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Oxide-based electrolyte-gated transistors: an emerging electrochemical platform for iontronic neuromorphics

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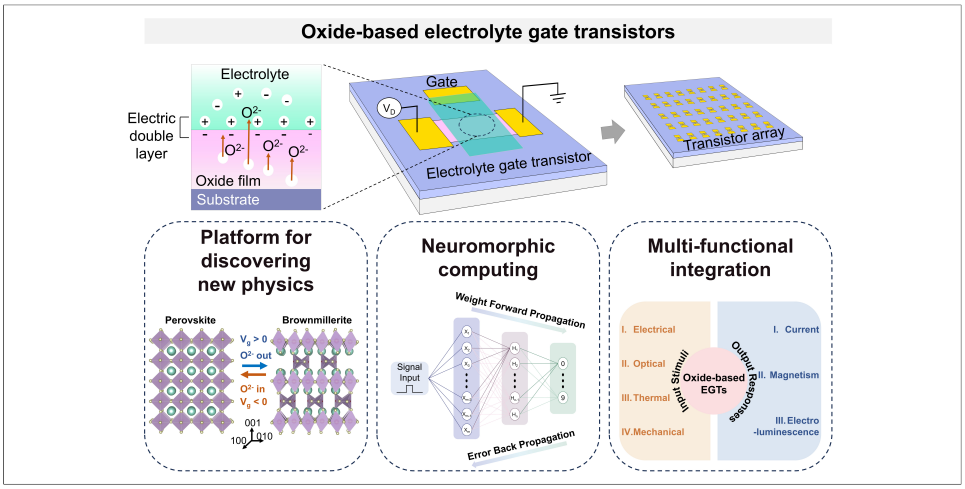

Electrolyte-gated transistors (EGTs) use an electrically insulating but ionically conductive electrolyte (e.g., ionic liquid, polymer electrolyte, ion gel) to induce charge in the transistor channel via an electric field effect. Under the gate bias, mobile ions accumulate at the electrolyte/channel interface, induce carriers in the channel, and form an electric double layer (EDL) [Figure 1A]. Because this EDL is essentially a nanometer-thin parallel-plate capacitor, it can induce a large specific capacitance (10-100 μF cm^{-2})[1], corresponding to induced charge carrier densities of 10^{14}-10^{15} cm^{-2} at a gate voltage (V_g) of only a few volts, exceeding conventional gate dielectrics by 1-2 orders of magnitude. This makes electrolyte gating especially effective in low-voltage and extreme modulations of material properties[2,3], providing access to new physics phenomena in two-dimensional crystals[1], metal oxides[4], and organic semiconductors[5], as well as wide applications in magnetoionics and optoelectronics owing to ion-electron coupling. In addition, electrolyte gating dynamics emulate ion transport in biological neural systems, indicating their potential for realizing low-energy neuromorphic computing.

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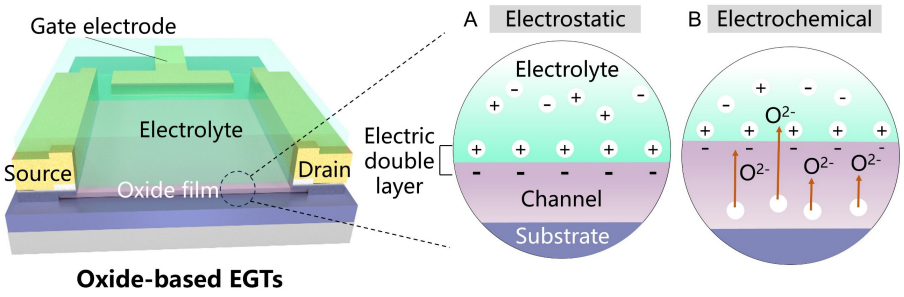

Figure 1. Device schematic of oxide-based EGTs, highlighting the difference between electrostatic (A) and electrochemical (B) gating mechanisms. EGT: Electrolyte-gated transistor.

