

Azobenzene-introduced organic electrochemical transistors with enhanced synaptic properties for neuromorphic computing

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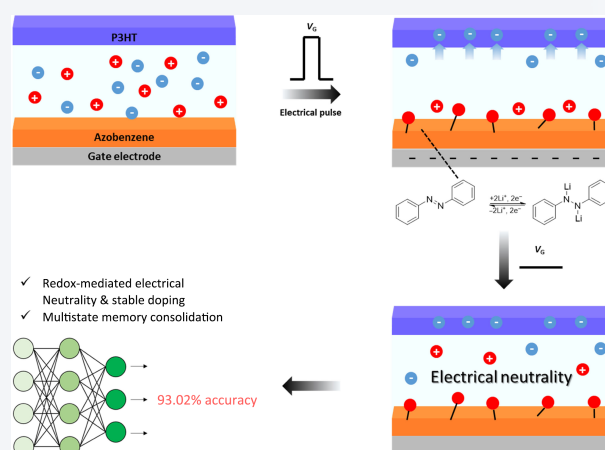
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ABSTRACT: The exponential growth of data-intensive applications has exposed the energy and latency bottlenecks of traditional von Neumann architectures, driving the demand for hardware-based neuromorphic systems. Organic electrochemical transistors (OECTs) have emerged as promising artificial synaptic devices. However, achieving robust long-term plasticity (LTP) remains a critical challenge due to the thermodynamically driven spontaneous de-doping of the active layer. Herein, we present a novel OECT-based synaptic device that overcomes this limitation by incorporating an azobenzene lithiation layer. This redox-active layer effectively compensates for charge imbalance during the electrochemical doping of the poly(3-hexylthiophene-2,5-diyl) (P3HT) channel, thereby maintaining the electrical neutrality of the electrolyte and significantly suppressing spontaneous de-doping. Consequently, the azobenzene-introduced Li-ion gel electrolyte transistor (AB-LGET) exhibits remarkably enhanced synaptic properties, including robust short-term plasticity (STP), prolonged LTP retention, and highly linear weight updates. Grazing-incidence wide-angle X-ray scattering (GIWAXS) analysis confirms that this enhanced plasticity originates from sustained molecular doping within the P3HT crystalline domains. Furthermore, the device achieves a high recognition accuracy of 93.02% in an artificial neural network simulation using the Modified National Institute of Standards and Technology (MNIST) dataset. This study provides a profound understanding of how managing interfacial electrical neutrality via redox reactions can fundamentally advance the synaptic performance of organic neuromorphic devices.

KEYWORDS: organic semiconductor, organic electrochemical transistor (OECT), organic redox transistor, neuromorphic device, synaptic device



1 Introduction

The explosive spread of data-intensive applications, driven by continuous advancements in artificial intelligence (AI) and big data, has precipitated a critical demand for energy-efficient computing paradigms. Conventional von Neumann architectures, characterized by physically separated memory and processing units, suffer from the inherent von Neumann bottleneck. This architectural limitation severely exacerbates energy consumption and latency during massive data transfers. Although state-of-the-art

AI systems exhibit remarkable capabilities, their reliance on software-based algorithms running on traditional hardware fundamentally limits their overall energy efficiency. Consequently, the development of hardware-based neuromorphic computing systems is imperative to achieve high-performance, low-power data processing [1].

Neuromorphic engineering, inspired by the parallel processing and highly efficient neural networks of the human brain [2], offers a compelling solution to these challenges. Artificial synaptic devices, which emulate the dynamic connectivity of biological synapses where neurotransmitters mediate signals between pre- and postsynaptic neurons [3], reside at the core of this paradigm [4, 5]. While two-terminal memristors have been widely investigated due to their structural simplicity, they frequently suffer from inherent drawbacks, including high spatiotemporal variability, low reliability,

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and sneak-path currents. To circumvent these stability issues, multi-terminal synaptic transistors have emerged as superior alternatives, offering decoupled read-write operations and enhanced signal processing capabilities [6, 7]. In particular, neuromorphic devices utilizing organic materials have gained immense attention as highly promising candidates within this paradigm. They offer distinct advantages over their inorganic counterparts, including versatile chemical tunability tailored for specific applications, cost-effective large-area scalability, and inherent compatibility with biological systems [8–12].

