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Ion-migration memristive synaptic devices: mechanisms, device architectures, and integration strategies

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Abstract: Ion-migration memristive synaptic devices provide a physical route for hardware neuromorphic computing by using ionic redistribution, conductive-filament evolution, and interfacial barrier modulation to regulate synaptic weights. However, existing studies are often discussed according to specific material systems or individual device demonstrations, which makes it difficult to compare how different ion-migration mechanisms determine synaptic behavior, device stability, and integration potential. This mini review organizes recent progress from the perspective of operating mechanism and device type. Electrochemical metallization (ECM)-based synaptic devices are discussed with emphasis on metallic-filament formation and the challenge of achieving gradual and reproducible conductance modulation. Filamentary valence-change memory (VCM)-based devices are reviewed in terms of oxygen-vacancy channel evolution, while interfacial VCM devices are examined through interfacial ionic modulation and barrier-controlled analog switching. Hybrid ECM-VCM devices are further discussed as integrated designs that couple multiple ionic processes to balance switching window, stability, and multifunctionality. By linking mobile ionic species, switching pathways, and synaptic functions, this review provides a mechanism-based framework for understanding ion-migration memristive synaptic devices and for identifying the material, interface, and device-level issues that remain before scalable neuromorphic hardware can be realized.

Key words: ion migration; memristor; artificial synapse; neuromorphic computing

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1. Introduction

The continued scaling of conventional semiconductor hardware has been increasingly constrained by quantum tunneling, heat dissipation, and fabrication cost[1–3]. In conventional von Neumann systems, the physical separation of processing and memory units gives rise to persistent data-transfer bottlenecks, often referred to as the "memory wall", which has become a major limitation for artificial-intelligence and big-data computing[4, 5]. The low-power and massively parallel information processing of the human brain, together with the central role of synapses in neural computation, provides an important reference for developing neuromorphic hardware[6, 7].

Memristive synaptic devices have become an important device class in neuromorphic electronics because their conductance states can be regulated by external stimuli and mapped to synaptic weights[8–11]. Among various memristive devices, ion-migration memristors are especially relevant to artificial synapses because their resistance modulation is governed by the redistribution of mobile ions or ionic defects[12, 13]. This feature is physically related to biological synapses, where ion fluxes and concentration-dependent relaxation processes participate in signal transmission, plasticity regulation, and memory transition. In ion-migration memristors, the motion, accumulation, diffusion, and recombination of mobile ionic species can record the history of electrical pulses and translate it into volatile or nonvolatile conductance evolution. Therefore, these devices can emulate typical synaptic functions, including short-term plasticity (STP), long-term plasticity (LTP), potentiation/depression, spike-timing-dependent plasticity (STDP), and the transition from short-term to long-term memory[14, 15]. More importantly, the type of mobile ionic species and their migration pathways determine whether conductance modulation is dominated by metallic-filament evolution, oxygen-vacancy channel formation, interfacial barrier modulation, or coupled ionic processes. This mechanism dependence is the main reason why ion-migration memristive synaptic devices are discussed separately from memristive synapses dominated mainly by purely electronic trapping, ferroelectric polarization, phase transition, or other non-ionic switching processes[12–16].

Recent studies have improved ion-migration memristive synaptic devices through material design, interface regulation, defect engineering, and device-structure optimization[12, 13, 16]. Electrochemical metallization (ECM) devices use metal-ion migration and metallic-filament formation to achieve low-voltage switching and large conductance modulation, whereas valence-change memory (VCM) devices rely mainly on oxy-

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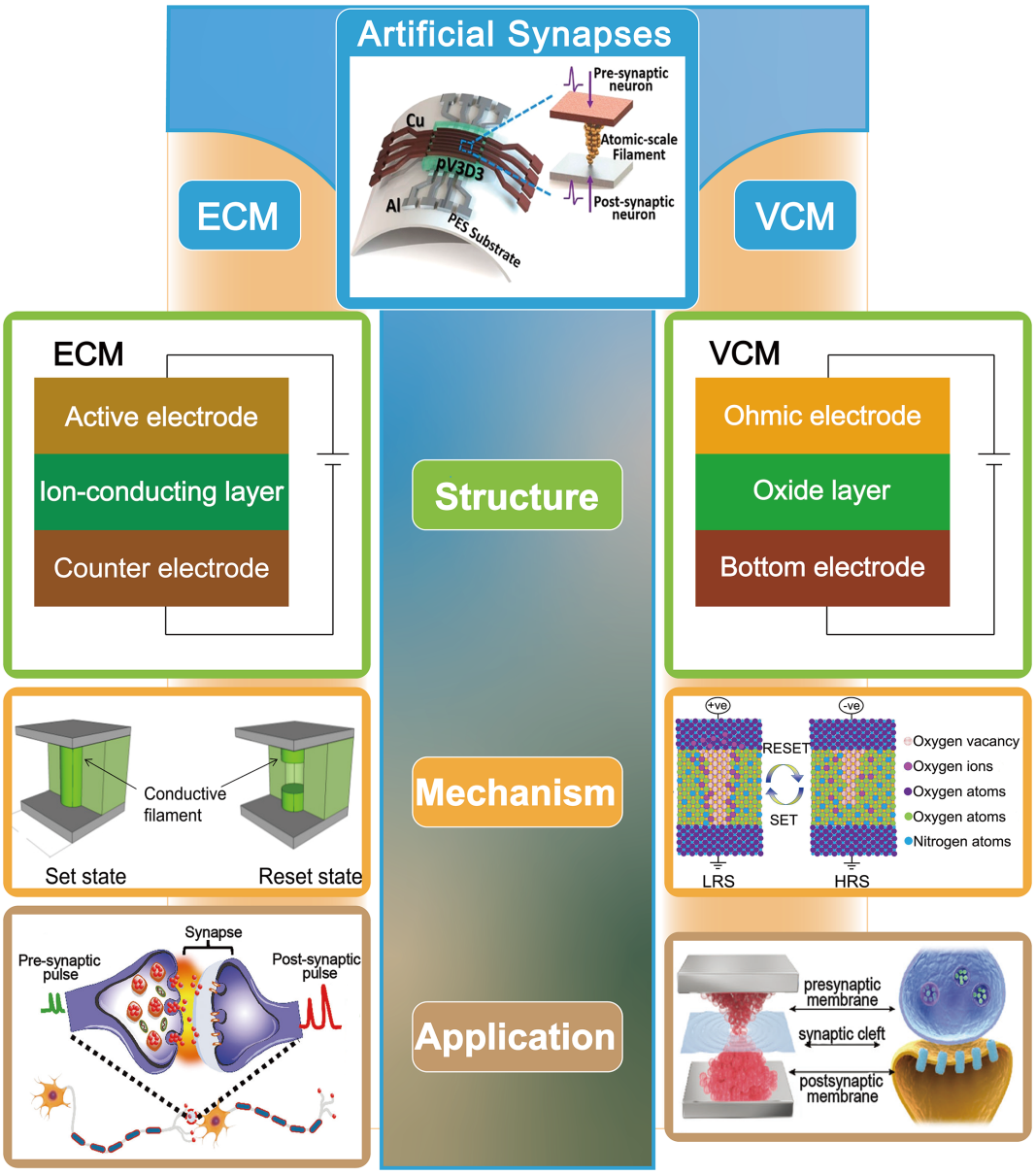