ELECTRIC-DOUBLE-LAYER GATING OF METAL OXIDES

Metal oxides exhibit great chemical versatility and diverse functionalities, and they are more mechanically and thermally compatible with complementary metal-oxide-semiconductor (CMOS) fabrication than two-dimensional crystals and organic materials. Moreover, metal oxides typically possess complex electronic phase diagrams as a function of doping with dense boundaries between different electronic states, and thus are ideal candidates for electrically regulating insulator-metal transitions, magnetic order, superconductivity, ferroelectricity, and catalytic activity via an electric field effect[2,4]. However, accessing these new regimes requires induced carrier densities of 10^{14}-10^{15} cm^{-2} to dope substantial fractions of an electron or hole per unit cell[1,2]. Therefore, since the early 21st century, EDL gating has been applied to metal oxides to explore doping-induced electronic phase transitions. Notable discoveries include the realization of superconductivity in $KTaO_3$ [Figure 2A and B][6], the doping-induced transitions from insulator to superconductor in $SrTiO_3$[7], and the insulator-metal transition in VO_2 [Figure 2C][8]. These unprecedented achievements demonstrate the use of electrolytic gating to obtain the precise property control of metal oxides.

ELECTROCHEMICAL GATING OF METAL OXIDES

It has gradually become clear that the true gating mechanism in metal oxides is not always purely electrostatic, as electrochemical responses can also occur [Figure 1B]. Oxide-based electrochemical EGTs typically involve O^{2-}/vacancy formation in oxides or the voltage-driven permeation of H^+ and Li^+, which can be reversibly extracted from or injected into the oxide channel under the gate bias. The electrochemical gating mechanism in VO_2 was first investigated in 2013, and it was confirmed by the valence changes of V and sophisticated ^{18}O tracer experiments[9]. Additional electrochemical mechanisms have been more recently reported in metal oxides, including $SrTiO_3$[10], TiO_2[11], $SrCoO_{3-\delta}$[12,13], $YBa_2Cu_3O_{7-x}$[14], $La_{1-x}Sr_xCoO_{3-\delta}$[15-18], and $SrFeO_{3-\delta}$[19,20]. These new discoveries benefit from advances in *in situ* probes such as *operando* synchrotron X-ray diffraction[12,21], X-ray absorption spectroscopy[14], neutron reflectometry[22], *in situ* environmental transmission electron microscopy (TEM)[13], and *operando*-Fourier transform infrared spectroscopy[16]. However, device speed, endurance, structural evolution, and the diffusion coefficients of vacancies must be further investigated, as well as which oxide types and conditions lead to electrochemical or electrostatic responses during electrolyte gating.

Nonetheless, reversible and extreme property modulations have recently been demonstrated in numerous oxides, especially those with topotactic transformations, indicating opportunities for novel electronic applications. For example, the electrochemical gating of $SrCoO_{3-\delta}$[12,13] and $La_{1-x}Sr_xCoO_{3-\delta}$[15-18,23,24] has attracted much attention because these cobaltites have extremely low enthalpies of formation for oxygen vacancies, ordered oxygen-vacancy channels, and distinct ground states between phases. Room-temperature, reversible, and tri-phase electrochemical switching has been reported between perovskite $SrCoO_{3-\delta}$ (a ferromagnetic metal), the oxygen-vacancy-ordered brownmillerite $SrCoO_{2.5}$ (an antiferromagnetic insulator), and the newly