Among these, organic electrochemical transistors (OECTs) have gained significant attention owing to their inherent biocompatibility, mechanical flexibility, and low-voltage operation [13–24]. OECTs operate via the permeation of mobile ions into an organic semiconductor channel, translating ionic signals into amplified electronic currents—a process closely mirroring biological synaptic transmission [12, 25]. While OECTs exhibit excellent short-term plasticity (STP) suitable for spatiotemporal information processing, achieving robust long-term plasticity (LTP), which is the fundamental basis for memory and learning remains a challenge [26, 27]. This limitation originates from the intrinsic electrochemical mechanism of conventional OECTs. When a presynaptic voltage pulse drives ions from the electrolyte into the polymeric active layer (i.e., electrochemical doping), it inherently disrupts the electrical neutrality of the electrolyte. Consequently, upon the removal of the electrical stimulus, a spontaneous de-doping process is thermodynamically driven to restore the system's electrical neutrality, causing the injected ions to diffuse back into the electrolyte. This rapid relaxation inevitably erases the conductance state, leading to the failure of long-term memory retention [28, 29]. To enhance LTP characteristics of OECT-based artificial synapses, researchers have explored various strategic interventions. For instance, previous studies have attempted to modulate the crystallinity of the active layer [30], incorporate ferroelectric materials into the electrolyte matrix [31], or precisely regulate ionic permeation utilizing a vertical three-terminal architecture designed for synaptic weight regulation [32]. Furthermore, the integration of an ionic floating gate within the transistor architecture has emerged as a highly effective approach to preserve the macroscopic electrical neutrality of the electrolyte, thereby yielding neuromorphic devices with excellent multistate retention capabilities [13, 33].

To fundamentally resolve this thermodynamic instability, paradigm-shifting strategies inspired by advanced energy storage systems are highly desirable. Organic battery electrode materials, which operate via highly reversible Faradaic redox reactions, offer a compelling pathway to stabilize ionic states [34–36]. When integrated with quasi-solid-state ion gel electrolytes utilizing salts like lithium bis(trifluoromethanesulfonyl)imide (LiTFSI)—whose large, bulky TFSI[−] anions provide robust electrochemical doping and structural stability within the polymer matrix—the interfacial kinetics of the active layer can be precisely modulated [37]. Unlike conventional OECT mechanisms that rely solely on the physically reversible permeation of anions, introducing a dedicated faradaic lithiation layer enables stable, localized charge compensation. By leveraging the robust oxidation-reduction kinetics of materials like azobenzene, we can intrinsically anchor the electrical neutrality of the electrolyte interface. This cross-disciplinary approach translates the exceptional cycle stability of battery materials into highly reliable long-term synaptic plasticity, effectively preventing spontaneous de-doping.

In this study, we propose a novel device architecture to overcome the fundamental limitations of conventional OECTs by introducing a redox-active lithiation layer. Specifically, we incorporate an azobenzene layer capable of undergoing reversible oxidation-reduction reactions with lithium ions [38, 39]. This redox mechanism effectively compensates for the charge imbalance during the doping process, thereby maintaining the electrical neutrality of the electrolyte and substantially suppressing the spontaneous de-doping of the active layer. The rationally designed azobenzene-introduced Li-ion gel electrolyte transistor (AB-LGET) demonstrates markedly enhanced synaptic behaviors, including robust STP, extended LTP retention, and highly linear/symmetric weight update characteristics. The underlying mechanism of this prolonged structural and electrical memory is comprehensively elucidated via grazing-incidence wide-angle X-ray scattering (GIWAXS). Furthermore, utilizing the highly linear conductance modulation of the AB-LGET, we successfully simulate an artificial neural network (ANN) that achieves a high recognition accuracy of 93.02% on the Modified National Institute of Standards and Technology (MNIST) handwritten digit dataset. Ultimately, this research provides vital insights into the critical role of interfacial electrical neutrality in modulating the synaptic plasticity of organic neuromorphic devices.