Fig. 1. (Color online) Mechanism-based overview of ion-migration memristive synaptic devices, including ECM and VCM structures, representative switching processes, and synaptic-function mapping. The ECM and VCM device-configuration schematics were drawn by the authors based on the device concepts described in Ref. [17]. Other schematics were adapted with permission from Refs. [18–22]. Copyright 2019, American Chemical Society; Copyright 2016, IOP Publishing Ltd.; Copyright 2024, Elsevier Ltd.; Copyright 2025, Wiley-VCH GmbH.

gen-vacancy migration, which can form localized conductive channels or modulate interfacial barriers[12, 13, 16]. These different ion-migration processes provide diverse routes for synaptic-function emulation, but they also lead to distinct limitations in analog tunability, device uniformity, retention, endurance, and array integration. At present, many reported studies are still discussed according to specific material systems or individual device demonstrations. As a result, the relationship among mobile ionic species, switching pathways, synaptic behavior, and device-level performance has not been sufficiently clarified. Therefore, a mechanism-based review is needed to compare these device types and summarize the design logic behind ion-migration memristive synaptic devices. An overview of the major device structures, ion-migration mechanisms, and their mapping to synaptic functions is provided in Fig. 1.

This review focuses on ion-migration memristive synaptic devices and organizes recent progress according to operating mechanism, device type, and integration strategy. First, the operating principles of ion-migration memristors are introduced, and the classification basis used in this review is established. Then, ECM-based synaptic devices are discussed with emphasis on how metal-ion migration and metallic-filament evolution determine conductance modulation. Filamentary VCM-based devices are reviewed by focusing on oxygen-vacancy channel formation and regulation, while interfacial VCM devices are summarized from the perspective of interfacial ionic modulation and barrier-controlled analog switching. Furthermore, hybrid ECM-VCM devices are discussed as integrated systems that couple multiple ionic processes to balance switching window, stability, and multifunctionality. By linking ion-migration mechanisms with synaptic functions and device-design strategies, this review aims to provide a clear reference for understanding current progress and remaining challenges in ion-migration memristive synaptic devices[13, 16].

2. Mechanisms and classification framework of ion-migration memristors

Ion-migration memristors change their resistance through electric-field-driven ionic transport[23–25]. In synaptic devices, this process is important because the movement, accumulation, diffusion, or recombination of mobile ions can be translated into conductance-state evolution under electrical pulses[26–28]. Unlike purely electronic switching, ion-mediated resistance modulation usually involves structural or interfacial changes inside the device, which makes the conductance history-dependent and suitable for emulating synaptic plasticity. The key question is therefore not only whether ion migration can induce resistive switching, but also which ionic species move, where they accumulate, and how their migration pathway determines the continuity, stability, and reversibility of conductance modulation.

According to the dominant mobile species and switching pathway, ion-migration memristors can be classified into electrochemical metallization memory (ECM) and valence-change memory (VCM)[23–25, 29–31]. ECM is governed by redox reactions of active metal species and their migration through the dielectric layer[25, 29, 30]. A typical ECM cell consists of an active electrode, an ion-conducting medium, and an inert counter electrode. Under an applied electric field, metal atoms at the active electrode are oxidized into cations and migrate toward the inert electrode, where they are reduced and accumulate to form metallic conductive filaments. This process switches the device from a high-resistance state to a low-resistance state. Reversing the polarity induces electrochemical dissolution or rupture of the metallic filament and returns the device to the high-resistance state[25, 29, 30]. Because metallic filaments can provide highly conductive pathways, ECM devices usually show low operating voltages and high switching ratios. However, the nucleation and growth of these filaments are often localized and stochastic, which can cause abrupt conductance jumps and limit smooth analog weight updating[29, 30].

VCM devices rely mainly on the migration and redistribution of oxygen vacancies and, in many oxide systems, the coupled motion of oxygen ions[25, 31, 32]. Compared with ECM, the mobile species in VCM are not metal cations from an active electrode but anion-related defects generated or redistributed inside the oxide. Depending on the spatial distribution and regulation mode of oxygen vacancies, VCM can be further divided into filamentary and interfacial types[25, 31–34]. In filamentary VCM, oxygen vacancies aggregate along localized paths to form conductive channels[25, 31, 32, 35]. This localized switching mode is attractive for fast operation and high-density integration, but it also suffers from variability because the formation and rupture of vacancy-rich channels are sensitive to local defects, electric-field concentration, and thermal effects. In interfacial VCM, oxygen vacancies accumulate or deplete over a broader region near the electrode/oxide interface, thereby modulating the Schottky barrier or interfacial resistance. This large-area modulation generally supports smoother and more reproducible conductance evolution, although the switching window is often more limited than that of filamentary devices[33, 34]. The representative switching modes of ECM, filamentary VCM, and interfacial VCM—including metallic-filament formation/rupture, oxygen-vacancy-channel evolution, interfacial ionic redistribution, and Schottky-barrier modulation—are summarized in Fig. 2.

Because the resistance change is driven by ionic motion, pulse parameters such as voltage amplitude, duration, and repetition frequency can be used to regulate the diameter of conductive filaments or the concentration of oxygen vacancies[11, 14, 26–29]. This feature underlies the emulation of multiple forms of synaptic plasticity. By adjusting ion-migration rate and filament morphology, the conductance can be increased or decreased progressively, corresponding to synaptic potentiation and depression[14, 27–29]. The diffusion and recombination kinetics of mobile ions determine the retention of resistance states and therefore support the transition from short-term to long-term memory[26, 28].

This mechanism-based classification also defines the main design targets of ion-migration memristive synaptic devices. ECM devices focus on controlling metal-ion injection, migration, and metallic-filament evolution. Filamentary VCM devices focus on regulating oxygen-vacancy generation, aggregation, and recombination within localized channels. Interfacial VCM devices focus on tuning defect distribution, electrode work function, and barrier modulation at the interface. These mechanisms are not simply ranked as better or worse; rather, they offer different balances among switching ratio, analog tunability, uniformity, retention, and integration suitability[23, 25, 33, 34]. Therefore, the following sections discuss ECM-based synaptic devices, filamentary VCM-based synaptic devices, interfacial VCM devices, and hybrid ECM-VCM designs according to their dominant ionic processes and corresponding regulation strategies.

3. ECM-based synaptic devices

In ECM-based synaptic devices, the central issue is how to convert stochastic metallic-filament growth into gradual and reproducible conductance modulation[23, 25, 30, 36, 37]. Metal-ion migration and reduction can produce fast low-voltage switching and high switching ratios, but the nucleation site, growth direction, and rupture position of metallic filaments are difficult to control[30, 36, 37]. This stochasticity often causes abrupt conductance changes and cycle-to-cycle variation, which are unfavorable for analog synaptic-weight updating. Current ECM studies therefore mainly focus on three regulation strategies: guiding metal-ion migration pathways, smoothing filament formation and rupture, and extending ECM devices toward multimodal synaptic functions[21, 36–38]. The following examples are discussed along this sequence to show how material design, interface regulation, and functional coupling address different stages of metallic-filament evolution[21, 36–38].

