emphasis is placed on analyzing these devices' superior biocompatibility and biodegradability, with in-depth discussions on how such properties enable implantable neuromorphic applications. Finally, we critically examine persisting challenges including environmental sensitivity/resistance state drift, stochastic ion migration pathways, and scalable integration hurdles. Potential countermeasures and feasible development pathways are systematically explored.

KEYWORDS: neuromorphic device, biomemristor, biomaterial, biocompatibility, biodegradability



1 Introduction

The rapid advancement of artificial intelligence and next-generation information technologies has triggered explosive growth in data volume [1]. Although von Neumann computing sys4OS scaling and multi-core parallelization, the separation between storage and processing units leads to data transfer energy consumption, which exceeds 60% of total system energy, a limitation referred to as the

“memory wall” [2]. As transistor feature sizes approach the 3-nm node, quantum tunneling-induced leakage currents significantly reduce energy efficiency, limiting further scaling and hindering system performance in the big data era [3]. In contrast, the human brain achieves exceptional energy efficiency, consuming only ~ 20 W during complex tasks [4]. This efficiency stems from its unique neuroarchitecture, with densely interconnected neurons and plastic synapses enabling event-driven operation, parallel processing, and in-memory computing. Inspired by this, neuromorphic computing has emerged, mimicking the brain’s ability to perform communication, computation, and memory functions simultaneously (e.g., adjusting synaptic weights based on signal history) [5], overcoming the energy-efficiency limitations of conventional computing.

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To translate the brain-inspired neuromorphic computing paradigm into practice, it depends on developing devices that emulate the dynamic behaviors of neurons and synapses. Among existing candidates, memristors are ideal for neuromorphic systems due to their nonlinear resistive switching, non-volatile memory, and memory-computation fusion [6]. Memristors modulate resistance via electrical stimuli (voltage/pulse), with retained post-stimulation resistance states that produce differentiated electrical responses [7]. Their ability to dynamically reconfigure resistance while encoding historical current flow replicates synaptic learning and forgetting in biological neural networks.

Mainstream memristors currently rely on inorganic materials such as metal oxides, which typically require high driving voltages (> 1 V) and exhibit rigid characteristics [8]. Additionally, their inherent biological incompatibility and non-biodegradability limit their potential in sustainable neuromorphic devices. In bio-integrated applications, the rigid interface of inorganic memristors mismatches the mechanical properties of living tissues (with an elastic modulus difference spanning 3–6 orders of magnitude), leading to signal distortion and potential device failure [9, 10]. To address these limitations, researchers have turned to organic-inorganic hybrid memristors, which combine the excellent electrical properties of inorganic materials with the flexibility and processability of organic materials [11]. By incorporating organic layers or molecules, hybrid memristors maintain high conductivity while significantly improving flexibility and reducing power consumption (e.g., 78 nW [12]), making them more suitable for wearable devices and flexible electronics. However, despite reducing rigidity, hybrid systems still face challenges related to biocompatibility, stability, and environmental degradation.

In this context, bio-based memristors, as an emerging technology, hold immense potential due to their biocompatibility, biodegradability, and mechanical flexibility. The unique advantages of biomaterials, such as polysaccharides and proteins, stem from their multi-level ordered structures and rich functional groups, which precisely regulate charge transport and mimic biological synaptic functions [13]. While organic-inorganic hybrid materials enhance device durability by combining the high performance of inorganic materials with the flexibility of organic ones, bio-based materials naturally offer biodegradability without the need for additional surface modifications or chemical crosslinking, making them more environmentally sustainable [14]. Moreover, biomaterials can be further enhanced through genetic engineering or composite with conductive materials (e.g., carbon-based materials), improving conductivity while maintaining their degradable characteristics through natural scaffolds or controlled crosslinking networks [15]. This strategy drives the application of bio-based memristors in neuromorphic computing and implantable devices [16].

Emerging as transformative functional devices for green electronics, biomaterial-based memristors have recently made significant strides in materials design and brain-inspired functionalities. This review systematically examines the technological advancements and application potential of these devices from four key perspectives (Fig. 1). Section 1 provides an overview of the latest research on bio-derived memristors for sustainable neuromorphic devices, with a focus on biological material sources, structural classifications, molecular characteristics, and their unique advantages in modulating memristive behaviors. Section 2 explores the operational principles of bio-memristive

systems, emphasizing the role of biological components in various switching mechanisms. Section 3 highlights the value of biological memristors in neuromorphic applications, evaluating their biocompatibility and environmental sustainability. Finally, Section 4 critically addresses the challenges hindering device development, offering interdisciplinary solutions and discussing their potential to drive breakthroughs in next-generation sustainable neuromorphic computing architectures.

2 Biomaterials types

Bio-derived materials from plants, animals, and microorganisms exhibit unique physicochemical properties shaped by evolutionarily refined molecular architectures. These biomass-based intelligent materials address the biotoxicity challenges of conventional electronics through inherent biodegradability and biocompatibility, while also demonstrating exceptional mechanical compliance. This unlocks transformative potential for neuromorphic devices in sustainable computing and wearable sensing technologies. Importantly, biological components in memristive devices rarely operate via a single mechanism; instead, they rely on synergistic cross-material interactions. This involves constructing heterogeneous composite architectures that integrate inorganic semiconductors, conductive polymers, and biomaterials to form cooperative molecular/mesoscale regulation networks, optimizing device performance and stability. In this section, we systematically explore the functional characteristics of bio-based materials, reviewing their natural origins, structural foundations, and mechanistic roles in memristive neuromorphic device applications.

2.1 Natural biopolymer materials

2.1.1 Polysaccharides

Monosaccharides, serving as core metabolites in living systems, represent the minimal structural unit of carbohydrates. Polysaccharides, which are hierarchically assembled from ≥ 10 monosaccharide subunits via $\beta(1\rightarrow4)$ - or $\alpha(1\rightarrow6)$ -glycosidic linkages, function as nature's energy reservoirs (e.g., glycogen, amylopectin) [17]. These polymers host abundant functional groups ($-\text{OH}$, $-\text{NH}_2$, $-\text{COOH}$) capable of electric-field-induced ionization/polarization [18], facilitating ion/charge transport while providing reactive sites for covalent integration with nanomaterials (e.g., graphene, carbon nanotubes) to form functional composites [19]. Their intrinsic biocompatibility and renewability further establish polysaccharides as ideal candidates for flexible memristors. Based on functional group chemistries, we classify them into cationic, neutral, and anionic polysaccharides [20].

Positively charged polysaccharides are polymer carbohydrates with a net positive charge, mainly due to the amino groups ($-\text{NH}_2$) that bind to protons (H^+) in acidic environments, forming ($-\text{NH}_3^+$). Chitosan, a rare natural positively charged polysaccharide, can be cheaply produced commercially by extracting it from chitin, which is abundant in the exoskeletons of crustaceans (Fig. 2(a)). Chitosan consists of long-chain structures made from D-glucosamine and N-acetyl-D-glucosamine [21]. Its versatile structure allows it to be formed into films, nanofibers, and nanoparticles. While pure chitosan has limited properties, it can be modified through physical or chemical methods to enhance its functionality [22].

The excellent memristive performance of chitosan results from the synergistic interplay of its key functional groups, including

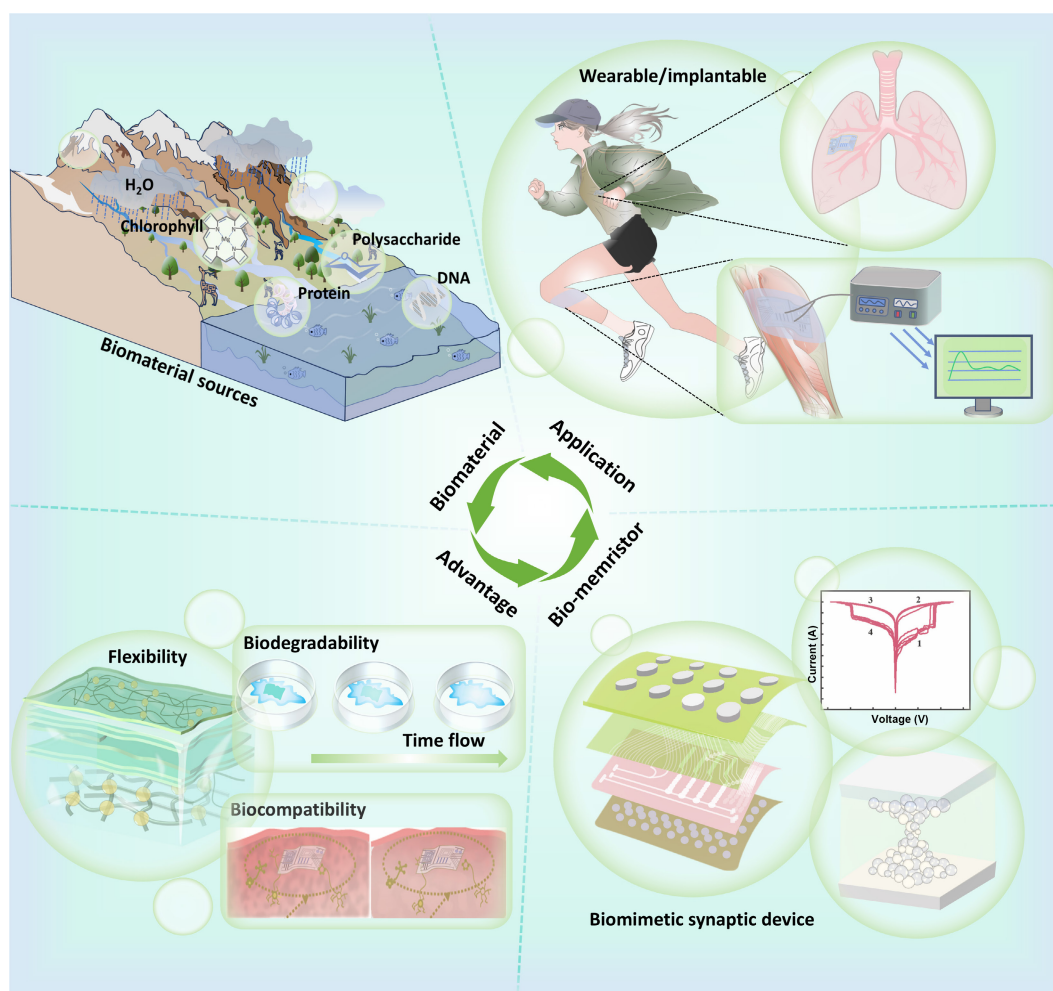


Figure 1 Schematic illustrating the fundamental characteristics and application potential of biomaterial-based memristors in sustainable neuromorphic devices, with annotated biological material sources and their intrinsic properties.