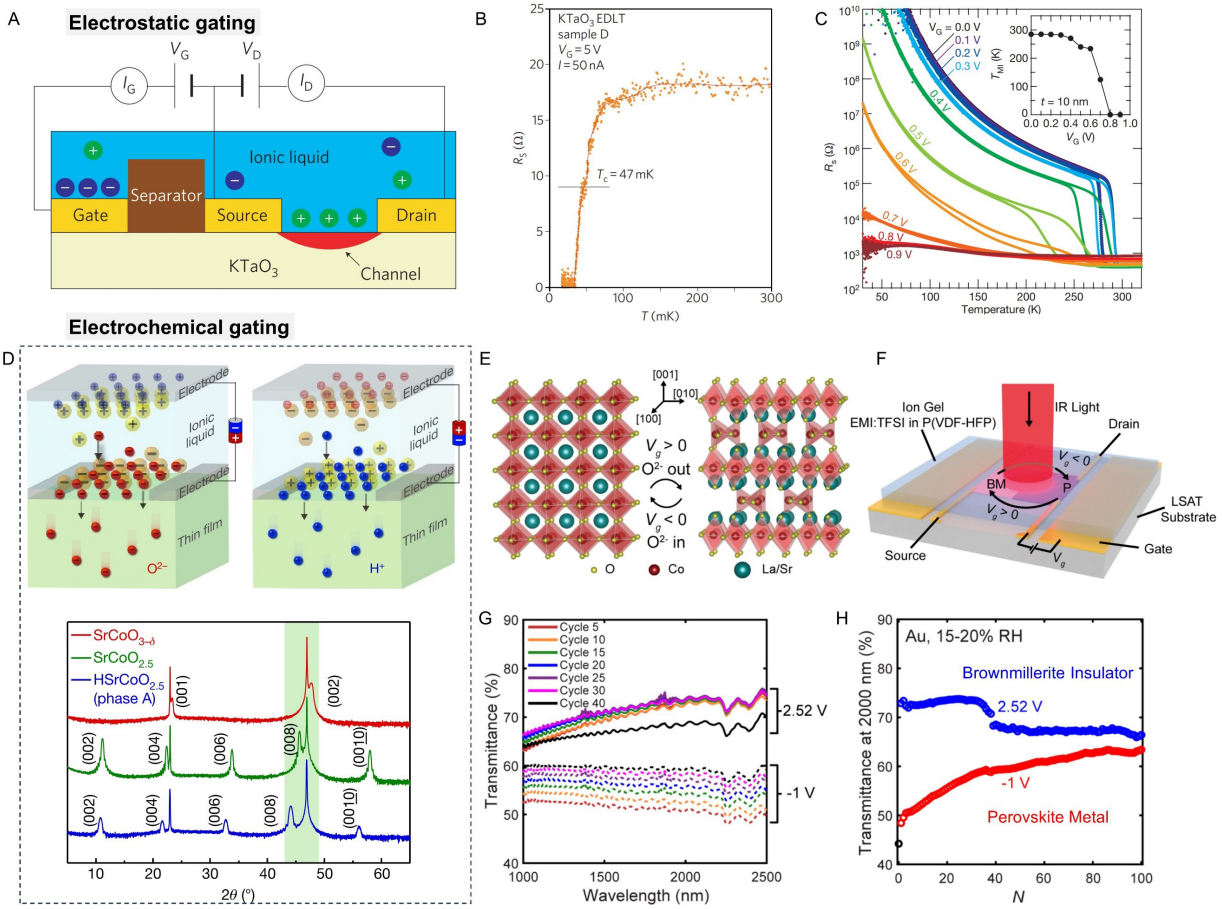

Figure 2. Examples of the electrostatic and electrochemical gating of oxides. (A and B) Ionic-liquid-gated $KTaO_3$ device designed to achieve superconductivity. Reprinted with permission. Copyright 2011, Springer Nature[6]; (C) Electrostatic-gating-induced insulator-metal transition in VO_2. Reprinted with permission. Copyright 2012, Springer Nature[8]; (D) Electrochemical-gating-induced phase transitions between $SrCoO_3$, $SrCoO_{2.5}$, and $HSrCoO_{2.5}$, as indicated by the peak shift in the X-ray diffraction results. Reprinted with permission. Copyright 2017, Springer Nature[12]; (E-H) Gated phase transitions between $La_{0.5}Sr_{0.5}CoO_3$ and $La_{0.5}Sr_{0.5}CoO_{2.5}$, including the structural schematic (E), *in situ* Fourier transform infrared spectroscopy setup (F), repeating cycles of phase transitions (G), and ON/OFF ratios of the transmittance (H). Reprinted with permission. Copyright 2025, ACS[16]. EDLT: electric double-layer transistor; EMI:TFSI: 1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)amide; P(VDF-HEP): poly(vinylidene fluoride-co-hexafluoropropylene); IR: infrared; RH: relative humidity.

discovered $HSrCoO_{2.5}$ (a weakly ferromagnetic insulator)[12]. The different electric and magnetic ground states of these phases enable the gated control of magnetic and optical properties without significantly altering the crystal structures. As such new oxide material systems are further developed, oxide-based electrochemical EGTs are evolving into multifunctional devices.