The first strategy is to regulate metal-ion migration by introducing composite active layers, conductive islands, or nanoparticles. These structures provide preferred nucleation sites or confined migration pathways, thereby reducing the randomness of filament growth[21]. Jana *et al.*[21] incorporated Ag nanoparticles into thermally exfoliated graphitic carbon nitride nanosheets to form a hybrid active layer (Fig. 3(a)). The Ag nanoparticles acted as conductive islands that confined mobile-ion diffusion and supported bipolar resistive switching at very low operating voltages. The device emulated synaptic functions such as STDP and spike-number-

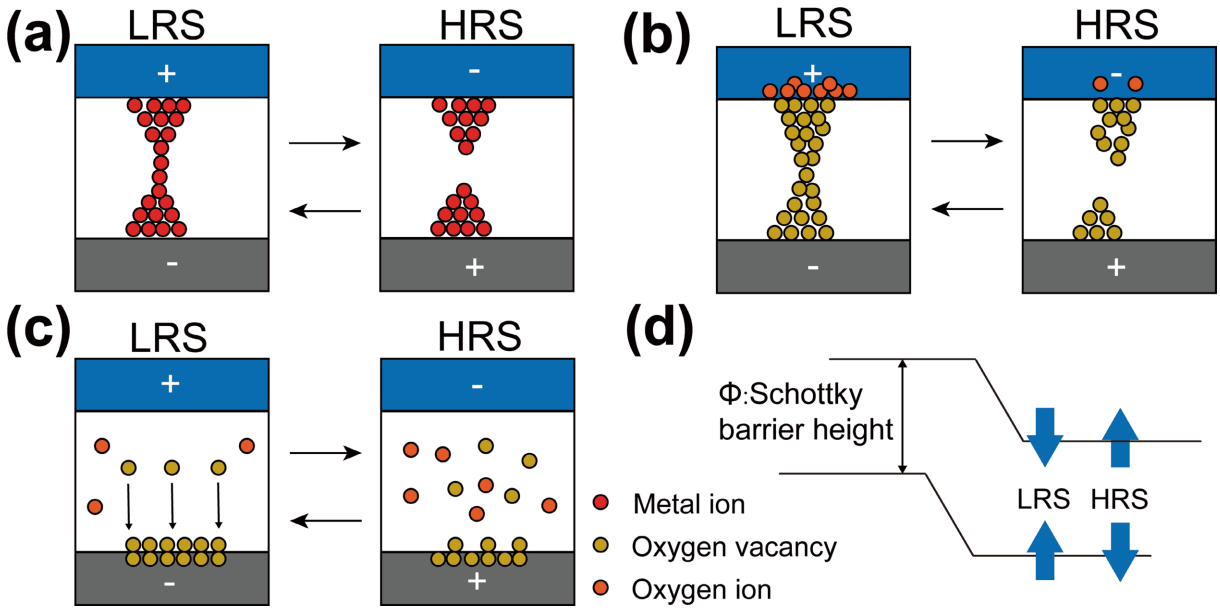

Fig. 2. (Color online) Representative ion-migration switching modes. (a) Metallic-filament formation/rupture in ECM, (b) oxygen-vacancy-channel formation/rupture in filamentary VCM, (c) interfacial ionic redistribution, and (d) Schottky-barrier modulation in interfacial VCM.

dependent plasticity (SNDP), and neural-network simulations based on its conductance weights achieved high image-recognition accuracy (Fig. 3(b))[21]. This type of composite-layer design is effective for improving filament controllability, but its uniformity still depends on the size, distribution, and interfacial contact of the introduced conductive domains. Because preferred nucleation sites alone cannot fully control cation injection and filament dissolution, interface engineering provides a complementary way to stabilize the same ECM process[29, 38].

The second strategy is interfacial, blocking-layer, or ferroelectric-layer engineering, which aims to regulate metal-ion injection, migration, and filament rupture[29, 38]. Compared with conductive-island design, this route focuses less on providing filament nucleation sites and more on slowing and stabilizing the electrochemical process. Dai *et al.*[38] inserted ferroelectric AlScN into an Ag/AlN/Pt stack to construct a trilayer device (Fig. 3(c)). The ferroelectric polarization modulated conductive-filament formation and rupture, converting sharp switching into smoother conductance evolution and enabling more accurate emulation of continuous synaptic-weight updates (Fig. 3(d))[38]. Similarly, Yoon *et al.*[29] demonstrated that an ultrathin ferroelectric $PbZr_{0.52}Ti_{0.48}O_3$ (PZT) electrolyte can control cation migration in ECM devices and support both threshold-switching neuron behavior and memory-switching synaptic behavior, depending on the active electrode. These studies show that interfacial polarization or barrier control can help bridge the gap between filamentary ECM switching and analog synaptic modulation[29, 38]. After electrical filament evolution is made more controllable, coupling ECM-type ion migration with optical or interfacial trapping processes further extends the device response beyond purely electrical weight modulation[39–43].

The third strategy extends ECM devices from electrical synapses to multimodal or optoelectronic synaptic systems. In these devices, ECM-type ion migration is coupled with photosensitive materials, interfacial trapping, or plasmonic effects. Bousoulas *et al.*[39] combined hybrid two-dimensional/three-dimensional FA_2PbI_4 perovskite with SiO_2/Pt-nanoparticle interfacial engineering to construct low-power optoelectronic memristors. The SiO_2/FA_2PbI_4 and FA_2PbI_4/Pt nanoparticle interfaces regulated ion migration, stabilized filament dynamics, and enabled optical neuromorphic functions[39]. Related Ag-nanoparticle/FA_2PbI_4 perovskite memristors also showed improved memory and optoelectronic synaptic behavior, supporting the use of nanoparticle/perovskite interfaces for improving perovskite-based ECM devices[40]. In addition, plasmonic Ag-oxide optoelectronic memristors demonstrated light-modulated synaptic plasticity and light-gated STDP, further supporting the feasibility of optical modulation in ECM-related synaptic systems[41]. These examples suggest that ECM mechanisms can be integrated with optical modulation, but their practical value still depends on whether filament dynamics, photoresponse, and device stability can be controlled simultaneously. For these optoelectronic ECM-related systems, mechanism analysis should distinguish metallic-filament evolution from interfacial trapping and photoinduced effects. *In situ* TEM/STEM, conductive AFM, EDS/EELS mapping, and ToF-SIMS can help verify metal-ion redistribution and conductive-filament formation, while wavelength- or light-intensity-dependent measurements, dark recovery tests, and transient photocurrent responses are useful for separating photogenerated-carrier effects from slower ion-migration processes. Kelvin probe force microscopy and impedance spectroscopy can further help evaluate interfacial charge trapping and barrier changes[16, 30, 39–45].

Overall, ECM-based synaptic devices are attractive because of their low operating voltage and high switching ratio[30, 36, 37]. Their main limitation is not the absence of synaptic behavior, but the difficulty of making metallic-filament evolution sufficiently gradual, reproducible, and device-uniform. Composite active layers, interfacial regulation layers, and optoelectronic hybrid designs address this problem from different directions. For analog neuromorphic hardware, the key task is to retain the strong conductance modulation of ECM while reducing the randomness of metal-ion migration and filament rupture[21, 36–38, 44].