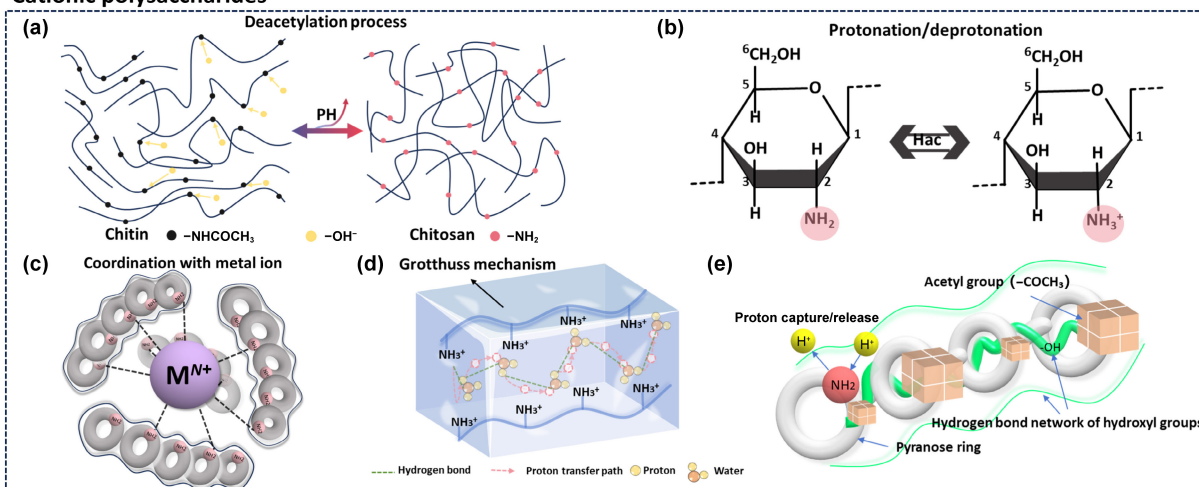
amino ($-\text{NH}_2$), hydroxyl ($-\text{OH}$), and acetyl ($-\text{COCH}_3$), which together form a “trinity” resistive switching system in sandwich-structured memristors. The amino and hydroxyl groups form hydrogen-bond networks that support proton conduction [23]. When a positive bias is applied, the amino groups undergo protonation ($-\text{NH}_2 \rightarrow \text{NH}_3^+$), trapping protons and releasing H^+ ions (Fig. 2(b)). These H^+ ions then undergo Grotthuss-type transport through the hydrogen-bond networks (Fig. 2(d)) [24]. Simultaneously, the amino groups coordinate with metal ions via their lone electron pairs, guiding conductive filament growth (Fig. 2(c)). The hydroxyl groups also drive the electrolysis of water ($\text{H}_2\text{O} \rightarrow \text{H}^+ + \text{OH}^-$), supplementing proton conduction while facilitating metal ion generation at the anode. These processes create dual conductive pathways—metallic filaments and proton transport—that lead to an abrupt reduction in resistance [25]. When a negative bias is applied, ion migration reverses, rupturing the conductive filaments. During this phase, the hydroxyl groups reorganize the hydrogen-bond networks to regulate ion diffusion, while protonated amino groups deprotonate ($-\text{NH}_3^+ \rightarrow \text{NH}_2$), reducing the molecular charge density and triggering the contraction of polymer chains, thus narrowing the proton channels. At the same time, acetyl groups in chitosan serve as structural stabilizers and ionic migration balancers, enhancing both flexibility and interfacial compatibility [26]. These components create

multilevel energy barriers that increase resistance, enabling stable and controllable resistive switching, making chitosan an ideal material for biocompatible, flexible memristors (Fig. 2(e)).

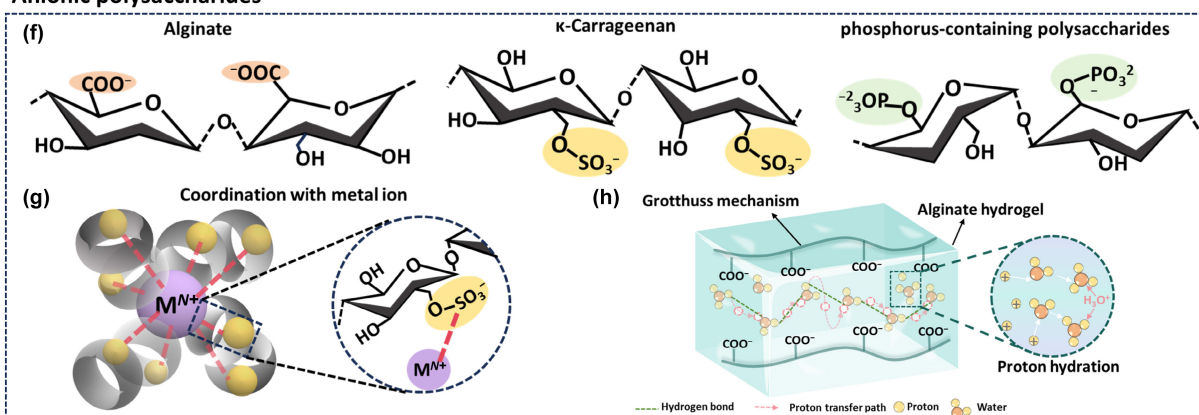
Pure chitosan-based memristive devices suffer from random conductive filament growth and brittleness, limiting performance uniformity and flexibility. To overcome these issues, Hosseini et al. (2021) embedded gold nanoparticles (Au NPs) within chitosan, forming a solid polymer electrolyte (SPE) layer in an Ag/Au-chitosan/Au crossbar memristor [27]. This modification enhances conductivity while maintaining biocompatibility, achieving a switching ratio of 10^3 , and simultaneously improving optical transparency and mechanical flexibility. Remarkably, the device maintains reliable bipolar resistive switching even after 1000 bending cycles at a 5 mm curvature radius, showcasing nanoparticle-reinforced chitosan as a promising platform for wearable flexible electronics.

Although metal-chitosan memristors have demonstrated excellent mechanical flexibility, their environmental stability and adaptability under extreme conditions remain limited. This is primarily due to the increased thermal vibration of chitosan molecular chains under electrical and thermal stress, which causes conformational drift and deformation of ion transport channels, leading to resistance state drift. Studies have shown that combining chitosan with carbon-based materials, such as graphene or carbon

Cationic polysaccharides



Anionic polysaccharides



Neutral polysaccharides

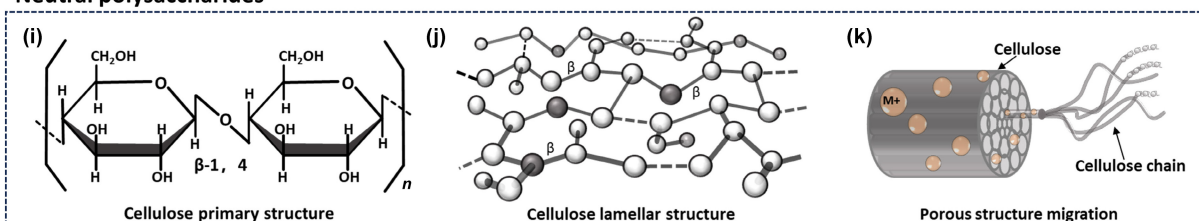


Figure 2 Structural and physicochemical properties of diverse polysaccharides. (a) Schematic illustration of the chitin deacetylation process converting to chitosan, with functional group changes represented by black and red spheres. (b) Schematic diagram of chitosan protonation/deprotonation process and chemical structure of chitosan. (c) Schematic illustration of coordination between amino groups in chitosan molecules and metal ions, with gray rings representing D-glucosamine residue units in chitosan. (d) Schematic diagram of the Grotthuss mechanism for proton transport through hydrated hydrogen-bonded networks. (e) Schematic illustration of synergistic interactions among amino, hydroxyl, and acetyl groups in chitosan molecules, where red spheres represent amino groups participating in proton capture/release, brown cubes denote the structural support role of acetyl groups, and green strands depict hydrogen-bond networks mediated by hydroxyl groups. (f) Chemical structures of three anionic polysaccharides. (g) Schematic illustration of the coordination between anionic polysaccharides and metal ions. (h) Proton transport via the Grotthuss mechanism within alginate hydrogels, with the magnified region highlighting proton hydration phenomena. (i) Chemical structure of the neutral polysaccharide cellulose. (j) Schematic illustration of the layered structure of cellulose. (k) Schematic of ion migration through the porous structure of cellulose, with the right panel highlighting structural features of cellulose chains.

nanotubes, can significantly enhance the device's flexibility and antioxidant performance. Kim et al. (2020) utilized a CS-graphene oxide (GO) composite to fabricate an Ag/CS-GO/FTO device structure. This improved operational stability, but suffered from inefficient ion transport [28]. Addressing this limitation, Zhao's group designed (2022) an interpenetrating CS-reduced GO (rGO) network electrolyte, achieving a flexible memristor with high reproducibility, superior ionic conductivity, and multi-level storage [29]. Compared to CS-GO, the CS-rGO electrolyte offers enhanced

conductivity, stability, flexibility, and memory diversity. The high oxygen content in CS-GO hinders ion migration and its disordered structure yields poor conductivity. In contrast, reducing GO to rGO increases sp^2 carbon domains, dramatically boosting ion mobility to $2.65 \times 10^{-5} \text{ S/cm}$ (7.4 times higher than CS-GO). Crucially, the 3D interconnecting network of the CS-rGO electrolyte enhances molecular ordering, establishing stable ion channels and suppressing the random formation of metallic conductive filaments. This device maintains stability without degradation for 24 h at

37 °C, with a switching ratio remaining stable around 10^6 , significantly improving both stability and reliability. By adjusting the compliance current (I_c) between 10 μ A and 1 mA, the device exhibits six distinct resistance states. Notably, each resistance state is maintained for over 10^4 s, demonstrating its multilevel storage capability.

Anionic polysaccharides are charged polyelectrolytes with three characteristic acidic groups ($-\text{COOH}$, $-\text{OSO}_3^-$, $-\text{OPO}_3^{2-}$), which dissociate to release mobile protons (H^+) while maintaining anionic backbones (Fig. 2(f)) [30]. These groups dictate specific bioresource associations: carboxylates dominate in alginate (marine algae) and pectin (plants), while sulfates prevail in carrageenan (red algae). The abundant carboxylic acid and sulfate ester groups are highly hydrophilic, and the negative charges cause the molecular chains to extend in solution, enhancing their contact with water and enabling stable hydrogen-bond network formation [31]. Released protons (H^+) move through this network, either hopping between sites or combining with water to form hydronium ions (H_3O^+), which migrate via random diffusion (vehicle mechanism) (Fig. 2(h)) [24]. These mechanisms contribute to the excellent proton conductivity of anionic polysaccharides. Additionally, the high density of negative charges facilitates strong electrostatic interactions and the formation of stable coordination bonds with protons and metal ions (Fig. 2(g)), further promoting ion movement. As a result, anionic polysaccharides effectively serve as the electrolyte or active layer in memristors, directly influencing resistance switching behavior.

Negatively charged polysaccharides (e.g., κ -carrageenan, sodium alginate (SA), pectin) form crosslinked 3D networks that provide structural integrity and enable ion/molecular regulation via intrinsic porosity and charge effects [32]. Wang et al. (2022) engineered an SPB hydrogel through PVA-SA-STB dynamic crosslinking, where borate ester bonds (PVA-STB) and hydrogen-bonded SA-PVA networks enhanced mechanical robustness and ionic conductivity. Notably, severed regions self-healed within 30 s and fully recovered in 1 h [33]. Chen's team (2023) extended this design to memristors, creating SA/PVA/STB-based devices with a $> 10^3$ ON/OFF ratio and 2000 s operational stability. After damage, switching current recovered to 12 ms (~ 15 ms original) within 1 h while maintaining memristive function [34].

Pectin and carrageenan, compared to SA, offer distinct advantages due to their unique structural motifs, enhancing ionic conductivity. Pectin enables dynamic ion modulation through pH-responsive methylesterification of carboxyl groups ($-\text{COOH}$), while carrageenan's sulfated esters ($-\text{OSO}_3^-$) provide 50% higher charge density than carboxyl-based polymers [35, 36]. Both are fully biodegradable under ambient conditions, making them ideal for eco-friendly electronics. Li et al. (2022) developed a synaptic transistor using Ca^{2+} -crosslinked pectin, where the high-density carboxyl network facilitated proton conduction, enabling ultralow-voltage operation (20 mV) and tunable synaptic plasticity. The device achieved complete hydrolysis in 100 s, with *in vivo* studies confirming biocompatibility (100% zebrafish survival) [37]. Similarly, Chang et al. (2023) engineered a carrageenan-based memristor by embedding K^+ ions to stabilize its double-helical structure, minimizing interfacial defects and localized electric fields, achieving a record switching ratio $> 10^7$ and 52-day environmental degradation [38].

Unlike acidic or alkaline polysaccharides, neutral polysaccharides feature non-dissociable hydroxyl groups ($-\text{OH}$) that prevent charge

generation in solution, resulting in low conductivity (Fig. 2(i)). However, their structural diversity and low hygroscopicity allow water adsorption in humid environments, forming hydrogen-bond networks that facilitate ion migration through porous structures or proton pathways. Cellulose, with its ordered nanofibrillar network, provides efficient ion-conducting channels (Fig. 2(k)). The chemical inertness of hydroxyl groups ensures stability by preventing reactions with metal electrodes [39]. Mechanically, cellulose exhibits high tensile strength (~ 2 GPa) and Young's modulus (~ 200 GPa) due to intermolecular H-bonding (Fig. 2(j)), while starch retains compressive strength despite lower crystallinity [40, 41]. Combined with biodegradability, these properties position neutral polysaccharides as strong candidates for flexible memory and neuromorphic computing technologies.