2 Experimental section

2.1 Materials

Regioregular poly(3-hexylthiophene-2,5-diyl) (P3HT) (M_n : 50,000–60,000, lot number: EM21208) was purchased from Electronic Material & Ink, Republic of Korea. Poly(vinylidene fluoride-co-hexafluoropropylene) (PVDF-HFP) (M_n : 130,000), azobenzene, LiTFSI, chloroform, and acetone were purchased from Sigma-Aldrich.

2.2 Device fabrication

As shown in Fig. S1 in the Electronic Supplementary Material (ESM), OECT-based neuromorphic devices were fabricated using indium tin oxide (ITO) coated glass (10 Ω) as substrate and gate electrode. On the substrate, a layer of azobenzene was applied through a spin-coating process, utilizing an azobenzene solution dissolved in chloroform solvent at a concentration of 50 mg/mL. On the other substrate, the LiTFSI gel was spin-coated using a solution which had PVDF-HFP:LiTFSI:acetone mass ratio of 8:1:56. After drying the gel film at 80 °C for 2 h, a P3HT solution in chloroform (5 mg/mL) was spin-coated onto the film. For better crystallinity, these samples were annealed at 120 °C for 30 min and cooled down to room temperature slowly. Gold source and drain electrodes (40 nm thickness) were thermally deposited through metal mask, and their patterns had a channel length (L) of 100 μm and a channel width (W) of 1 cm ($W/L = 100$). After peeling the integrated ion-gel, P3HT, source/drain electrode film from the substrate, the bottom-gate, top-contact device was completed by placing the integrated film onto the azobenzene-coated gate substrate.

2.3 Characterization

For Synaptic electrical characterizations, all measurements were conducted under vacuum using a Keithley 2612A System SourceMeters and a Keithley 4200-SCS parameter analyzer

(Keithley Instrument Inc.). GIWAXS data were acquired at the PLS-II 9A U-SAXS beamline of the Pohang Accelerator Laboratory in Korea. With an X-ray beam at 11.09 keV ($\lambda = 1.11798 \text{ \AA}$), GIWAXS patterns were recorded by a two-dimensional (2D) charge-coupled device detector with a sample-to-detector distance of 222.2257 mm. The incident angle of the X-ray beam on the sample was fixed at 0.12° . AFM images were obtained using an atomic force microscope (NX-10; Park Systems) installed at the Research Institute of Advanced Materials (RIAM).

2.4 Artificial neural network simulation

Artificial neural network simulations were performed utilizing the open-source “NeuroSim+ V3.0” platform [40]. The system employed a multi-layer perceptron (MLP) architecture configured with dimensions of $400 \times 300 \times 10$. This structure consisted of 400 input neurons to process 20×20 -pixel images, 300 hidden neurons, and 10 output neurons corresponding to the digit classes 0 through 9. Adjacent layers were interconnected via a synaptic weight matrix, which corresponded to the effective conductance values of the devices. Upon feeding an input image into the network, an output vector was generated through the synaptic weight matrix (W) combined with a sigmoid activation function. To enhance digit recognition accuracy, the network iteratively updated these synaptic weights utilizing a back-propagation algorithm, driven by the variance between the predicted outputs and the labels. The MLP model was trained on a dataset of 60,000 patterns and subsequently evaluated for classification performance on an independent set of 10,000 test images. For all numerical computations and modeled synaptic devices, the initial learning rate was set to 0.01, accompanied by a learning rate scheduler that applied a decay factor of 0.75 every 20 epochs.