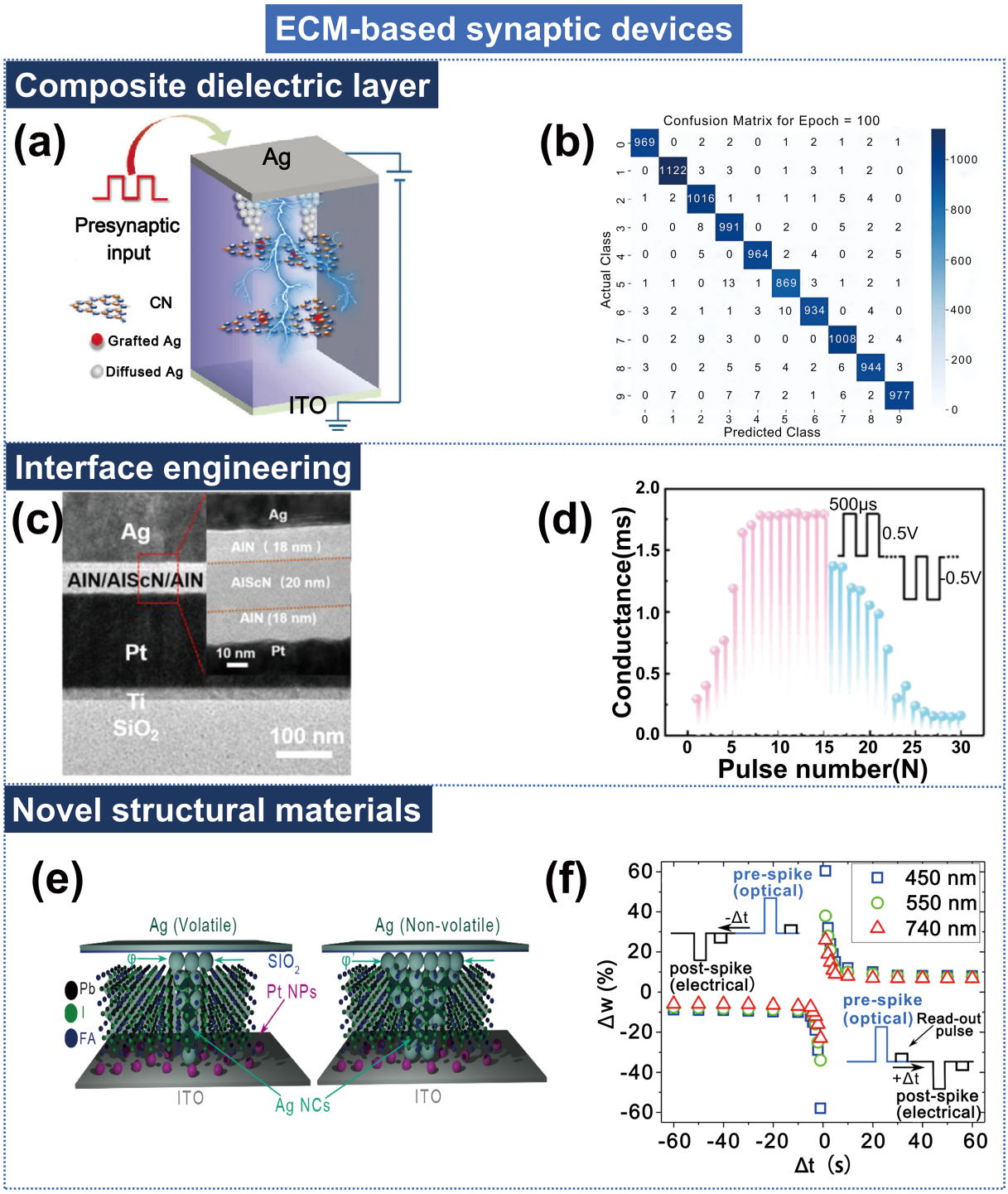

Fig. 3. (Color online) Materials and interfacial regulation strategies for ECM-based synaptic devices. (a) and (b) Conductive-island-assisted switching, (c) and (d) interface-engineered analog conductance modulation, and (e) and (f) optoelectronic/structural-material designs for multimodal synaptic behavior. Panels (a) and (b) adapted with permission from Ref. [21]. Copyright 2025, Wiley-VCH GmbH. Panels (c) and (d) adapted with permission from Ref. [38]. Copyright 2024, Elsevier Ltd. Panels (e) and (f) adapted from Ref. [39] under the terms of the Creative Commons Attribution 4.0 International License. Copyright 2025, The Authors, published by American Chemical Society.

4. VCM-based synaptic devices

VCM-based synaptic devices regulate conductance mainly through the migration and redistribution of oxygen vacancies in oxide dielectrics[23, 35, 45]. Unlike ECM devices, which rely on metal-ion migration and metallic-filament formation, VCM devices are governed by anion-related defect dynamics within the oxide or near the electrode/oxide interface[23, 35, 45]. Building on the classification established in Section 2, the following discussion distinguishes filamentary VCM, based on localized oxygen-vacancy channels, from interfacial VCM, based primarily on broader ionic redistribution and barrier modulation[23, 35, 45–47]. This distinction provides the basis for comparing their regulation strategies and synaptic characteristics.

4.1. Filamentary VCM

The synaptic behavior of filamentary VCM originates from the formation, expansion, contraction, and recombination of oxygen-vacancy channels[20, 22, 23, 35, 45]. Its advantages

include small characteristic dimensions, fast switching, and compatibility with high-density integration. Its main limitation is variability: oxygen-vacancy generation and migration are strongly influenced by initial defects, local electric fields, and Joule heating[23, 35, 45]. The central design target is therefore to make oxygen-vacancy filament evolution more controllable in position, concentration profile, and dynamic response[20, 22]. For filamentary VCM, the key mechanism is oxygen-vacancy generation, migration, aggregation, and recombination. Chemical-state and defect-distribution analyses, such as XPS, EELS, and ToF-SIMS, can provide evidence for oxygen-vacancy redistribution, while conductive AFM can map localized conductive channels. Impedance spectroscopy and temperature-dependent *I–V* measurements can further help distinguish vacancy-assisted filament conduction from electronic trapping, interface-limited transport, or possible electrode-related ion migration. Accordingly, the following discussion moves from regulating the initial vacancy distribution, to tuning defect-generation kinetics, and then to confining vacancy-migration pathways[20, 22, 32, 35, 45, 48].

One route is to predefine the spatial distribution of oxygen vacancies. Zou *et al.*[22] constructed a multilayer HfO_x memristor with a "V"-type oxygen-vacancy distribution to mimic Ca^{2+}-related modulation during synaptic transmission (Fig. 4(a)). By adjusting the applied voltage and compliance current, the device regulated different levels of voltage-controlled Ca^{2+}-like channels and enabled controlled conductive-filament formation. The hourglass-like filament formed in the middle functional layer improved filament-position control and supported fast, low-power synaptic behaviors, including STP, LTP, and STDP-like responses (Fig. 4(b))[22]. This design shows that spatially engineered vacancy profiles can reduce filament-position randomness and improve the biological relevance of conductance modulation. After the vacancy profile is spatially predesigned, repeated synaptic operation still depends on whether vacancy generation and recombination remain controllable under pulse stimulation[20, 22, 35, 45].