In 2022, Hussain's team developed a cellulose nanocrystal (CNC)/AgNP hybrid memristor, combining high ON/OFF ratio digital storage with biological synaptic plasticity (LTP/LTD/PPF) [42]. The CNC's layered structure offers two advantages: (i) It serves as a template for silver ion filament growth at low voltage, and (ii) the hydroxyl passivation layer prevents electrode corrosion. In 2023, Professor Sun's team developed a corn starch-PVDF device integrating storage, sensing, synaptic, and logic functions [43]. This porous starch-based network enables efficient silver ion migration (operating voltage < 1 V) and exhibits excellent flexibility, with over 200 bending cycles. The system achieves a switching ratio over 10^2 and remains stable after 100 cycles, meeting the requirements for sustainable electronics.

Overall, polysaccharide-based memristors offer significant advantages in biodegradability and energy efficiency, making them promising candidates for sustainable electronic devices and biomedical applications. While these devices exceed traditional inorganic memristors in terms of biocompatibility and energy consumption control, their switching speed and stability under extreme conditions remain limitations, restricting their expansion in high-performance applications. The incorporation of carbon-based materials (such as graphene, carbon nanotubes, etc.) for composite modification effectively enhances charge transport efficiency and stability while preserving the intrinsic advantages of the material, providing a viable solution to these challenges.

2.1.2 Proteins

Compared to other natural biomaterials, proteins have more intricate architectures due to their unique amino acid compositions and hierarchical four-level structures (primary to quaternary). The hierarchy begins with linear amino acid sequences (primary structure, Fig. 3(a)), progresses through hydrogen-bond-stabilized secondary structures, and achieves stable tertiary conformations (Fig. 3(b)) via spatial folding coordinated by hydrogen bonds and disulfide bridges ($-\text{S}-\text{S}-$) [44]. This complexity enables programmable functionality, where precise modulation of each level through genetic engineering or chemical modification tailors material properties, highlighting proteins' potential for high-performance memristor development [45]. Functionally, protein secondary structures primarily adopt β -sheet or α -helix configurations, which often coexist in varying proportions. Tertiary structures diverge into globular or fibrous forms for functional specialization. The pairing of secondary structures (β -sheet/ α -helix) with morphological types (globular/fibrous) expands functional diversity, yielding four distinct categories: β -sheet + fibrous, β -sheet + globular, α -helix + fibrous, and α -helix + globular [46].

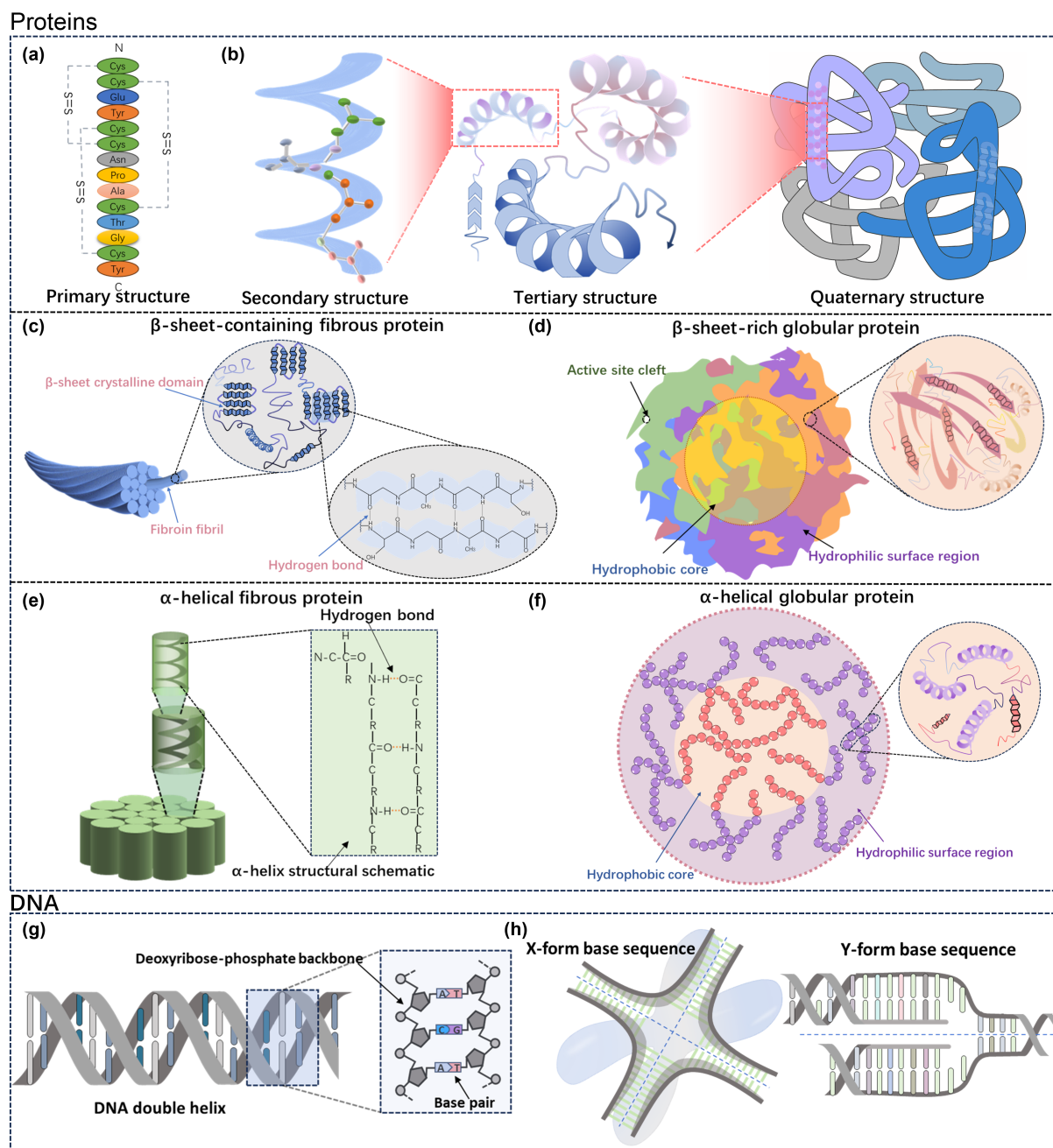


Figure 3 Structures and properties of different types of proteins and structural characteristics of DNA. (a) Schematic of short peptide sequence (with S-S bonds): composed of Cys, Glu, Tyr, Asn, etc. (specific arrangement) with distinct N-terminus (amino) & C-terminus (carboxyl). (b) Protein hierarchical structures. (c) Schematic illustration of silk fibroin protein containing fibrillar silk precursors and β -sheet crystalline domains. (d) Schematic of β -sheet-rich globular protein: undulating surface (with functional sites: concave/convex); central yellow region = hydrophobic core, surrounding = hydrophilic surface. (e) Structural illustration of α -helical fibrous protein: right magnified region highlights characteristic H-bonds (between C=O & N-H groups, 3-4 residues apart along α -helix backbone). (f) Schematic of 3D structure of α -helix-rich globular protein: central red region & purple outer layer = hydrophobic core & hydrophilic surface (respectively). (g) Schematic diagram of the DNA double helix structure. (h) Schematic diagram of X/Y-shaped double-stranded units formed by designing specific base sequences.

In β -sheet + fibrous protein architectures, the fibrous organization serves as a structural scaffold, with tightly packed fibrillar networks enhancing material extensibility and toughness. The interwoven β -structured polypeptide chains form hydrogen-bonded layers, creating a rigid framework that provides mechanical strength (Fig. 3(c)) [47], while inter-sheet gaps facilitate ion transport. Among these proteins, silk fibroin stands out due to its unique molecular design: alternating glycine (Gly) and serine (Ser) residues drive dense β -sheet stacking, while Ser's hydroxyl groups

(-OH) enhance hydration through hydrogen bonding, imparting plasticity and flexibility. Silk fibroin predominantly adopts antiparallel β -sheet conformations (~50%–60% content), forming a highly ordered nanofibrillar network with exceptional mechanical properties and biocompatibility [48, 49].

In 2021, Zhang's team reviewed silk fibroin (SF) in memristor applications, highlighting its mechanical flexibility and biodegradability, while noting challenges such as high power consumption and instability in conventional SF-based devices [50].

To address these, they integrated silver nanoparticles (AgNPs) into SF, followed by ethanol treatment, which induced stable microcrystalline structures. These crystalline domains provided shorter, less resistant carrier transport pathways, reducing power consumption by over 50%. Building on this, Li's team (2024) developed an SF-graphene oxide (GO) composite memristor, incorporating GO into the SF matrix [51]. GO introduced tunable oxygen vacancies at the interface, which confined vacancy migration to interlamellar pathways, promoting uniform conductive filament nucleation. This not only reduced power consumption but also increased the switching ratio to 10^3 , with negligible degradation over 100 cycles. GO doping further stabilized filament formation, reducing protein hydrolysis risks and extending environmental stability to 2 weeks (compared to < 1 week for pure SF devices). Furthermore, The optoelectronic modulation capability also enabled dual-mode electrical/optical synaptic operation, mitigating thermal degradation from Joule heating.

β -sheet-containing globular proteins share structural features with fibrous proteins but achieve tighter molecular packing through highly ordered folding into compact spherical conformations. Their surfaces exhibit irregular morphologies with multiple functional sites (Fig. 3(d)) [52], and their hydrophobic-core/hydrophilic-shell architecture ensures aqueous stability. Under electric field stimulation, these proteins undergo reversible β -sheet-to-alternative-structure transitions [53], driving conductive pathway switching. Pathway A, stabilized by conformation A, dissipates as Pathway B emerges through conformation B [54]. This dynamic modulation significantly enhances ion transport controllability. Proteins like trypsin and ferritin demonstrate functional variations in memristive devices due to their distinct physicochemical properties.

The abundant polar residues in trypsin (such as serine and aspartic acid) facilitate proton conduction and induce conformational changes responsive to electric fields, enabling precise control over ion transport. Based on this mechanism, Desai developed a hydrogel memristor in 2021, achieving frequency-dependent hysteresis and synaptic pulse recognition ($\tau \sim \text{ms}$) through the trypsin-FTO interface's bilayer effect [55]. However, trypsin-based memristors face degradation issues, including hydrogel dehydration and enzyme active site oxidation damage under high voltage (> 4 V). To address these challenges, limiting the operating voltage to $\leq 3 \text{ V}$ effectively prevents the generation of $-\text{OH}$ radicals, while using Tris-Cl buffer (pH 8.5) competitively stabilizes the enzyme's conformation, maintaining > 95% activity after 50 cycles and extending the hydrogel's structural stability for up to 48 h.

In contrast, ferritin stores approximately 4500 $\text{Fe}^{2+}/\text{Fe}^{3+}$ ions in its hollow nanostructure, with external electric fields triggering controlled Fe^{3+} release via a redox gating mechanism mediated by ferritin. In 2016, Zhang developed a ferritin-based memristor with dual-mode (volatile/non-volatile) operation by modulating compliance current (I_{cc}) to stabilize filament formation [56]. At low I_{cc} (< 5 μA), Fe^{2+} -formed filaments become fragile, leading to resistance degradation and threshold switching. Increasing I_{cc} to 100 μA stabilizes the iron filaments, enabling non-volatile memory characteristics and achieving a high switching ratio (> 10^5) over 70 cycles.

In α -helix+fibrous proteins, the α -helical structure arises from hydrogen bond formation between the C=O group of each amino acid and the N-H group of the fourth subsequent residue along the axis, creating a periodic helical arrangement (Fig. 2(e)). This highly

regular linear conformation provides more efficient proton migration pathways compared to the sheet-like structure of β -sheets [57, 58]. Furthermore, fibrous proteins such as ferritin and collagen not only exhibit exceptional mechanical robustness, but their well-ordered linear architectures enable the formation of conductive channels, which are crucial for achieving low power consumption and high on/off ratios in memristive devices.