2.5 Nonlinearity (NL) calculation

The NL values of the long-term potentiation (NL_{LTP}) and depression (NL_{LTD}) were calculated using the following equations (Eqs. (1)–(3)) [40].

$$G_{Pot} = B \times \left(1 - \exp \left(-\frac{P}{A_{Pot}} \right) \right) + G_{min} \quad (1)$$

$$G_{Dep} = B \times \left(1 - \exp \left(-\frac{(P - P_{max})}{A_{Dep}} \right) \right) + G_{max} \quad (2)$$

$$B = \frac{(G_{max} - G_{min})}{\left(1 - \exp \left(-\frac{P_{max}}{A_{Pot, Dep}} \right) \right)} \quad (3)$$

The parameters G_{Pot} and G_{Dep} represent the conductance during potentiation and depression, respectively. The current and maximum number of applied pulses are denoted by P and P_{max} . Furthermore, A_{Pot} and A_{Dep} serve as the key parameters governing the linearity of these processes. The specific relationship between these coefficients and the resulting nonlinearity is detailed in Table S1 in the ESM.

3 Results and discussion

3.1 Mechanism of azobenzene layer-introduced OECTs

In order to enhance the short-term and long-term plasticity of

OECT-based synaptic devices as well as overcome the limitations originated from the mechanisms of conventional OECTs, we introduced lithiation layer which positioned between gate electrode and electrolyte. In this device, azobenzene, which has been studied as an organic battery electrode, was chosen as lithiation layer due to its great redox property and solution processability to build film-stacked OECT structure. As shown in Fig. 1(a), on the ITO gated glass substrate, azobenzene layer, electrolyte and active layer were stacked sequentially. The electrolyte containing Li ions and existing in gel form was composed of LiTFSI and PVDF-HFP as a matrix for Li salts. And P3HT, which has been reported in the field of organic thin film transistors (OTFTs) and organic field effect transistors (OFETs) was chosen as an active layer of p-type OECT due to its high crystallinity and ability to molecular doping [37, 41–43]. On the top of the device, source and drain gold electrode was located. The molecular structures of P3HT, PVDF-HFP, and LiTFSI were displayed in Fig. 1(b). To establish a robust multi-layer architecture and provide a stable underlayer, the device was fabricated by first depositing the Li ion gel onto an ITO/glass substrate, followed by the sequential stacking of the P3HT film. The corresponding atomic force microscopy (AFM) topography reveals a highly uniform and continuous surface without noticeable pinholes or defects (RMS roughness of 0.979 nm), indicating that this sequential deposition process yields a high-quality interface crucial for efficient electrochemical doping (Figs. S2 and S3 in the ESM).

Compared to conventional Li-ion gel electrolyte transistors (LGETs), the introduction of the azobenzene layer (AB-LGET) introduces a distinct Faradaic redox mechanism at the interface, which is highly advantageous for preserving electrical neutrality within the solid-state electrolyte. As illustrated in Fig. 1(c), the application of a negative presynaptic voltage pulse (-2.5 V) induces profound ionic polarization. Driven by the internal electric field, mobile Li^+ cations migrate toward the gate electrode, while TFSI $^-$ anions are injected into the P3HT active layer. This anion permeation electrochemically dopes the P3HT film, generating polarons and bipolarons that proportionally increase the postsynaptic drain current—manifesting as synaptic potentiation. In a conventional LGET, the removal of the gate bias triggers rapid, spontaneous anion de-doping to resolve the macroscopic charge imbalance within the electrolyte, severely degrading long-term memory. In stark contrast, within the AB-LGET architecture, the accumulated Li^+ ions engage in a reversible redox reaction with the azobenzene layer, reducing it to its lithiated state. This localized electrochemical reduction effectively accommodates the positive charge carriers, ensuring that the overall electrical neutrality of the electrolyte is strictly maintained. Consequently, the thermodynamic driving force for the spontaneous back-diffusion of TFSI $^-$ anions is substantially mitigated. This redox-mediated charge compensation fundamentally stabilizes the doped state of the P3HT channel, transforming transient STP into robust and prolonged LTP.