A second route is defect chemistry engineering, especially through elemental doping[20, 35, 45]. Mahata *et al.*[20] used plasma-induced nitrogen doping in indium gallium zinc oxide (InGaZnO) to optimize the nitrogen/oxygen ion concentration, which reduced the randomness of oxygen-vacancy generation and filament evolution. In the ITO/InGaZnO:N/ITO memristor (Fig. 4(c)), volatile and nonvolatile memory behaviors could coexist depending on the SET current compliance. The device demonstrated pulse-induced short-term memory learning, STP, paired-pulse facilitation (PPF), spike-rate-dependent plasticity (SRDP), and experience-dependent synaptic weight modification (Fig. 4(d))[20]. Compared with spatial vacancy-gradient design, doping mainly regulates the generation and recombination kinetics of oxygen vacancies, thereby improving the continuity of state evolution. In addition to tuning defect chemistry, structural confinement can further determine where the vacancy-rich channel evolves during switching[48].

Nanoparticles and multilayer composite structures provide another way to guide vacancy migration pathways[48]. Mahata *et al.*[48] embedded localized Au nanoparticles into an $HfTiO_x/TiSiO_x$ bilayer switching layer (Fig. 4(e)). The Au nanoparticles enhanced the local electric field near the $HfTiO_x$/Au-NP interface and promoted controlled filament formation, while the oxygen-vacancy-rich $TiSiO_x$ layer provided a stable conduction pathway after electroforming. This spatial confinement improved cycle-to-cycle uniformity and supported metaplasticity, STDP, pulse-width-dependent plasticity, pulse-frequency-dependent plasticity, and reservoir-computing functions (Fig. 4(f))[48]. This strategy differs from doping because it regulates where vacancy channels form rather than only how vacancies are generated.

Overall, filamentary VCM is suitable for applications requiring fast response, small device dimensions, and relatively large conductance modulation. However, its value as an analog synapse depends on whether vacancy-channel evolution can be made sufficiently reproducible[20, 22, 23, 35, 45, 48]. Vacancy-gradient design, elemental doping, and nanoparticle-assisted confinement all improve filament controllability, but they target different sources of variability: spatial defect distribution, defect-generation kinetics, and migration-pathway localization[20, 22, 48]. Further optimization should therefore focus on matching vacancy-filament control with the linearity, symmetry, and noise tolerance required by neuromorphic systems[20, 22, 23, 35, 45, 48].

4.2. Interfacial VCM

Interfacial VCM differs from filamentary VCM by replacing localized conductive-channel formation with barrier modulation at the electrode/oxide interface[46, 47]. In this mode, oxygen vacancies or interfacial charges accumulate and deplete over a broader region, changing the Schottky barrier height, barrier width, or interfacial resistance. This surface-like modulation avoids the random formation and rupture of narrow vacancy filaments and is therefore more favorable for gradual and reproducible conductance updates[46, 47]. Its limitation is that the modulation depth of the interface is finite, so the switching window is often smaller than that of filamentary devices. This change in switching region also shifts the design focus from local filament control to interface and barrier engineering[46, 47].

Ju *et al.*[46] demonstrated a $Pt/TaO_x/TiN$ memristor with neuromorphic behavior mediated by interfacial ionic coupling (Fig. 5(a)). The work-function difference between Pt and TiN produced an asymmetric Schottky barrier and self-rectifying behavior, which helped suppress sneak currents in crossbar arrays. Field-driven migration of oxygen ions in TaO_x modulated the Schottky barrier and produced volatile, gradual switching (Fig. 5(b)). The coupling between interfacial effects and ionic dynamics allowed the device to emulate synaptic plasticity, nociceptor functions such as thresholding, relaxation, and sensitization, and reservoir-computing behavior[46]. This work illustrates the advantage of interfacial VCM in combining analog conductance modulation with array-relevant rectification. The same barrier-modulation logic can also be extended to interfacial systems in which charge trapping or asymmetric contacts dominate the conductance change[46, 47].

Beyond classical oxygen-vacancy-mediated interfacial VCM, related barrier-modulated interfacial devices based on charge trapping/detrapping can also provide useful references for analog synaptic switching. Two-dimensional or heteromeltal-contact structures provide another platform for interfacial switching. Fejzaj *et al.*[47] reported a forming-free vertical MoS_2 memristor based on asymmetric metal/MoS_2 con-