Collagen, a structural protein abundant in animal connective tissues, exhibits remarkable biocompatibility and biodegradability. In 2024, Raeis-Hosseini et al. developed a dissolvable bio-memristor by extracting collagen from fish scales and integrating it with Mg electrodes and ITO/PET substrates [59]. In this device, collagen facilitates efficient Mg^{2+} migration due to (i) abundant polar groups (e.g., hydroxyl) that adsorb ions in aqueous environments and (ii) an intrinsic helical structure providing ion migration channels. Additionally, the collagen thin film combined with flexible PET substrates enables high mechanical durability, allowing the device to retain stable performance (< 15% current fluctuation) after 1000 bending cycles. Notably, the presence of hydrophilic groups (e.g., hydroxyl) ensures complete degradation in aqueous solutions within 2 min. In 2025, Zheng et al. further advanced this concept by aligning collagen into periodically staggered fibrils and infiltrating the interstitial spaces with CdCO_3 , which grows along the fibril's long axis to form highly ordered ion transport channels [60]. This structural modification not only reduced ion migration by 60%, but also suppressed conformational drift under electric field and thermal stress, enabling the device to achieve an ultra-low switching voltage (0.65 V) and an exceptionally high switching ratio ($\sim 10^6$) at 37 °C, with no significant decay over 24 h.

The final protein category integrates α -helical and globular structures (Fig. 2(f)), exemplified by whey protein and ovalbumin. Whey protein's sulfhydryl ($-\text{SH}$) groups enable unique metal-ion coordination, undergoing field-induced redox reactions that reduce ions to atomic metals to initiate conductive networks [61]. Synergy with other polar functionalities enhances ion transport via multivalent interactions, substantially boosting conductivity. Conversely, ovalbumin contains abundant disulfide bonds but only four $-\text{SH}$ groups, yielding devices with high operating voltage and exceptional stability [62].

In 2022, Wang's team developed a flexible, physically transient memristive device by spin-coating a thin film of silver ion-doped egg white protein (ovalbumin) on a flexible substrate [63]. The ovalbumin's polar groups served as effective silver ion carriers through adsorption, and the incorporated Ag^+ ions provided an exceptional switching ratio (2×10^5) via constrained electron tunneling. The dried film exhibited remarkable flexibility, maintaining functionality even with a bending radius as small as 12 mm. Additionally, the protein's water solubility enabled complete dissolution within 30 min. In 2023, Xiao et al. developed an ovalbumin-based memristor with complementary switching characteristics using non-inert Al electrodes [64]. Under applied bias, oxygen anions (O^{2-}) released from ovalbumin reacted with Al electrodes to form a Schottky barrier, which could be modulated by varying voltages to regulate resistance switching. The flexible polypeptide chains in ovalbumin also conferred exceptional endurance, with stable performance over 140 continuous cycles.

Overall, the abundant polar residues, reconfigurable conformations, and inherent ion-binding sites in protein materials enable stable operation under low voltage conditions, making them particularly suitable for applications that require high

biocompatibility and flexibility, such as biointerfaces and wearable devices. In contrast, traditional inorganic memristors typically operate at higher voltages and have poor biocompatibility and environmental sustainability. However, protein-based memristors also face certain limitations, especially regarding degradation under high temperature and prolonged operation. At higher electric fields or elevated temperatures, protein materials may experience conformational relaxation, dehydration, or oxidation, leading to performance degradation. Compared to polysaccharide-based memristors, although protein-based memristors exhibit superior electrical performance (e.g., several studies have demonstrated protein-based memristors with switching ratios exceeding 10^5 and lower operating voltages [60]), they still face challenges in stability and long-term durability. Polysaccharide materials, with their enhanced degradation and stability, are more suitable for short-term applications, while protein-based materials are better suited for applications that require high electrical performance and biocompatibility.

2.1.3 DNAs

DNA, nature's primary genetic material, encodes heredity via precisely ordered nucleobases. Its iconic double helix (Fig. 3(g)) features deoxyribose-phosphate backbones: the negatively charged phosphates electrostatically attract mobile metal cations, which then migrate directionally along the strands under electrical stimulation [65]. Simultaneously, nucleobase heteroatoms (O/N) coordinate metal ions, steering their movement along the π -stacking axis via field-induced polarization [66]. This dual mechanism (backbone cation drift + base ion coordination) creates efficient conductive pathways. Critically, DNA's 1–2 nm nanoconfinement shortens charge transport distances for low-power operation [67], while its atomic scale enables memristor densities unattainable with silicon photolithography—positioning it as a transformative platform for ultrahigh-density memory (Fig. 3(h)).

Conventional double-stranded DNA films fabricated via solution processing typically exhibit disordered ion transport pathways due to random molecular stacking and nanoscale channel width constraints. Such structural defects inevitably lead to elevated switching ratios and increased energy consumption, making them unsuitable for neuromorphic computing applications demanding ultra-low power operation. In 2023, Wang's team developed a porphyrin-DNA hybrid memristor achieving exceptional performance parameters (39.56 nW power consumption and $> 10^5$ stable cycles) [68]. Nevertheless, this device relied on DNA-material hybridization, necessitating the development of purely DNA-based functional layers. By 2025, the team engineered a novel memristor employing three-dimensional DNA brick crystals (DNAbc) as the active component [69]. Unlike conventional dsDNA, the 15-nm-thick DNAbc features a compact, ordered three-dimensional architecture that significantly enhances ion transport efficiency. This pure DNA-based device achieved a switching ratio of 10^4 , an ultralow programming power of 2.17 μ W, and robust cycling stability without requiring composite materials.

Overall, DNA materials, with their unique double-helix structure and negatively charged phosphate groups, form effective conductive channels with metal ions, enabling ultra-low power consumption (e.g., as low as 2.17 μ W [69]), making them highly suitable for applications such as ultra-high-density 3D integrated memory chips. Leveraging their inherent self-assembly properties at the molecular level, DNA can form highly ordered nanostructures, significantly surpassing the storage density of traditional inorganic

memristors. Moreover, compared to protein and polysaccharide-based memristors, DNA materials typically exhibit superior ion conductivity and electrical performance, providing the device with higher storage capacity and lower operating voltage. However, in practical applications, DNA materials are highly sensitive to environmental factors such as temperature and humidity, which can lead to resistance state drift and performance degradation. Therefore, techniques such as chemical crosslinking or material hybridization are necessary to enhance device stability. This also reflects that DNA-based memristors are less stable and adaptable to environmental changes compared to the other materials.

2.1.4 Other biomaterials

Recent investigations have identified unconventional biological substances exhibiting remarkable promise for resistive switching applications. Among lesser-explored candidates, naturally derived compounds including photosynthetic pigments (e.g., chlorophyll) [70], plant polyphenols (lignin) [71], apicary products (propolis) [72], floral biomass [73], *Azadirachta indica* extracts [74], and hydrophyte epicuticular waxes demonstrate particularly interesting molecular configurations that facilitate memristive behavior [75]. These organic systems typically feature redox-active moieties such as phenolic groups and formyl functionalities, which enable reversible charge transport phenomena under applied electric fields, consequently modulating conductive states [76]. Their appeal stems not only from structural advantages but also from compelling practical benefits: abundant natural sourcing combined with straightforward fabrication protocols. Furthermore, their intrinsic physiological compatibility and environmental degradability present transformative opportunities for biointegrated neuromorphic devices.

Chlorophyll, a photoactive metallo-organic hybrid molecule, features a magnesium-centered tetrapyrrole macrocycle coordinated with an aliphatic hydrocarbon tail [77, 78]. Its conjugated tetrapyrrole ring enables delocalized electron clouds that enhance interfacial charge transport, while the peripheral hydrocarbon tail modulates van der Waals interactions, promoting amorphous thin-film formation and suppressing leakage currents [79]. Beyond its structural advantages, chlorophyll exhibits light-harvesting capabilities within the visible spectrum. Photoexcitation drives transitions from ground to excited states, generating localized polaronic electron pairs that spatially redistribute interfacial barriers and dynamically tune localized electric fields [80]. These unique optoelectronic properties position chlorophyll as a promising candidate for synaptic transistors and bioinspired neuromorphic devices.

In 2020, Yang's team, inspired by plant photosynthesis, developed a multifunctional field-effect transistor based on natural chlorophyll and organic semiconductors [81]. This transistor combines the functions of a photodetector (PD) and a light-stimulated synaptic transistor (LSST), with a switching ratio as high as 2.7×10^6 under light illumination of 0.271 mW/cm² in photodetection mode. Interestingly, the device can dynamically switch between these two modes by adjusting the gate voltage. Under the cooperative control of the light grid, hole accumulation in the dielectric layer localizes photogenerated carriers, forming a stable space charge region. This charge layer effectively screens the gate electric field, causing a positive shift in the channel threshold voltage. Moreover, chlorophyll molecules exhibit excellent light-capturing properties, inducing a significant photoresponse even under extremely low light conditions, reducing single pulse energy

consumption to 0.25 fJ in synaptic mode. The device shows potential applications in light imaging (5×5 sensor array), neuromorphic computing (82% accuracy in MNIST recognition), and image contrast enhancement. In 2021, Ou's team developed a low-energy, biodegradable light-controlled synaptic transistor using chlorophyll-a and single-walled carbon nanotubes (SWCNTs) [82]. Chlorophyll-a, with its ultra-low exciton binding energy, works synergistically with the high mobility properties of SWCNTs, enabling the device to achieve an ultra-low energy consumption of 17.5 fJ/pulse at ultra-low voltages (100 μ V). Its unique energy level arrangement and charge trapping mechanism successfully mimic the plasticity characteristics of biological synapses.

2.2 Synthetic biopolymer materials

Poly(lactic acid) (PLA), derived from renewable biomass like corn, requires synthetic polymerization, unlike enzymatically produced biopolymers (e.g., proteins, chitosan). As a structurally designable biomaterial, PLA permits precise control of chain length, stereochemistry, and functional groups, enabling optimization of ionic/charge transport for memristors [83]. Specifically, adjusting polymerization yields short-chain PLA with high amorphous content (enhancing ion mobility) or long-chain entangled structures (improving mechanical stability). The L-/D-lactide ratio critically governs crystallization: L-rich configurations slow degradation via ordered packing, while D-incorporation disrupts alignment to accelerate hydrolysis [84, 85]. This exceptional balance between tunable charge transport and programmed degradation enables PLA to serve as a versatile platform for next-generation biodegradable electronics and adaptive memory devices.

In 2024, Ren's team developed a synaptic device based on fibrous PLA and MXene, combined with ME-ICE ion-conducting elastomers to construct a biomimetic tactile nervous system [86]. In this design, the biodegradable PLA fiber substrate provides the device with high flexibility and bending characteristics. Its cylindrical structure enables the Pt fibers to form a coiled contact with the MXene layer, significantly reducing the conductive area and achieving the ultra-low power consumption (17 nW) required for wearable medical applications, such as radial artery pulse monitoring and joint motion tracking, while maintaining stability over 50 cycles. The device, with its ability to linearly regulate synaptic current through pressure, temperature, and bending angle, can be directly applied to smart prosthetics and environmental

interaction systems, supporting multimodal sensory integration. Concurrently, Wang's team designed a low-power, photosensitive artificial synapse by compositing retinoic acid (RA) with PLA [87]. PLA simultaneously stabilizes the device by suppressing RA crystallization and insulates against leakage currents, directing all carriers to RA. There, conjugated double bonds generate polarization fields at 60 fJ energy inputs for efficient conductance control. Crucially, PLA's transparency synergizes with RA's solar absorption to enable exceptional photoresponsivity.

Overall, synthetic biopolymer-based memristors offer significant advantages in terms of environmental sustainability and biodegradability, making them suitable for applications in smart wearable medical devices, such as real-time pulse monitoring and joint motion tracking. Compared to traditional inorganic memristors, PLA-based memristors excel in low power consumption (with a single pulse consuming only 60 fJ at 10 mV [87]) and biocompatibility, but their electrical performance is relatively weak, with a switching ratio typically around 100. In comparison to natural biopolymer-based memristors, PLA materials demonstrate better thermal stability (operating stably at 55 $^{\circ}$ C [86]), but still fall short in terms of electrical performance and biodegradability.