3.2 Synaptic performances of AB-LGET

In biological neural networks, the frequent stimulation of excitatory synapses fundamentally strengthens synaptic weight. To evaluate the efficacy of the azobenzene lithiation layer in modulating synaptic behavior, we first investigated STP. Paired-pulse facilitation (PPF), a form of STP wherein the postsynaptic response is amplified by temporally correlated stimuli, was systematically analyzed. We applied paired presynaptic gate voltage pulses ($V_G =$

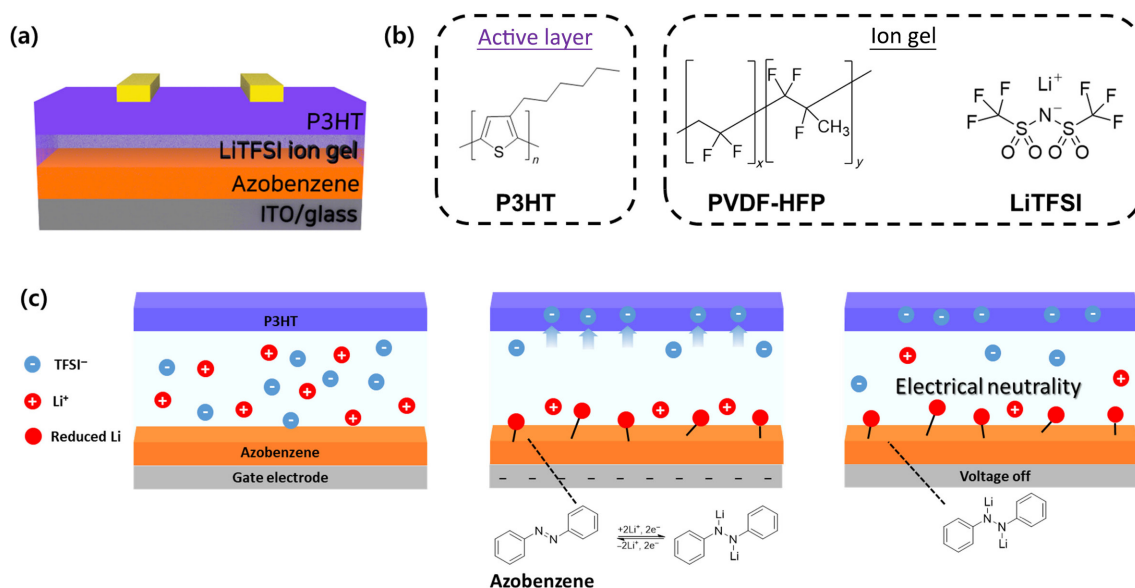


Figure 1 Schematic illustration and operating mechanism of the AB-LGET. (a) 3D device structure of the AB-LGET integrating the azobenzene lithiation layer and the P3HT channel. (b) Chemical structures of the materials composing the AB-LGET, including the P3HT channel and the Li-ion gel composition, the PVDF-HFP as a polymer matrix and LiTFSI as a lithium salt. (c) Schematic representation of the mechanism of interfacial redox reactions maintaining electrical neutrality during the electrochemical doping process. The lithiation of the azobenzene layer maintains electrical neutrality within the ion gel, thereby accommodating the accumulated Li⁺ ions and effectively mitigating the thermodynamic driving force for the back-diffusion of TFSI⁻ anions.