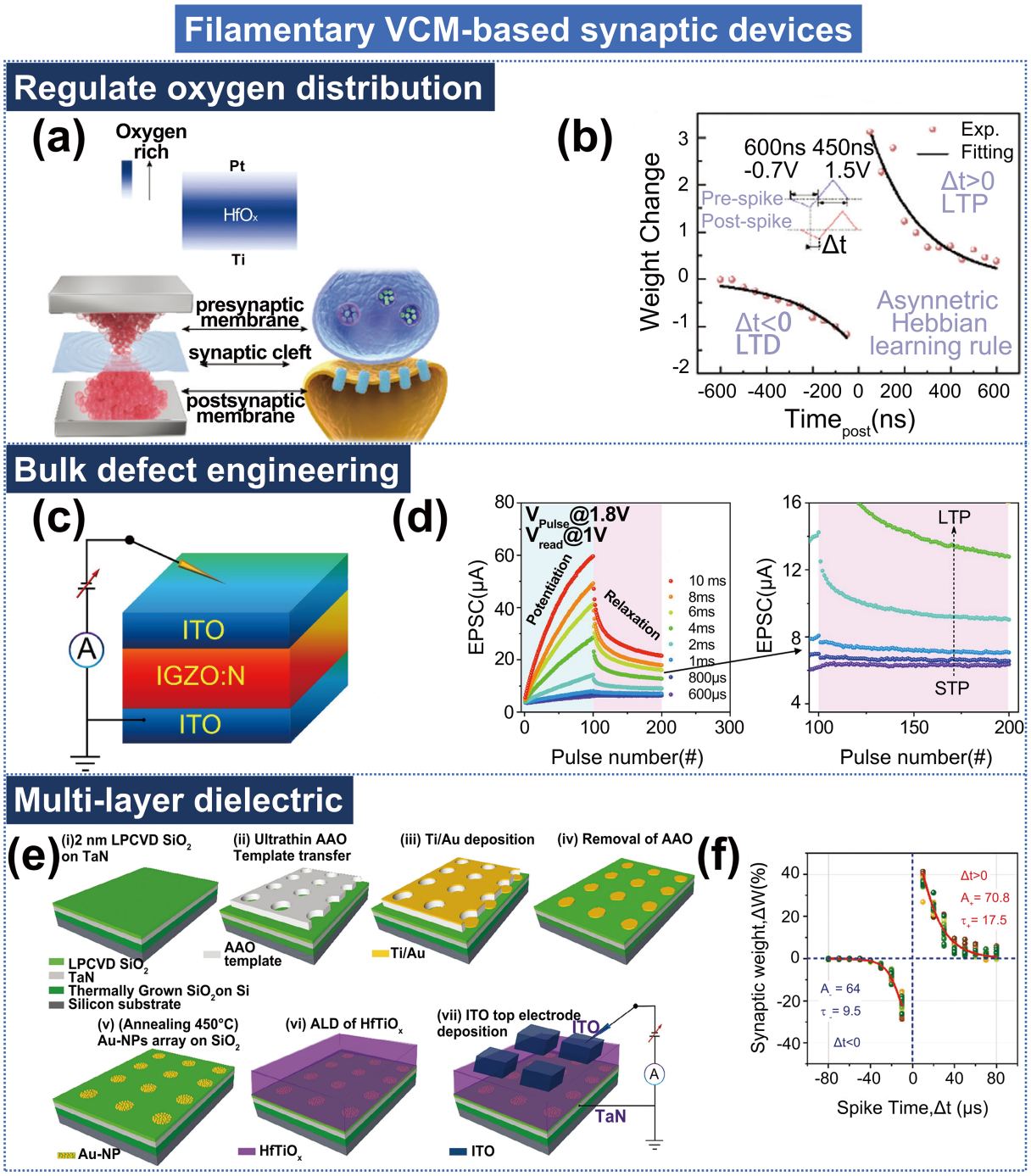

Fig. 4. (Color online) Strategies for regulating oxygen-vacancy channels in filamentary VCM synaptic devices. (a) and (b) Oxygen-vacancy-gradient design, (c) and (d) N-doping-assisted defect engineering, and (e) and (f) nanoparticle/multilayer confinement for reproducible filament evolution. Panels (a) and (b) adapted with permission from Ref. [22]. Copyright 2025, Wiley-VCH GmbH. Panels (c) and (d) adapted with permission from Ref. [20]. Copyright 2024, Elsevier Ltd. Panels (e) and (f) adapted with permission from Ref. [48]. Copyright 2025, Wiley-VCH GmbH.

tacts (Fig. 5(c)). The switching was attributed to charge trapping and detrapping at the MoS_2/metal interface, which modulated the Schottky barrier rather than forming metallic or oxygen-vacancy filaments. The device exhibited gradual resistive switching and synaptic functions, including potentiation, depression, and spike-amplitude-dependent plasticity (Fig. 5(d))[47]. This type of device further shows that interfacial barrier control can support analog synaptic behavior without relying on localized filament formation. Because barrier modulation is sensitive to the defect population near the interface, doping provides a materials-level route to tune this interfacial process[49].

Doping can also be used to tune interfacial VCM by regulating the concentration and distribution of oxygen vacancies. Su *et al.*[49] used Sn doping in β-Ga_2O_3 memristors to tune oxygen-vacancy concentration, enabling sharp negative differential resistance (NDR), analog resistive switching, and artificial synaptic functions in the same device (Fig. 5(e)). The optimized device reproduced paired-pulse facilitation, with the PPF index decreasing as the pulse interval increased (Fig. 5(f))[49]. This example shows that defect engineering in interfacial devices can be used not only to tune switching behavior, but also to couple synaptic plasticity with additional nonlinear electrical characteristics. In interfacial VCM systems, gradual conductance modulation may originate from oxygen-vacancy redistribution, charge trapping/detrapping,

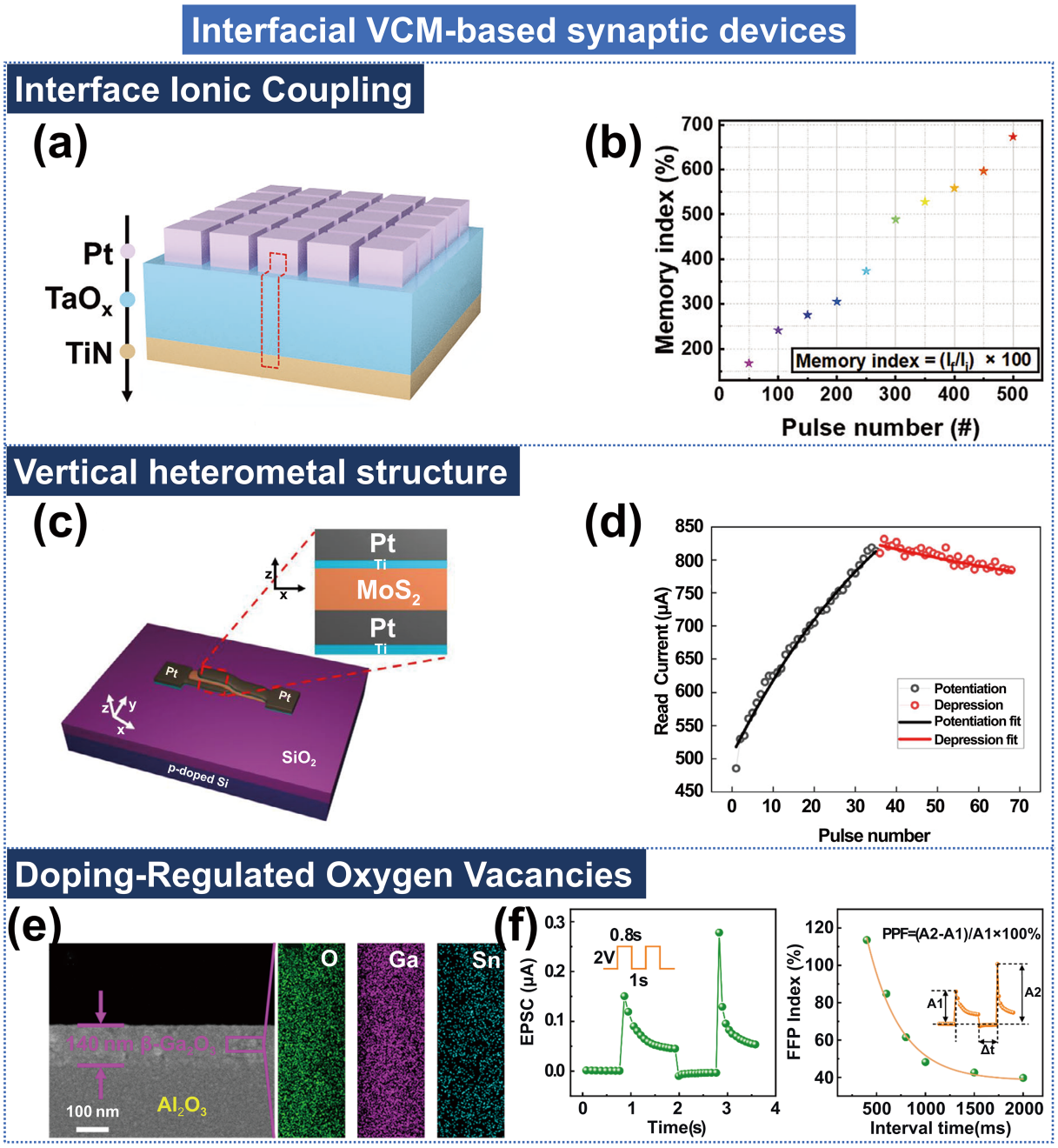

Fig. 5. (Color online) Device-design strategies for interfacial VCM synaptic devices. (a) and (b) Interfacial ionic coupling, (c) and (d) heterometal-contact barrier modulation, and (e) and (f) doping-regulated oxygen-vacancy engineering. Panels (a) and (b) adapted with permission from Ref. [46]. Copyright 2025, Wiley-VCH GmbH. Panels (c) and (d) adapted from Ref. [47] under the terms of the Creative Commons Attribution 4.0 International License. Copyright 2026, The Author(s), published by Wiley-VCH GmbH. Panels (e) and (f) adapted with permission from Ref. [49]. Copyright 2025, American Chemical Society.

asymmetric contact barriers, or their combined effects. XPS depth profiling, EELS, and ToF-SIMS can provide evidence for oxygen-vacancy accumulation or redistribution near the interface. Kelvin probe force microscopy, impedance spectroscopy, and temperature-dependent *I–V* fitting can be used to evaluate Schottky-barrier modulation and transport mechanisms. Transient current measurements and retention/recovery tests are helpful for identifying charge-trapping-related contributions, while conductive AFM and device-area-dependent measurements can help exclude localized filamentary switching[46, 47, 49].