3 Mechanisms of bio-memristors

Bio-based memristors offer unique advantages in neuromorphic engineering due to their intrinsic biocompatibility, biodegradability, and dual-carrier (ion/electron) transport. Current understanding of memristive switching mechanisms includes various models, such as ion migration, charge trapping/detrapping, phase transition, and valence change [88, 89]. However, the inherent ionic conductivity, abundant functional groups, and complex topological structures of biomaterials predominantly lead to switching behaviors governed by ion migration and charge capture/release in biological memristors [90]. Table 1 summarizes the key performance parameters of some typical biological memristors according to the working mechanism[91, 92].

3.1 Ion migration mechanism

3.1.1 Conductive filament mechanism

The conductive filament (CF) mechanism enables reversible

Table 1 The working mechanisms and performance parameters of typical bio-based memristors

Device structure	Endurance (cycles)	HRS/LRS ratio	VSET/RESET (V)	Retention time (s)	Mechanism	Ref.
Ag/CS-GO/FTO	$> 10^3$	$\sim 10^2$	-4.5 / 1.12	$> 10^4$	Charge trapping/detrapping	[35]
Glass/Ag/pectin/Ag	$> 10^3$	200	-0.5/3	$> 10^4$	Conductive filament	[50]
Au/CQD-chitosan/Al	160	$\sim 10^6$	-0.75/0.75	$> 10^4$	Conductive filament	[91]
Ag/chitosan/FTO	200	4	—	$> 10^4$	Oxygen vacancy migration	[92]
Ag/cellulose/Ag/Si	100	10	0/2	—	Conductive filament	[96]
Al/CS: MWCNTs/ITO	100	395	-1/2.7	$> 10^4$	Proton migration	[105]
Al/agarose@AuNPs/ITO	$> 10^5$	$\sim 10^4$	-0.84/1.95	$> 10^5$	Charge trapping/detrapping	[110]
Al/chitosan-pnipam/ITO	400	$\sim 10^3$	-2.35/0.75	$> 10^4$	Oxygen vacancy migration	[112]
Al/pectin-GO/ITO	100	$\sim 10^4$	-1.21/2.97	$> 10^4$	Conductive filament	[113]
In/carrageenan/Ag/ICAC	100	$\sim 10^6$	-0.65/1.4	$> 10^4$	Conductive filament	[114]
Al/NaCas/ITO	180	20	-0.9/1	$> 10^5$	Charge trapping/detrapping	[115]
Au/IC/ITO	38	$\sim 10^4$	-0.96/1.37	$> 10^4$	Conductive filament	[116]
Al/honey/ITO	200	$\sim 10^4$	-1/1.5	$> 10^4$	Conductive filament	[117]

resistive switching in memristors through bidirectional bias, driven by the dynamic formation and rupture of metallic filaments under electric fields, combining ionic migration (a physical process) and redox reactions (a chemical process). Under positive bias, active metal anodes oxidize to release ions that migrate through the bio-mediating layer toward the cathode, where reduction yields atomic-scale metal deposition. At critical local concentrations, these atoms nucleate into metallic nanoclusters, which gradually grow (akin to stalactite formation in caves) until bridging the electrodes, triggering abrupt resistance drop to a low-resistance state (LRS). Conversely, negative bias induces Joule-heating-assisted thermal oxidation of CFs, resetting the device to a high-resistance state (HRS) (Fig. 4(a)) [93].

Biomaterials play a pivotal role in the conductive CF mechanism owing to their inherent functional groups and structural features. Polar moieties ($-\text{NH}_2$, $-\text{OH}$, $-\text{COOH}$, $-\text{SO}_3\text{H}$) in biomaterials enable precise regulation of ion transport and CF growth via electrostatic adsorption of metal ions. For instance, amino groups ($-\text{NH}_2$) in chitosan stabilize metal ions through chelation, restricting their random diffusion and guiding directional CF formation [94]; carboxyl groups ($-\text{COOH}$) in sodium alginate achieve coulombic trapping of metal ions, facilitating ordered CF

growth [95]. Additionally, natural multiscale structures of biomaterials (e.g., porous networks in cellulose, β -sheet crystalline domains in silk fibroin) provide favorable pathways for metal ion diffusion and transport [96]. These structural merits synergize with the functional group properties to collectively modulate ion transport, nucleation, and CF growth.

The use of bio-materials provides an effective way to optimize the switching characteristics of memristors. However, metal ion migration in bio-based media is still influenced by defects (such as vacancies), leading to uncontrolled nucleation sites for metal clusters. Concurrently, inhomogeneous electric field distributions and thermal gradients within devices promote stochastic conductive filament rupture, significantly degrading operational stability. To overcome these limitations, researchers achieve critical improvements by modulating metal-ion migration barriers to suppress random dispersion [91]. Li et al. employ thermal scanning probe lithography (t-SPL) in 2023 to create nanopatterned cone arrays on silk fibroin films [97]. High-curvature cone tips concentrate electric fields, selectively ionizing Ag^+ to confine filament growth strictly within tip regions while suppressing random Ag^+ diffusion. Geometrically isolated cones further eliminate competitive multi-filament formation. Most recently,

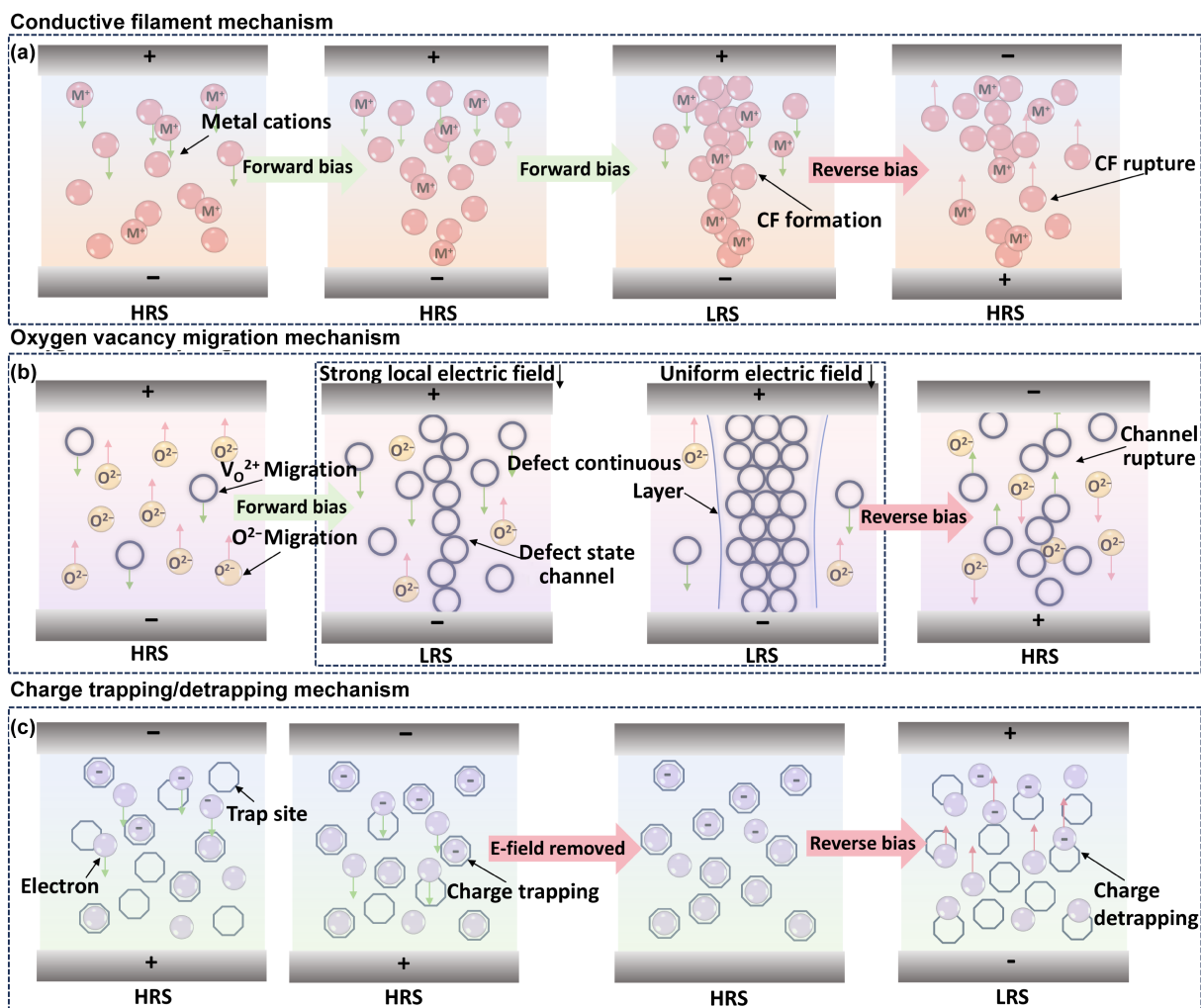


Figure 4 Schematic illustration of the mechanism in biological memristors. (a) Schematic of CF dynamics: Formation and Rupture Regulated by external electric field. (b) Schematic of oxygen vacancy migration process: Middle two diagrams show different defect-state channels (thin filament under strong local E-field; wide filament under uniform E-field). (c) Schematic of charge trapping/detrapping process: left to right—electron trapping/release; trapped charges remain bound (no immediate release) post-trapping (even with E-field removed), keeping device in HRS.

Zhang et al. (2024) blend AgNO_3 with tea polyphenol (TP) solutions to form TP- Ag^+ complexes [71]. TPs reduce Ag^+ to Ag^0 nanoparticles that self-assemble into uniformly dispersed AgNPs. Under applied fields, these AgNPs act as controlled nucleation centers, guiding directional silver deposition into orderly arranged filaments that minimize disordered growth.

The dynamic tunability of CF diameter and resistance enables precise emulation of biological synaptic weight modulation, while pulse-timing-dependent CF formation/rupture kinetics offers unique advantages for neuromorphic devices. Liu et al. (2025) engineered an Al/pectin-GO/ITO synapse, achieving comprehensive synaptic functionalities, including paired-pulse facilitation (PPF), long-term potentiation/long-term depression (LTP/LTD), spike-timing-dependent plasticity, and associative learning [98]. Cyclic pulses triggered gradual saturation, natural decay, and rapid reconfiguration of current responses, mimicking the biological “learning-consolidation-forgetting-relearning” memory cycle.

3.1.2 Oxygen vacancy migration mechanism

In addition to metal ion migration, electric field modulation can dynamically regulate the local conductivity of materials through the directed transport of other ions, such as oxygen vacancy defects (VO^{2+}) and protons (H^+). Under forward bias, local Joule heating causes VO^{2+} to dissociate from lattice oxygen and migrate toward the cathode. When the VO^{2+} concentration exceeds a critical threshold, it forms a quasi-continuous defect state channel, significantly enhancing electron transport and driving the device into the LRS. In contrast, under reverse bias, VO^{2+} migrates toward the anode, leading to the rupture of the defect state channel and restoring the device to the HRS (Fig. 4(b)) [99]. Notably, this conductive behavior exhibits two distinct modes: localized nanowire conduction and extended two-dimensional interface conduction. The dominant mechanism depends on both the material properties and external field conditions. Under strong local electric fields, the field concentration effect promotes VO^{2+} migration, forming discrete conductive filaments, which typically exhibit stronger localized conductivity. Due to the accumulation of oxygen vacancies in a small area, this results in random conductive pathways. In contrast, when the electric field is more evenly distributed, VO^{2+} spreads uniformly across the material interface, forming a continuous conductive layer. This migration in a uniform electric field allows oxygen vacancies to expand evenly along the interface, enhancing the overall conductivity of the device and leading to smoother and more stable resistance state transitions [100].