EMERGENCE OF OXIDE-BASED ELECTROCHEMICAL EGTS FOR NEUROMORPHIC COMPUTING

Because oxide electrochemical EGTs typically operate at kilohertz frequencies and possess signal transmission behaviors similar to biological nervous systems, these devices are well-suited for replicating neural ion dynamics and biological connections between neurons and synapses[25,26]. In fact, oxide electrochemical EGTs have been suggested as three-terminal, nonvolatile synaptic devices for creating artificial neural networks (ANNs), which can enable multiply-and-accumulate operations in deep-learning systems and advanced spiking neural networks (SNNs). Specifically, oxide EGTs provide decoupled write/read lines, in which the gate drives ions (e.g., O^{2-}[27-33], Li^+[34-41], H^+[42-46]) into the oxide channel to mimic

the migration of neural transmitters between pre- and postsynaptic neurons, and the channel conductance corresponds to synaptic weight. Compared to memristor-based two-terminal artificial synapses, this decoupled write/read operation improves programming energy, synaptic weight precision, and the linearity of weight updates, which are critical components of the computing accuracy of ANNs. Additionally, oxide EGTs generally exhibit high ON/OFF ratios with an ultralow OFF current, good CMOS compatibility, and better chemical/thermal stability than many organic electrochemical transistors (OECTs) or electrolyte-gated two-dimensional materials, thereby reducing the challenges associated with fabricating large-scale device arrays.

Important advancements in oxide-based electrochemical EGTs for neuromorphic computing are summarized in Figure 3. Differing from early electrostatic attempts using indium-zinc-oxide (IZO) channels[47,48], $SmNiO_3$-based electrochemical EGTs first demonstrated nonvolatility and a learning function in ANNs due to oxygen-vacancy formation in their channel[27]. Other EGTs that use oxygen vacancies, such as ZnO[28], ITO[29], WO_3[30,31], and $SrCoO_x$[32,33], have also demonstrated a low programming energy, long-term potentiation and depression, and spike-timing-dependent plasticity and logics. Generally, oxygen-vacancy-based EGTs excel in terms of their nonvolatility (on the order of days to years), wide conductance modulation range, and CMOS compatibility. However, they also typically suffer from relatively large driving voltages and low speeds due to the oxygen migration energy barrier and limited oxygen diffusivity found in many oxides. Moreover, injecting oxygen ions back into oxides often involves water splitting[16], which requires careful consideration of electrolyte selection and device endurance. Large-scale device arrays are needed to further realize advanced neural functions, as current demonstrations typically rely on using ionic liquids in a single device.

Li^+ ion intercalation has also been applied in numerous oxide EGTs such as $Li_{1-x}CoO_2$[34], Li_xTiO_2[35], MoO_3[36], WO_{3-x}[37], Nb_2O_5[38-40], and Li_xInO_x[41], and it has demonstrated potential for use in neuromorphic devices. Li *et al.*[38] investigated Nb_2O_5 and demonstrated the first instance of a 32 × 32 EGT array, which exhibited quasi-linear updating, good endurance (10^6) and retention, a 100 ns switching speed, and a low readout current. Owing to their intermediate ion-migration barrier, oxide EGTs based on Li^+ intercalation can achieve a balanced performance between speed (microsecond-level response), reversibility, and nonvolatility (on the order of days). However, Li^+-based electrolytes (e.g., LiPON[34], Li_3PO_4[37], and Li_xSiO_2[38,39]) typically require additional encapsulation to avoid water and cannot be easily integrated into CMOS fabrication processes. Oxides that allow Li^+ intercalation are also relatively rare compared to other mechanisms, and the issue of lattice-distortion-induced current drift due to repeated inter-/deintercalation requires more investigation.

In recent years, high-*k* oxides such as AlO_x[42], ZrO_x[43], ScO_x[44], and HfO_x[45] have also been adopted as proton electrolytes that are compatible with CMOS technologies. For example, Cui *et al.*[43] proposed H_xWO_3/ZrO_2 electrochemical EGTs, which modulate multilevel conductance by converting H^+ between the H_xWO_3 channel and gate through the ZrO_2 electrolyte, demonstrating an operating frequency of 1 MHz and endurance exceeding 10^8 cycles. Additionally, introducing a proton reservoir (e.g., MgO) between the channel and electrolyte can suppress interfacial defects and improve device durability[45]. Generally, the proton intercalation mechanism demonstrates promising high-speed, reversibility, and low-voltage characteristics, partially due to the small ionic radius and fast migration of protons. Future efforts may focus on improving the state retention, ON/OFF ratios, and expanding the choice of materials for these systems. Overall, these different oxide-EGT mechanisms increase the number of available materials for building EGT-based ANNs and larger-scale device arrays, providing a solid foundation for realizing all-solid-state neuromorphic computing hardware.