Overall, interfacial VCM is better suited than filamentary VCM for continuous analog weight updates, device uniformity, and stable multilevel conductance control[46, 47]. Its weakness lies in the limited switching window and possible retention constraints caused by the finite range of barrier modulation. Thus, future interfacial VCM design should not simply increase defect concentration, but coordinate interfacial defect density, electrode work function, barrier profile, and read/write voltage window[46, 47, 49]. In this sense, filamentary and interfacial VCM represent two complementary routes: the former prioritizes localized high-contrast switching and scaling potential, whereas the latter prioritizes smoother conductance evolution and array-level stability.

5. Integration of ECM and VCM

Single-mechanism ECM and VCM devices each empha-

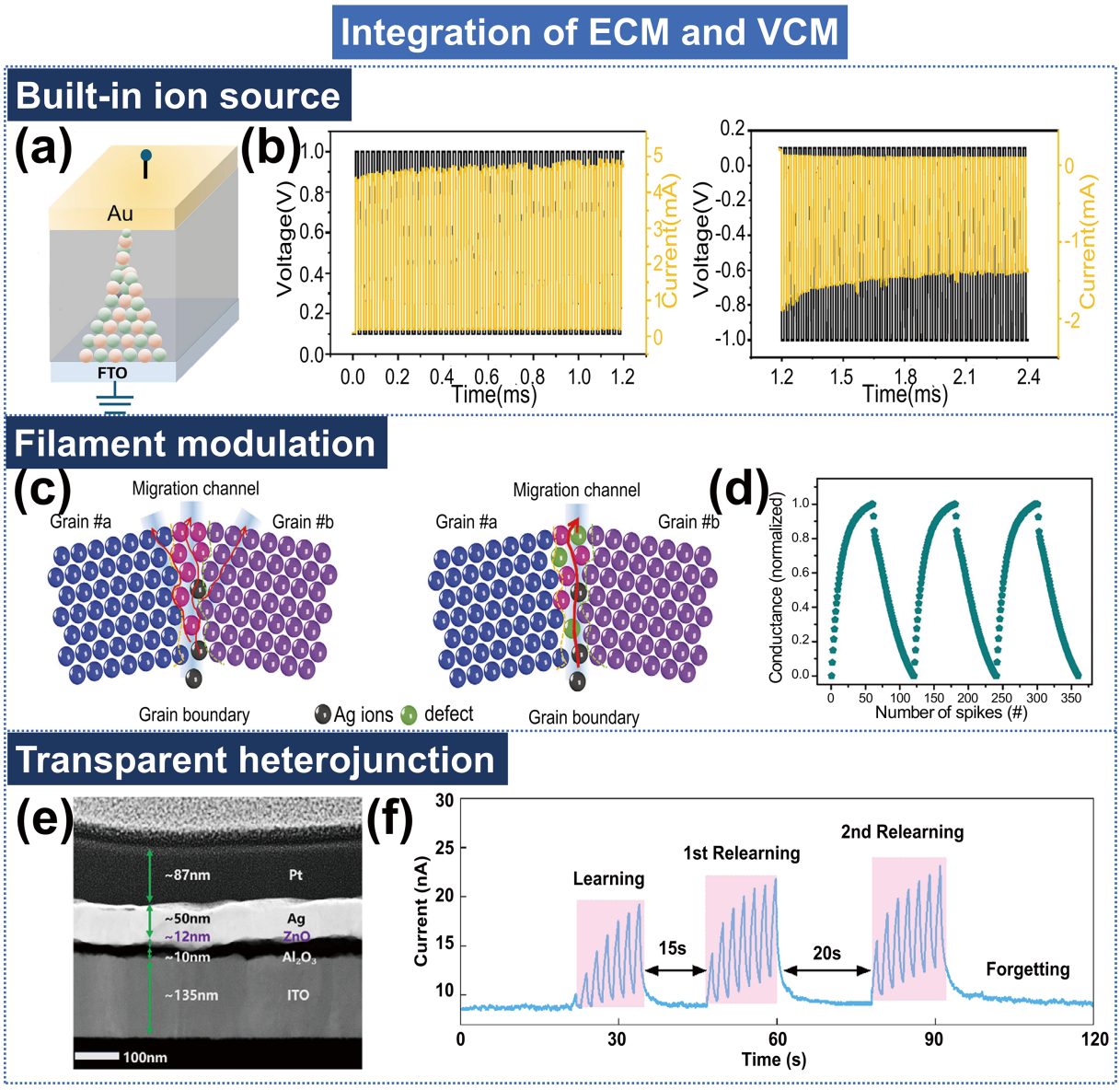

Fig. 6. (Color online) Hybrid ECM-VCM device-design strategies. (a) and (b) Built-in ionic sources, (c) and (d) defect-rich filament modulation, and (e) and (f) transparent heterojunctions for electrical/optical synaptic responses. Panels (a) and (b) adapted with permission from Ref. [50]. Copyright 2025, Wiley-VCH GmbH. Panels (c) and (d) adapted from Ref. [51] under the terms of the Creative Commons Attribution 4.0 International License. Copyright 2024, The Authors, published by Wiley-VCH GmbH. Panels (e) and (f) adapted with permission from Ref. [55]. Copyright 2026, Wiley-VCH GmbH.

size different advantages, but none of them alone can easily satisfy all requirements of synaptic hardware. ECM devices offer low-voltage operation and high switching ratios, but stochastic metallic-filament evolution limits stable analog weight updating[30, 36, 37]. Filamentary VCM devices are favorable for fast switching and scaling, but oxygen-vacancy channels also suffer from variability[23, 35, 45]. Interfacial VCM devices provide smoother conductance evolution, yet their switching window is usually limited[33, 34, 46–48]. Hybrid ECM-VCM design is therefore not merely a combination of two switching mechanisms; it is a strategy to redistribute the roles of metal ions, oxygen vacancies, interfaces, and defects so that switching window, analog tunability, stability, and multifunctionality can be balanced within one device[36, 37, 50–52]. The following examples therefore follow how ECM and VCM contributions are coupled within the active layer, the defect-rich migration medium, and the optoelectronic heterointerface[50–55].

One integration route is to construct active materials containing more than one ionic source. In this design, metallic ions provide strong filamentary conduction, while oxygen-vacancy regulation is used to tune the stability and continuity of switching[50]. Mukherjee *et al.*[50] synthesized $AgHfO_{3-x}$ perovskite, in which Ag atoms embedded in the lattice acted as a stable source for conductive-filament formation (Fig. 6(a)). By deliberately introducing oxygen during deposition, the oxygen-vacancy concentration was regulated and the randomness of vacancy-related switching was reduced, making Ag ions the dominant filament-forming species. This design improved switching uniformity and cycling endurance and enabled more precise conductance modulation during synaptic potentiation and depression (Fig. 6(b))[50]. The key value of this approach is that the active layer itself defines the relative contribution of metal-ion and oxygen-vacancy processes. For such built-in-ion-source systems, the relative contributions of lattice-sourced Ag filaments and oxygen-vacancy regu-

lation should be carefully distinguished. Cross-sectional TEM/STEM and EDS/EELS mapping can directly examine filament morphology and Ag distribution, while XPS and ToF-SIMS can track oxygen-vacancy-related chemical changes. Comparative electrical measurements under different oxygen contents, electrode configurations, or temperature conditions can further clarify whether switching is dominated by Ag-filament formation or vacancy-assisted modulation. After defining the ionic source in the active material, regulating the migration medium becomes another way to stabilize hybrid switching[50].