The integration of biomaterials serves as an “oxygen vacancy buffer layer”, dynamically regulating the generation/annihilation equilibrium of VO^{2+} through their unique molecular architectures and physical properties. This enables precise control over defect-state conductive channels, reducing device parameter fluctuations and prolonging operational longevity. For instance, chitosan exhibits dual functionality as both a VO^{2+} source and trap: Under electric field or humidity stimulation, protonation of its amino groups ($-\text{NH}_2$) releases H^+ ions, inducing VO^{2+} formation via the reduction reaction ($\text{O}^{2-} + 2\text{H}^+ \rightarrow \text{VO}^{2+} + \text{H}_2\text{O}$) [101]. Simultaneously, chitosan's extensive hydrogen-bonding network temporarily immobilizes VO^{2+} , restricting migration to achieve defect-state stabilization. Beyond chitosan, protein-based materials like soy protein leverage porous structures and abundant polar

groups (e.g., $-\text{COOH}$) for enhanced vacancy modulation. During film formation, molecular chain folding creates microcrack regions that preferentially adsorb oxygen ions, while polar groups generate VO^{2+} through dynamic oxygen exchange ($\text{O}^{2-} \leftrightarrow \text{VO}^{2+}$) [102, 103]. The spatial arrangement of these groups establishes hydrophilic channel networks that guide directional VO^{2+} migration, effectively suppressing performance variations caused by random diffusion.

The dynamic behavior of VO^{2+} exhibits significant temperature dependence. High temperatures accelerate the generation/migration rate and annihilation of VO^{2+} , leading to a decrease in device conductivity stability, while low temperatures inhibit the migration rate of VO^{2+} , potentially hindering switching operations. To mitigate this temperature sensitivity, researchers have optimized device structures through strategies such as confining ion diffusion with nanoscale channels and incorporating nanomaterials. In 2023, Hee Lee et al. innovatively developed a transistor featuring polycrystalline silicon (poly-Si) nanochannels integrated with chitosan [104]. This design leverages H^+ release from chitosan's amino groups to directly regulate VO^{2+} formation, circumventing the conventional oxide-based memristors' reliance on high-temperature thermal activation ($> 100^\circ\text{C}$) for vacancy generation. The nanochannels spatially confine proton diffusion paths, effectively suppressing high-temperature disorderly H^+ permeation and consequent electrical parameter drift. Furthermore, this geometric confinement promotes uniform VO^{2+} distribution, enabling linear conductance-pulse response essential for emulating neurosynaptic functionalities like LTP/LTD and spike-timing-dependent plasticity (STDP). The pulse neural network with STDP learning achieved over 91% accuracy on the MNIST benchmark.

3.2 Charge trapping/detrapping mechanism

The charge trapping/de-trapping mechanism leverages functional groups or defect states in biomaterials to achieve purely electronic resistive switching through carrier (electron/hole) capture-release. Under forward bias, carriers are trapped by localized states, drastically reducing free charge density in conductive channels and transitioning the device to the HRS for data storage [105]. Reverse bias facilitates carrier de-trapping, enhancing mobility to restore the LRS for information erasure (Fig. 4(c)). The key feature lies in the nonvolatility of HRS—the resistance persists post-field removal, requiring reverse bias to trigger switching [106, 107]. This mechanism synergizes with space-charge-limited current (SCLC) to modulate switching characteristics: when injected carrier density exceeds intrinsic levels, space-charge effects dominate charge transport, deviating from Ohm's law [108]. SCLC double-logarithmic analysis quantitatively reveals trapping dynamics: linearity at low fields ($n \approx 1$) reflects intrinsic transport; $n > 2$ at higher fields indicates trap-filling; and $n \rightarrow 2$ signifies trap saturation, where carriers overcome barriers to form de-trapping pathways [109].

Recent studies have exploited the energy-level characteristics of biomacromolecules to engineer novel memristive devices through functional-group-mediated charge defect centers. In agarose, the polar hydroxyl groups ($-\text{OH}$) form dynamic hydrogen-bond networks that modulate charge trapping, while the polarity of glycosidic bonds ($\text{C}-\text{O}-\text{C}$) reduces carrier transport barriers, synergistically enabling high-stability, low-power resistive switching. In 2023, Kim et al. developed a flexible memristor by embedding gold nanoparticles (Au NPs) into agarose, achieving ultra-low-power (pJ-level) synaptic plasticity modulation [110]. This device

ingeniously combines agarose's efficient proton transport with Au NPs' deep charge-trapping capability. Remarkably, it maintains LTP/LTD characteristics after 5000 mechanical bending cycles and exhibits a 10^5 -second data retention at 80 °C.

3.3 Multimechanism

Recent developments in advanced composites and memristor architectures have demonstrated that synergistic multi-physics mechanisms rather than single conduction modes govern device performance. In 2024, Yeoh et al. reported a breakthrough pectin-AgNPs composite memristor exhibiting dual modulation mechanisms: (i) Ag^+ ion-migration mediated conductive filament formation enabling robust switching, and (ii) charge trapping at polar functional groups stabilizing the high-resistance state [111]. This cooperative operation yielded exceptional metrics including a switching ratio of 7.2×10^3 , 6.5 V resistive window, and retention exceeding 10^4 s. Systematic SCLC analysis provided definitive mechanism assignment: ohmic conduction (slope = 1.02 ± 0.05) dominated the LRS, while HRS characterization revealed distinct transport regimes—from Ohmic (slope ≈ 1) to trap-filled-limited (slope ≈ 2) to pure space-charge-limited conduction (slope = 3.8 ± 0.4), conclusively validating the dual operational pathways.

In bio-materials, the abundance of polar functional groups, hydrogen-bonded networks, and porous structures not only provides channels for metal ions or defect migration but also introduces numerous trap states, forming a spatial charge layer that enables natural synergy between ion transport and electronic modulation [112–114]. For example, in systems dominated by the filament formation mechanism, charge traps and interface barriers can effectively limit the random growth of filaments, improving device durability and retention characteristics. Compared to the simpler mechanisms found in traditional inorganic memristors, the multi-mechanism synergy in bio-materials, while increasing the complexity of analysis (e.g., the pectin-AgNPs composite system simultaneously demonstrating filament conduction and trap-stabilized high-resistance state dual-channel control [111]), offers greater design flexibility for achieving high switching ratios, low power consumption, and editable bio-memristors [115–117]. However, to address the challenges posed by extreme external conditions (such as temperature, humidity, and strong electric fields), strategies such as multi-layer structural designs [91], carbon-based material composites [98] and surface modification are required to enhance device reliability.

4 Application characteristics and directions of bio-memristors

As an essential component for neuromorphic computing, memristors show unique advantages in implantable medical devices and wearable electronics. Conventional electronic implants often cause immune rejection and may release toxic heavy metal ions, while non-degradable devices staying in the body long-term could potentially lead to tissue cancerization, usually requiring additional surgery for removal. These limitations highlight the urgent need to develop biocompatible neuromorphic electronics. Biomaterials, with their inherent biocompatibility, biodegradability, and excellent electrical properties, have become ideal candidates for building next-generation neuromorphic devices. This chapter will delve into the crucial role of biocompatibility and biodegradability in neuromorphic devices, outlining the relevant evaluation methods

and analyzing the unique performance advantages offered by biomaterials. Additionally, it will discuss the application directions of bio-based memristors, as well as the advantages and challenges they present in current applications.

4.1 Biocompatibility

In practical neuromorphic device applications, especially those requiring direct contact, implantation, or tight integration with neural tissue, such as high-precision brain-machine interfaces (BMIs) for prosthetic control or communication, excellent biocompatibility is essential [118]. First, all materials and potential byproducts must meet stringent cytotoxicity standards to prevent adverse interactions with host cells/tissues. Second, both materials and device architectures should minimize risks of acute immune responses, avoiding failure caused by complex physiological environments [119]. For memristors as key neuromorphic components, superior biocompatibility becomes paramount. Biomaterials inherently offer low toxicity and metabolic compatibility, providing new opportunities for biocompatible neuromorphic memristors [120, 121]. Currently, bio-compliant neuromorphic devices remain in early-stage development, with most studies validating safety only through *in vitro* tests, while a limited subset employs animal models for *in vivo* assessment of true physiological responses [122].

In vitro testing typically employs material immersion in cell culture media to simulate biological responses under physiological conditions [123, 124]. This approach bypasses complex ethical approvals required for animal studies while enabling efficient preliminary screening by monitoring critical parameters like cell viability and inflammatory mediator secretion [125]. In 2022, Zhang et al. utilized single-celled *Paramecium* cultures to assess the cytotoxicity of chlorophyll-a-based synaptic transistors embedded in wood-derived cellulose nanofiber (WCN) substrates [126]. Under strictly controlled environmental parameters, parallel cultures were established, containing either synaptic transistor arrays (experimental group) or blank controls. Results showed normal *Paramecium* viability in both groups after 5 days, with partial survival persisting in the experimental group by day 10. This demonstrates the material's negligible cytotoxicity and excellent biocompatibility.

The L929 cell line, derived from mouse subcutaneous connective tissue, has emerged as an ideal *in vitro* model for evaluating the biocompatibility of neuromorphic devices such as memristors, owing to its exceptional electrical responsiveness, distinct morphological characteristics, and glia-like metabolic properties [127]. Furthermore, this cell line has been designated as a recommended testing model by ISO 10993-5 standards, offering a reliable benchmark for biomaterial assessment due to its stable *in vitro* culture characteristics and standardized biological parameter system [128, 129]. In 2025, Kim et al. developed a lactose-based implantable memristor device [130]. To validate its suitability for bio-implantable electronic applications, the team conducted *in vitro* cytotoxicity assessments using L929 mouse fibroblasts. Experimental data revealed cell viability rates of 97.2% after 24 h, 88.8% after 48 h, and consistently above 80% after 72 h (Fig. 5(a)). Additionally, device performance optimization studies demonstrated that memristors fabricated with 3 wt.% lactose concentration maintained both electrical functionality and excellent biocompatibility, sustaining > 90% cell viability after 120 h—fully meeting implantable application requirements. In the same year, Moon's research team verified the biocompatibility of a mucin-

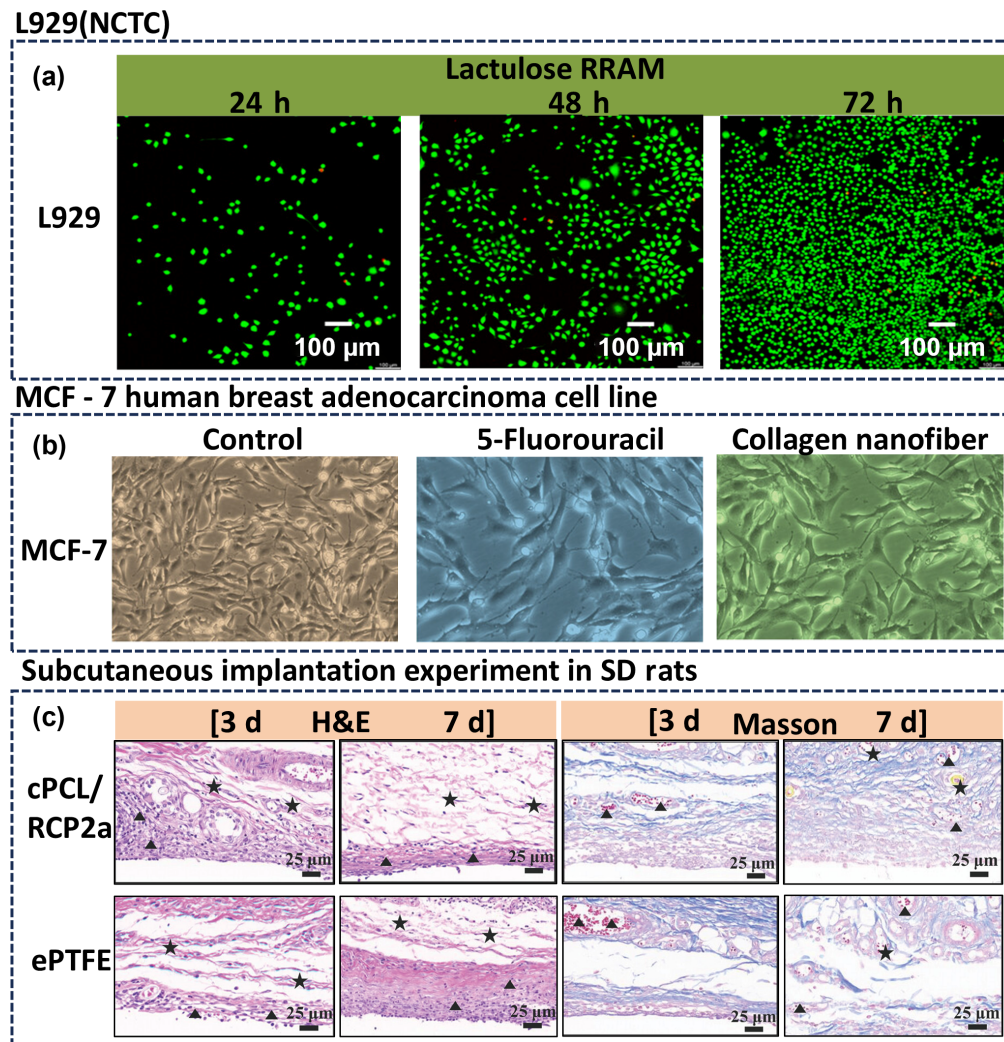


Figure 5 Biocompatibility evaluation/testing of bio-based memristive devices and biomaterials. (a) Effect of device (immersed in L929 cell line) on L929 cell proliferation. Reproduced with permission from Ref. [130], © American Chemical Society 2025. (b) Antitumor effect of collagen nanofiber memristor, referenced to control group & 5-fluorouracil. Reproduced with permission from Ref. [133], © Wiley-VCH GmbH 2024. (c) *In vivo* biocompatibility of cPCL/RCP bilayer composite (SD rat subcutaneous implantation; control: commercial ePTFE). Reproduced with permission from Ref. [134], © Wiley-VCH GmbH 2024.

based memristor network through L929 cell culture experiments [131], reporting remarkable cell viability exceeding 100% (108.46%) after 72 h of cultivation—compelling evidence of superior cellular adaptability within this material system.