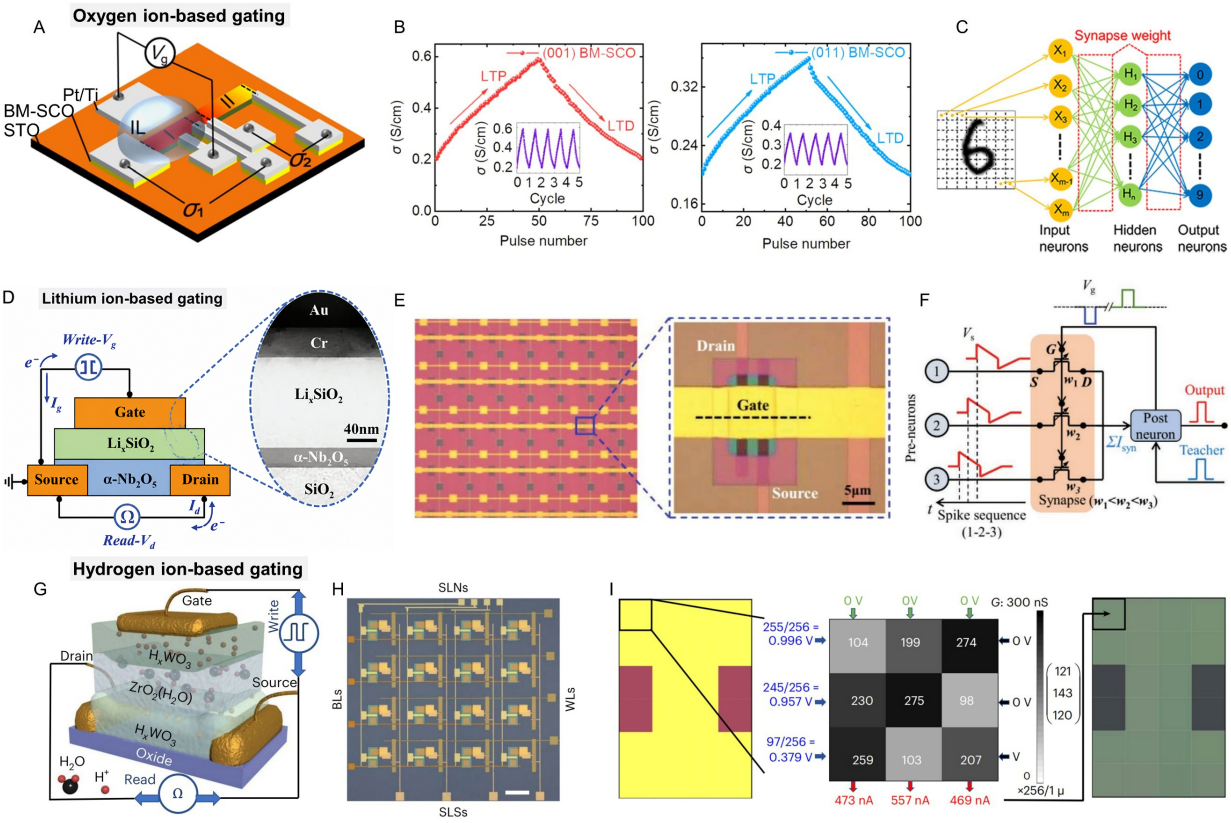

Figure 3. Examples of oxide-based electrochemical EGTs for neuromorphic computing. (A-C) Oxygen-ion-based gating in Sr_xCoO_3, including the device schematic (A), long-term potentiation and long-term depression (B), and ANN for image recognition (C). Reprinted with permission. Copyright 2023, WILEY-VCH[33]; (D-F) Lithium-ion-based gating in Nb_2O_5: device schematic (D), EGT arrays (E), and a simple 3 × 1 SNN (F). Reprinted with permission. Copyright 2020, WILEY-VCH[38]; (G-I) Hydrogen ion-based gating in H_xWO_3, showing a device schematic (G), device arrays (H), and the color transformation of a letter "I" by the arrays (I). Reprinted with permission. Copyright 2023, Springer Nature[43]. BM-SCO: brownmillerite $SrCoO_{2.5}$; STO: $SrTiO_3$; LTP: long-term potentiation; LTD: long-term depression; BL: bit line; SLN: source line for weight update; WL: word line; SLS: source line for weight sum; EGT: electrolyte-gated transistor; ANN: artificial neural network; SNN: spiking neural network.