A second route is to engineer defect-rich oxide media in which metallic-cation migration and oxygen-vacancy-related defect migration are synergistically regulated[51]. Wang *et al.*[51] constructed an Ag/SnO_2/p^{++}-Si ion-based memristor using oxygen-rich SnO_2 nanoflowers as the switching medium (Fig. 6(c)). In this device, grain-boundary defects in SnO_2 provided preferential one-dimensional migration channels that confined Ag^+ diffusion and reduced random ion migration. Meanwhile, the interplay between Ag^+ and oxygen vacancies effectively modulated the formation and rupture of conductive filaments, leading to a high on/off ratio, long retention, reduced switching variability, and stable analog conductance modulation[51]. The device further exhibited multiple synaptic functions, including PPF, LTP/LTD, and STDP, and showed nearly symmetric and linear conductance updating under pulse programming (Fig. 6(d))[51]. Compared with single-ion switching, this defect-rich oxide strategy uses microstructural channels and vacancy-related defects to improve the controllability of metallic filament evolution. In defect-rich oxide media such as SnO_2 nanoflowers, Ag+ migration, oxygen-vacancy transport, and grain-boundary-assisted conduction may be coupled. Elemental mapping and ToF-SIMS can trace Ag redistribution, while XPS/EELS analysis can evaluate oxygen-vacancy-related defect states. High-resolution TEM and conductive AFM are useful for correlating grain-boundary structures with local current paths. Impedance spectroscopy and temperature-dependent transport measurements can further help separate ionic-defect-assisted conduction from metallic-filament-dominated switching. On this basis, photoresponsive heterostructures add optical excitation to the coupled ionic processes, enabling sensory-memory functions[51].

A third route introduces photoresponsive materials into hybrid ECM-VCM structures to achieve electrical/optical dual modulation[53–55]. Zhang *et al.*[55] constructed an Ag/ZnO/Al_2O_3/ITO transparent heterojunction (Fig. 6(e)). Under electrical stimulation, Ag^+ ions and oxygen vacancies cooperatively contributed to conductive-filament formation and rupture, mimicking dynamic neuronal behavior[55]. Under optical stimulation, ultraviolet absorption in ZnO generated carriers that were trapped by defects in Al_2O_3, thereby producing synaptic plasticity and memory behavior (Fig. 6(f))[55]. This design extends hybrid memristors from purely electrical synapses toward sensory-memory devices. However, in such systems, metal-ion migration, oxygen-vacancy modulation, photogenerated carriers, and charge trapping may all contribute to resistance changes, so mechanism separation becomes especially important[53–55]. In transparent heterojunctions such as Ag/ZnO/Al_2O_3/ITO, metal-ion migration, oxygen-vacancy redistribution, photogenerated-carrier transport, and charge trapping may coexist. *In situ* TEM/STEM, conductive AFM, EDS/EELS mapping, and ToF-SIMS can be used to identify Ag-related filament formation and ionic redistribution. XPS, EELS, and temperature-dependent *I*–*V* measurements can help evaluate oxygen-vacancy modulation and defect-assisted transport. Wavelength-dependent illumination, light-intensity-dependent measurements, dark recovery tests, and transient photocurrent responses are particularly useful for distinguishing photogenerated carriers and charge trapping from slower ionic migration. Combining these analyses is necessary to determine whether the synaptic response is dominated by ECM-type filaments, VCM-type defect migration, interfacial trapping, photoinduced carriers, or their coupled effects[42, 43, 53–55].

Overall, ECM-VCM integration provides a practical route to combine the large switching window of ECM, the defect-regulation capability of VCM, and additional functions from heterostructures or photoresponsive materials[36, 37, 50–52, 55]. Its main contribution is not that hybrid devices are universally superior, but that they offer more degrees of freedom in ionic-source, defect, and interface design. At the same time, hybrid devices introduce new challenges in mechanism identification, parameter control, interface reliability, and array-level scalability[23, 25, 36, 51, 52]. Future work should therefore move from simply demonstrating coupled functions toward clarifying dominant switching processes, stabilizing device-to-device performance, and verifying whether the proposed hybrid mechanisms remain effective in integrated neuromorphic arrays. Moreover, for flexible or wearable implementations, mechanical deformation such as repeated bending or tensile strain can disturb ion-migration pathways and electrode/dielectric interfaces. Mechanical robustness must therefore be assessed alongside electrical performance to ensure that hybrid switching mechanisms remain reliable under realistic operating conditions[52].

6. Conclusion

Ion-migration memristive synaptic devices should be assessed by how controllably their ionic processes translate into synaptic-weight modulation. ECM devices are advantageous for low-voltage operation and large conductance modulation, but their main bottleneck remains the stochastic formation and rupture of metallic filaments[30, 36, 56]. Filamentary VCM devices offer fast switching and high-density scaling potential, yet their synaptic performance is limited by fluctuations in oxygen-vacancy channel evolution[23, 35, 45, 56]. Interfacial VCM devices provide smoother and more stable analog conductance modulation through barrier regulation, although their switching window is usually narrower[46, 47, 56]. Hybrid ECM-VCM and optoelectronic devices expand the design space by coupling metal-ion migration, oxygen-vacancy redistribution, charge trapping, and interfacial modulation, but this coupling also increases the difficulty of identifying the dominant switching mechanism and maintaining long-term device stability[36, 37, 50–53, 56, 57]. The practical deployment of these devices, however, demands that their switching stability be examined under the thermal and mechanical constraints of real-world operation. ECM devices suffer from Joule-heat-driven filament coarsening that exacerbates cycle-to-cycle variability; filamentary VCM devices must cope with local heating that accelerates stochastic vacancy redistribu-

tion; interfacial VCM devices may experience temperature- or strain-induced barrier shifts that narrow the already limited modulation window; and hybrid ECM-VCM designs face compounded reliability concerns as multiple ionic species respond differently to thermal activation or mechanical deformation. Future research must therefore move beyond demonstrations of synaptic functions such as STP, LTP, and STDP, and directly integrate thermal and mechanical reliability into the design loop. Stability evaluation should be coupled with the dominant switching mechanism—for example, through variable-temperature retention/endurance tests, thermal cycling, and, where flexible device structures are involved, cyclic bending or stretching tests. Only by ensuring predictable ion migration under both electrical and environmental stress can ECM, filamentary VCM, interfacial VCM, and hybrid devices serve as reliable building blocks for scalable, sensory-interactive neuromorphic hardware[56–63].

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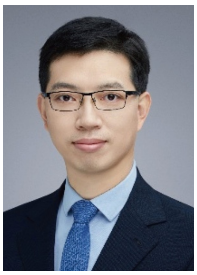

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