Long-term implantation of biomaterials may trigger persistent localized inflammation and, in extreme cases, promote tumorigenesis. Therefore, neural implant devices designed with anti-inflammatory and anti-tumor properties can significantly enhance long-term biocompatibility and clinical applicability [132]. In 2024, Kasturi et al. developed a bio-memristor based on collagen nanofibers (Col NFs) [133]. The team evaluated the biocompatibility of Col NFs using an *in vitro* culture system of MCF-7 breast cancer cells, with a parallel control group treated with the standard chemotherapeutic drug 5-fluorouracil (5-FU) to assess anti-tumor effects. Experimental observations revealed a 50% inhibition rate of MCF-7 cancer cells after 24 h, though cancer cell proliferation rebounded to 90% by 48 h (Fig. 5(b)). These results demonstrate that the device exhibits excellent biocompatibility with mild anti-cancer activity, making it suitable for neuromorphic implant development.

While *in vitro* testing offers simplicity and rapid turnaround,

biomaterials with true clinical applicability must first undergo rigorous *in vivo* biocompatibility validation. In 2024, Zhang et al. developed a novel moisture-responsive bilayer vascular graft (cPCL/RCP) structured with crosslinked polycaprolactone (cPCL) as the inner layer and regenerated chitosan/polyvinyl alcohol composite (RCS/PVA) as the outer layer [134]. Through subcutaneous implantation experiments in Sprague-Dawley (SD) rats, the team comparatively evaluated the *in vivo* biocompatibility of the cPCL/RCP bilayer composite versus commercial ePTFE material (Fig. 5(c)). The cPCL/RCP group demonstrated superior tissue response at 3- and 7-days post-implantation: H&E staining revealed reduced inflammatory cell infiltration (Δ) alongside robust fibroblast proliferation ($\star$), indicating controlled inflammatory response; Masson's trichrome staining showed more organized collagen fiber alignment with significant neovascularization ($\star$); CD68 immunostaining further confirmed lower macrophage infiltration (Δ) in this group. These collective findings demonstrate cPCL/RCP's exceptional biocompatibility and tissue regenerative capacity, positioning it as an ideal candidate for implantable neuromorphic devices.

4.2 Biodegradability

The annual global generation of 53.6 million tons of electronic waste poses significant ecological threats through heavy metal contamination (e.g., Cd, Pb) [135–137]. Concurrently, chronic biocompatibility concerns persist regarding permanently implanted medical electronics. These challenges underscore the critical need for developing neuromorphic electronic devices with controlled degradability and biosafety. Degradability refers specifically to the progressive breakdown of materials into non-toxic constituents under defined environmental conditions [138–140]. Researchers generally evaluate this property systematically using standardized test media (Table 2), including deionized water, pH buffer solution, and hydrated soil [141, 142]. Biomaterials exhibit intrinsic degradability through cleavage of ester/peptide bonds in their molecular structures via hydrolysis, oxidation, or enzymatic action. Unlike conventional electronics, their degradation byproducts undergo complete environmental assimilation without persistent pollution [143–152]. These attributes establish biomaterials as ideal candidates for eco-friendly degradable electronics development.

Most current bio-memristors employ partially biodegradable designs where only the dielectric layer fully degrades while retaining bottom electrodes and substrates for recycling. This approach ensures both superior electrical performance and clinical practicality [153, 154]. In 2024, Hosseini et al. developed a Mg/Collagen/ITO memristor using fish scale-derived collagen [59]. Under hydration, the collagen layer first dissolved to form a protein hydrogel, followed by complete hydrolysis of the Mg electrode within 2 min, and achieving full device degradation (excluding ITO electrodes) within 5 min (Fig. 6(a)). Concurrently, Li's team engineered a Mo/ADM/Mo memristor based on acellular dermal matrix (ADM) [145]. Experiments demonstrated complete ADM active layer degradation in soil within 3 days, a process mediated by soil microbiota (Fig. 6(b)). Enzymatic reactions progressively decomposed collagen into nontoxic intermediates (amino acids, short peptides), ultimately mineralizing them into harmless small molecules (CO_2 , H_2O , NH_3).

To realize fully biodegradable bio-memristors, researchers used the gelatin-based composite hydrogel films as flexible supporting layers. These films, synthesized by integrating functional nanomaterials with gelatin, simultaneously exhibit excellent

mechanical support and environmental friendliness [155]. In 2023, Shaikh's team fabricated a fully degradable W/Mg/PVP/Mg/CHS memristor using chitosan hydrogel film substrates and magnesium electrodes [147]. Full device degradation was achieved within 300 s (Fig. 6(c)). Advancing this work in 2024, Wang's group developed freestanding artificial optoelectronic synaptic devices using gelatin-polydopamine nanoparticle (PDA NP) composite films as substrate replacements [143]. Remarkably, these devices demonstrated complete degradation in 60 °C deionized water within just 5 minutes (Fig. 6(d)). Crucially, all components (gelatin matrix, glycerol, PDA NPs) exhibited exceptional biodegradability, ensuring full environmental compatibility.

Overall, bio-memristors, with their inherent biocompatibility and biodegradability, are particularly well-suited for applications that require direct contact with biological systems, such as wearable health monitoring devices and smart implantable medical instruments. Compared to traditional rigid inorganic memristors, biomaterials not only exhibit lower toxicity but also simulate the interface microenvironment of natural tissues through factors such as surface hydrophilicity, molecular functional groups, and more, significantly reducing the risk of immune rejection by the body. Meanwhile, the biodegradability of bio-materials aligns perfectly with the growing demand for “green electronics”, making them ideal for disposable medical sensors and short-term environmental monitoring devices that need to degrade naturally after use, avoiding electronic waste. However, these materials face challenges under high temperature, humidity, or long-term operation conditions. The conformational changes in biological molecules can lead to resistance state drift and performance degradation. Researchers have explored diversified solutions, such as chemical crosslinking (e.g., using PVP as a thermal stabilizer to strengthen collagen fiber networks through hydrogen bonding [133]), surface functionalization (e.g., the formation of MgO_x layers through Mg-lactose interfacial reactions to resist enzymatic degradation [130]), and material composites. These strategies have significantly improved the electrical stability and durability of bio-memristors.

4.3 Application of bio-memristors

With the continuous development of neuromorphic computing, bio-based neuromorphic devices have achieved system-level integration with biological components, establishing a functional

Table 2 Degradation characteristics of memristive devices based on biomaterials^a

Device structure	Biomaterial	Degradative environment	Degradation mechanism	Degradation time	Complete degeneration	Ref.
Au/pectin-DNTT/Au	Pectin	DI water	Hydrolytic degradation	100 s	—	[37]
Mg/collagen/ITO	Collagen	DI water	Hydrolytic degradation	5 min	—	[59]
Cu/PDA-IGH/IGH/Cu	Gelatin	DI water (60 °C)	Hydrolytic degradation	5 min	○	[143]
Si/Li-IGH/IGZO/Al	Gelatin	DI water (40 °C)	Hydrolytic degradation	10 min	—	[144]
Mo/ADM/Mo	ADM	Active soil matrix	Enzymatic hydrolysis	72 h	—	[145]
Mg/Ag-Chitosan/ITO	Chitosan	DI water	Hydrolytic degradation	10 min	—	[146]
W/Mg/PVP/Mg/CHS	Chitosan	DI water	Hydrolytic degradation	10 min	○	[147]
Ag/PVA-ECM/ITO	PVA	PVA dehydrogenase	Microbial biodegradation	30 s	—	[148]
Ag/p-SF/ITO	SF	Papain	Enzymatic hydrolysis	10 min	—	[149]
Al/gelatin/Ag/BC	Gelatin/collagen	Active soil matrix	Enzymatic hydrolysis	120 h	○	[150]
Au/dextran-DNTT/Au	dextran	DI water	Hydrolytic degradation	17 s	—	[151]
Al/C ₃ N ₄ -chitosan/IZO/ITO	Chitosan	DI water	Hydrolytic degradation	10 min	—	[152]

^a “○”: Full degradation of the device under the corresponding conditions. “—”: Partial degradation, indicating that the device is not fully degraded under the test conditions, and part of the structure remains.

Biodegradability

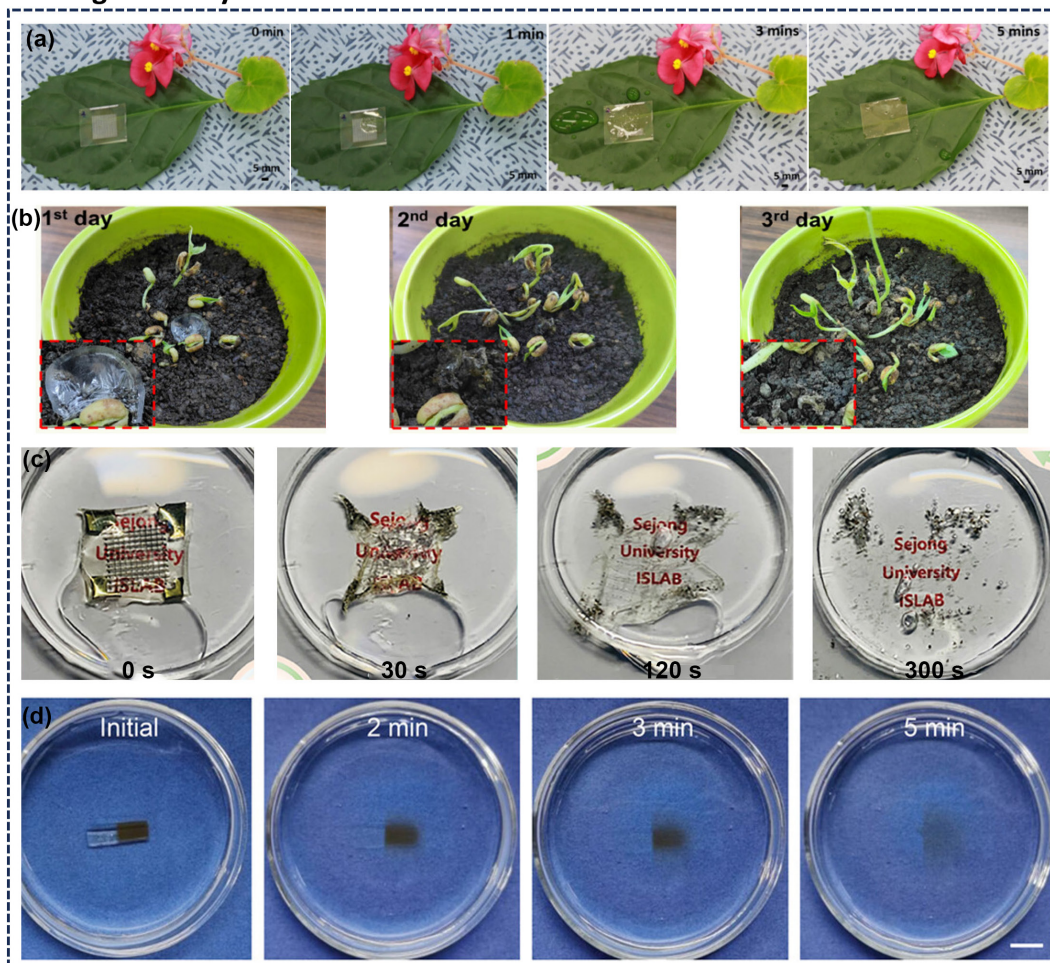


Figure 6 Testing the biodegradability of bio-based memristive devices. (a) Entire memristive device (excl. ITO electrode), fully degraded in deionized water. Reproduced with permission from Ref. [59], © American Chemical Society 2024. (b) Memristor active layer (ADM-based), gradual degradation in active soil. Reproduced with permission from Ref. [145], © American Chemical Society 2024. (c) Memristive device on chitosan hydrogel substrate, fully degraded in deionized water. Reproduced with permission from Ref. [147], © Shaikh, M. T. A. S. et al. 2023. (d) Optoelectronic synaptic device with gelatin-PDA NPs composite substrate, fully degraded in deionized water at 60 °C. Reproduced with permission from Ref. [143], © Wiley-VCH GmbH 2024.