PROSPECTS, CHALLENGES, AND POTENTIAL APPLICATIONS

Oxide-based electrochemical EGTs offer diverse device mechanisms based on the incorporation of various ions such as O^{2-}[27-33], Li^+[34-39], and H^+[42-46]. Moreover, they typically provide stronger modulation of electronic and magnetic properties as well as the realization of nonvolatility compared to electrostatic gating or conventional CMOS devices, thus offering numerous advantages for iontronic neuromorphics. First, oxide EGTs can leverage gating-induced phase change to achieve power-efficient multifunctionality and nonvolatile computing, whose capabilities require the use of many conventional transistors in circuits. Second, oxides that exhibit good thermal stability are more compatible with CMOS technology compared to organic or two-dimensional materials. Third, compared to two-terminal oxide memristors, the device geometry of oxide EGTs decouples the read/write operations, enhancing synaptic computing precision and reducing static power consumption, and these systems can further mimic neuron behaviors with multiterminal inputs. These advantages demonstrate the potential of oxide EGTs for achieving stable, accurate, energy-efficient, and large-scale neuromorphic computing devices that are compatible with CMOS technology.

Although promising results have been achieved for oxide EGTs, issues such as device scalability, speed, and endurance must be addressed. The most critical challenges are fabricating large-scale device arrays (e.g., arrays larger than 32 × 32[38]) and demonstrating advanced neural functions. The integration of inorganic

solid electrolytes is necessary to achieve these goals, as existing ionic liquids or polymer electrolytes cause incompatibility with CMOS fabrication technology (e.g., instability at a CMOS back-end-of-line temperature of ~ 400 °C[49]). Fortunately, extensive knowledge from batteries and ionic conductors research may be used to overcome these issues. Another challenge concerns the slow switching speed of many electrochemical oxide EGTs, particularly those featuring gated phase changes (on the order of seconds)[15]. Although oxide EGTs are typically faster than OECTs, megahertz to gigahertz frequencies are highly desirable in neuromorphic computing. The speed of these systems can be improved by improving the ion diffusivity, ionic pathway, gate geometry, and device miniaturization of the oxide. However, it should be noted that there is often a tradeoff between device speed and nonvolatility. Finally, ion diffusion, moisture, and lattice distortion due to repeated ion intercalation can lead to device instability, negatively affecting device reversibility and endurance. Strategies such as engineering ordered ionic pathways in the oxides, device packaging, and ion reservoirs of these systems may be used to overcome problems related to structural instability, external interference, and the source/destination of ions.

By combining the beneficial physical properties of oxides with nonvolatile control, oxide EGTs have the potential to achieve multifunctionality by integrating sensing, memory, and computing operations in a single device. For example, a recent study of $BaSnO_3$-based EGTs used electrolyte-gated proton injection to achieve short- and long-term plasticity[46]. Ultraviolet (UV) light was used to modulate oxygen vacancies to realize the synergistic control of the current and sensing capability, demonstrating the achievement of optical sensing, nonvolatility, and neuromorphic computing. With the advancement of the gated phase switching of oxides, further research is required to explore its applications in multimodal optoelectronics, magnetoionics, biological and chemical identification, and in/near-sensor nonvolatile computing, so as to effectively mimic biological multisensory systems and neuromorphic architectures.

Overall, oxide electrochemical EGTs show great promise due to their significant application potential in emulating biological nervous systems and achieving the power-efficient voltage control of various material functions. In the future, oxide electrochemical EGTs may enable the integration of sensing, memory, and computing operations into a single optoelectronic transistor for multimodal recognition. Combined with their inherent nonvolatility and compatibility with CMOS technology, oxide electrochemical EGTs are promising hardware candidates for next-generation bioinspired computing systems.

DECLARATIONS

Authors' contributions

Conceptualization: Xu, H.; Wang, Z.
Writing: Liu, W.; Liang, J.
Visualization: Liu, W.; Liang, J.
Supervision: Wang, Z.
Funding acquisition: Xu, H.; Wang, Z.
All authors have discussed and agreed on the published version of the manuscript.

Availability of data and materials

Not applicable.

AI and AI-assisted tools statement

Not applicable.

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Conflicts of interest

All authors declared that there are no conflicts of interest.

Ethical approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

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