bridge between artificial sensory nerves and biological systems. Specifically, these studies focus on bio-based memristors as the core components to accurately simulate signal transmission and synaptic plasticity in biological neural systems [110]. One key research branch involves mimicking the visual processing and synaptic functions of the human eye, integrating biological components into artificial retinas and neuromorphic vision systems, thus bringing their visual signal processing and memory capabilities closer to biological systems [156]. For example, by integrating bio-based memristors made from chlorophyll and glucose, a low-power neuromorphic vision system can be developed that intelligently processes visual signals and performs color recognition [157]. This neuromorphic vision system closely emulates the biological visual system's function—processing RGB color information and regulating synaptic plasticity, achieving a color recognition accuracy of 91.4%, with an energy consumption of only 102.1 fJ (Fig. 7(a)). Furthermore, after 30 days of environmental aging (25 °C/65% RH), the chlorophyll-glucose memristor retains 93.6% of its initial switching ratio. This stability is attributed to the molecular self-protection mechanisms of the biomaterials. The conjugated structure of the chlorophyll porphyrin ring inhibits photo-oxidative

damage, while the coordination bond (Mg–O, ~ 214 kJ/mol) formed between the magnesium core and glucose hydroxyl groups prevents ion migration, maintaining 87.3% of the device's effectiveness in saline solution. This long-term stability and low power consumption make it particularly suitable for wearable devices and smart robots, with significant application potential in simulating the learning and memory functions of biological neural systems.

Recently, bio-based memristors have been applied in advanced fields such as automatic control systems and smart robotics [158]. For instance, the silk fibroin (SF)@tungsten disulfide (WS₂) memristor plays a crucial role in automatic braking systems [159]. The device controls ion migration with light, mimicking synaptic plasticity, and adjusts its resistance state in response to red light stimuli. When the light signal is input, the memristor's resistance changes, affecting the control system's electrical signals. This enables the integration of light-controlled sensing, storage, computation, and control, allowing the system to respond accurately to light signals and make decisions. For example, upon detecting a red light signal, the system initiates the automatic braking function based on the memristor's state change to ensure safety (Fig. 7(b)). The SF@WS₂ memristor operates stably at 0.65 V,

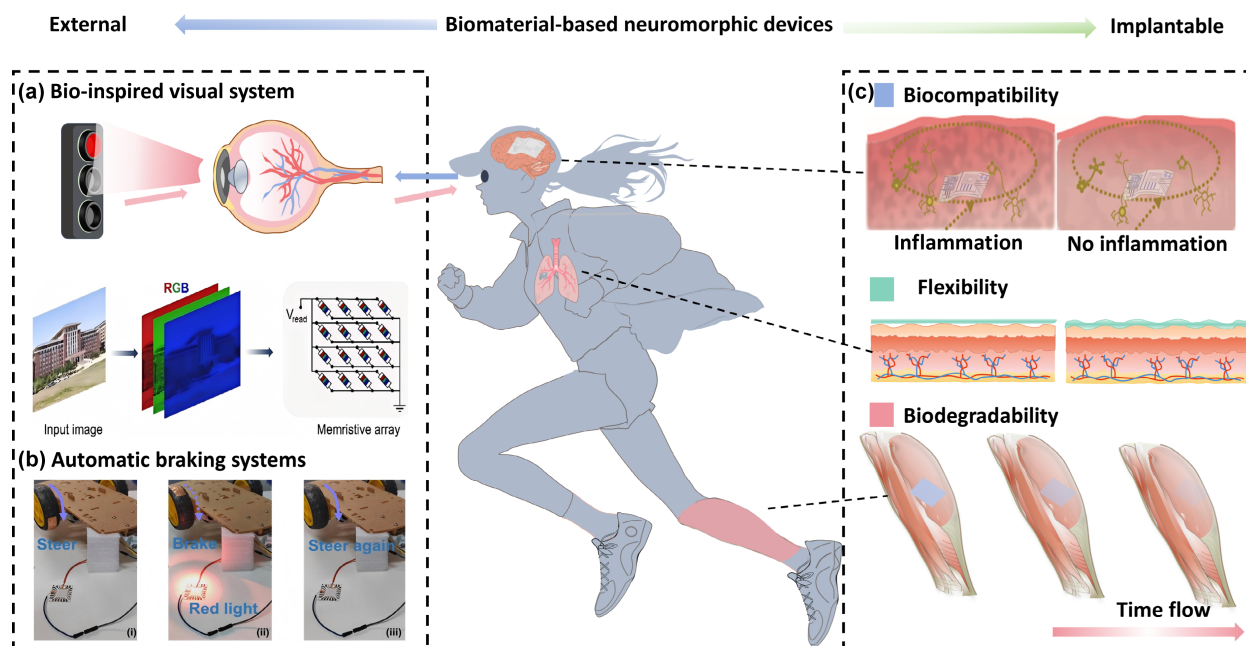


Figure 7 Potential applications and advantages of bio-based memristive devices. (a) The processing process of the bio-based memristor-based bio-inspired visual system made from chlorophyll and glucose. Reproduced with permission from Ref. [157], © Wiley-VCH GmbH 2025. (b) Automatic control system based on silk fibroin material memristors. Reproduced with permission from Ref. [159], © Elsevier B.V. 2025. (c) Schematic diagram demonstrating the applications and advantages of biomaterial-based neuromorphic memristors in implantable and wearable devices.

making it ideal for low-power, long-term applications like autonomous driving and smart robotics, while its integration of sensing, storage, and computing functions enhances flexibility and intelligence in control systems.

Biomaterials, with superior biocompatibility, biodegradability, long-term stability, and low power consumption, show great potential in implantable medical devices such as smart prosthetics and health monitoring systems. The neuromorphic memristor based on natural mucin (MNM array) demonstrates superior performance in these applications [131]. This device successfully mimics the effective learning and memory functions of biological synapses, as evidenced by a pair-pulse facilitation (PPF) experiment, where the PPF index reached 122.65%, showing efficient signal transmission and plasticity. Most importantly, the mucin-based bio-memristor operates stably at low power (1.06 nJ per pulse) and exhibits significant biocompatibility, with cell viability reaching 108.46% after 72 h of cell culture, proving its long-term stability and safety within biological systems (Fig. 7(c)). However, bio-memristors still face challenges, particularly in their electrical characteristics [114]. The inherent structure of the material can lead to some randomness, affecting the long-term stability of the device. To address this, researchers have enhanced the device's stability by employing interface engineering and material composites, such as introducing an MgO interface layer between the mucin and metal electrodes, which reduces ion migration issues. Additionally, improving the mechanical flexibility and biological adaptability of the device to suit the complex physiological environment of the human body remains a key challenge for its use in implantable devices.

5 Conclusions and outlook

This review summarizes recent advances in biomaterial-based memristors, thoroughly examining their potential for sustainable neuromorphic electronics while systematically analyzing their

performance advantages over conventional materials. Biopolymeric memristors have established a well-defined material classification system (e.g., protein-based, polysaccharide-based, DNA-based, and synthetic biopolymer categories). Their performance is precisely tunable through biomolecular structural design and hybrid ion/electron transport mechanisms [24]. Such devices show unparalleled biomedical advantages, including dynamic resistance switching faithfully simulating synaptic function, and superior biocompatibility reducing immune responses (such as inflammation) and toxicity in implantable systems [104]. Critically, programmable degradation, enabled by material engineering (e.g., gelatin-ion/PDA nanoparticle composites), spans partial degradation (selective component breakdown) to full-device disintegration (including substrates) [142], offering an eco-innovative solution for electronic waste management.

In recent years, bio-based memristors have gained significant attention in neuromorphic electronics due to their potential in smart prosthetics, bionic sensing systems, and implantable medical devices. The development of memristive devices with high biofidelity, controllable degradability, and 3D integrability is key to advancing next-generation wearable and implantable neuromorphic systems. However, the environmental sensitivity of biomaterials, while enabling controlled degradation, also makes their electrical properties vulnerable to factors like temperature and humidity, leading to resistance state drift, performance degradation, and even functional failure. Additionally, the amorphous nature of biomaterials causes random ion migration, which affects long-term device stability [160]. To overcome these issues, researchers have used molecular engineering strategies such as material composites and chemical cross-linking, including covalent modifications and bioinspired ion-channel designs, improving both environmental stability and electrical performance. For example, controlling the degree of substitution (DS) of cellulose esters and blending them

with PHBV and PLA enhances their environmental stability, mechanical, and thermal properties [161]. Moreover, the precise self-assembly of porphyrin molecules created ion channels with high selectivity, maintaining an ion selectivity of 18.6 even after 100 h of continuous operation, improving the reliability of bio-based memristors [162].

Although bio-based memristors have exhibited improved short-term stability, current optimization strategies still fail to meet the long-term reliability requirements for demanding applications such as implantable medical devices and smart robotics. Simple “patchwork” solutions cannot address the fundamental challenges in long-term reliability, rendering multidisciplinary collaborative design at the material, interface, and device levels essential for achieving significant progress. For instance, integrating the photosensitive proton pump function of bacteriorhodopsin (bR) with memristor arrays enables the development of high-efficiency light-controlled neuromorphic chips, where light-modulated ion migration mimics synaptic plasticity to realize low-power computing [163]. Additionally, optimizing thermal and humidity tolerance via CRISPR protein engineering while retaining biodegradability and self-powering capabilities offers great promise for resolving stability issues in complex environments [164]. Nevertheless, with continuous technological iterations, bio-based memristors hold substantial potential for widespread applications in smart healthcare, wearable devices, and neuromorphic computing [165].

Overall, bio-based memristors, with their excellent neuromorphic characteristics, biocompatibility, and degradability, offer innovative approaches for the development of next-generation neuromorphic devices. These devices show significant application potential in fields such as bio-inspired neural systems, flexible wearable technology, and implantable biomedical devices. In the future, bio-material-based neuromorphic devices are expected to integrate molecular electronics and cellular engineering to build cell-level functional circuits, enabling efficient interconversion between biological and electrical signals, thus driving the integration of perception, computation, and regulation. With the deepening of interdisciplinary collaboration, bio-based memristors hold the potential for breakthroughs in sustainable neuromorphic electronics, pushing the field toward more efficient and sustainable development.

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Declarations conflict of interest

The authors declare no interest conflict. They have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Author contribution statement

X. C. Z.: Writing – original draft, visualization. H. T. W.: Writing – original draft, project administration. X. Z. Z.: Review, project administration. D. Y. W.: Review, project administration.

Use of AI statement

During the preparation of this work, the authors used ChatGPT to polish the English language and improve the readability of the manuscript. After using this tool, the authors reviewed and edited the content as needed and take full responsibility for the content of the publication.

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