−2.5 V) with varying inter-spike intervals (Δt). The pulse amplitude was determined based on the reduction potential of azobenzene in the LiTFSI system, confirmed via cyclic voltammetry (Fig. S4 in the ESM). Furthermore, to systematically determine the optimal presynaptic voltage, we conducted cyclic potentiation and depression tests by gradually increasing the pulse amplitude from 0.25 to 1.5 V in increments of 0.25 V (Fig. S5 in the ESM). The results indicated that sub-optimal operating voltages yielded poor linearity and a restricted dynamic range. Consequently, the final pulse amplitude was carefully optimized to maximize these synaptic performance metrics while avoiding the electrochemical degradation of the Li ion gel at excessively high voltages and maintaining low operating energy consumption. The PPF index was calculated as the percentage ratio of the second current peak (A_2) to the first (A_1) (Eq. (4) and Fig. 2(a)).

$$\text{PPF}(\%) = \frac{A_2}{A_1} \times 100 \quad (4)$$

The AB-LGET device exhibited a profoundly enhanced PPF index across all Δt ranges compared to the conventional LGET. Notably, the AB-LGET maintained a PPF index of approximately 110% even at prolonged intervals exceeding 30 s, whereas the LGET's synaptic facilitation rapidly decayed to baseline within 10 s as shown in Fig. 2(b), indicating rapid memory loss. Furthermore, the AB-LGET exhibited a maximum PPF of approximately 150%, which is highly consistent with the behavior of biological synapses [44]. In addition, the temporal decay of the PPF index for the AB-LGET was well-fitted to a double-exponential decay model (Eq. (5)), yielding rapid (τ_1) and slow (τ_2) relaxation times of 921 ms and 6624 ms, respectively.

$$\text{PPF} = 1 + C_1 \exp\left(-\frac{\Delta t}{\tau_1}\right) + C_2 \exp\left(-\frac{\Delta t}{\tau_2}\right) \quad (5)$$

These retention times are significantly extended compared to biological synapses ($\tau_1 \sim 40$ ms, $\tau_2 \sim 300$ ms) [45], indicating robust

short-term and long-term memory retention that effectively delays the forgetting process as τ_1 and τ_2 represent the relaxation times associated with STP and LTP, respectively [46]. In alignment with the aforementioned results, the abrupt decay observed in the synaptic facilitation of the LGET is consistent with the rapid relaxation time ($\tau_1 \sim 80$ ms) derived from Eq. (5). While this value exceeds the relaxation time of biological synapses by a factor of two, it represents a marked decrease compared to the AB-LGET. Such behavior is ascribed to the doping and de-doping mechanism facilitated by redox reaction of the azobenzene layer. To further elucidate the robust STP performance enabled by this redox mechanism, we investigated the multistate conductance modulation under varying presynaptic voltage amplitudes and the spike duration-dependent plasticity (SDDP). Specifically, for the multistate conductance test, single potentiation and depression pulses were sequentially applied with amplitudes incrementally increasing from 0.5 to 2.5 V in steps of 0.5 V (Fig. S6(a) in the ESM). Notably, the AB-LGET exhibited highly linear responses during both potentiation and depression even under these single-pulse stimuli. This excellent linearity was further corroborated by linearly fitting the postsynaptic current plotted against the pulse number (Fig. S6(b) in the ESM). Furthermore, the SDDP analysis revealed that the magnitude of the postsynaptic current increased significantly as the applied presynaptic pulse width was extended from 2 to 36 s (Fig. S7(a) in the ESM). These comprehensive evaluations not only reaffirm the exceptional STP characteristics of the AB-LGET but also strongly indicate its capability to achieve highly stable LTP. In stark contrast, the conventional LGET failed to retain any potentiation after just a few pulses under identical SDDP conditions (Fig. S7(b) in the ESM). In terms of kinetics, the addition of the Faradaic redox step from the azobenzene layer does not induce significant further delays. As demonstrated in the SDDP results (Fig. S7 in the ESM), even a short presynaptic pulse of 2 s successfully elicits a distinct excitatory postsynaptic current peak. Furthermore, the relaxation kinetics immediately after the voltage

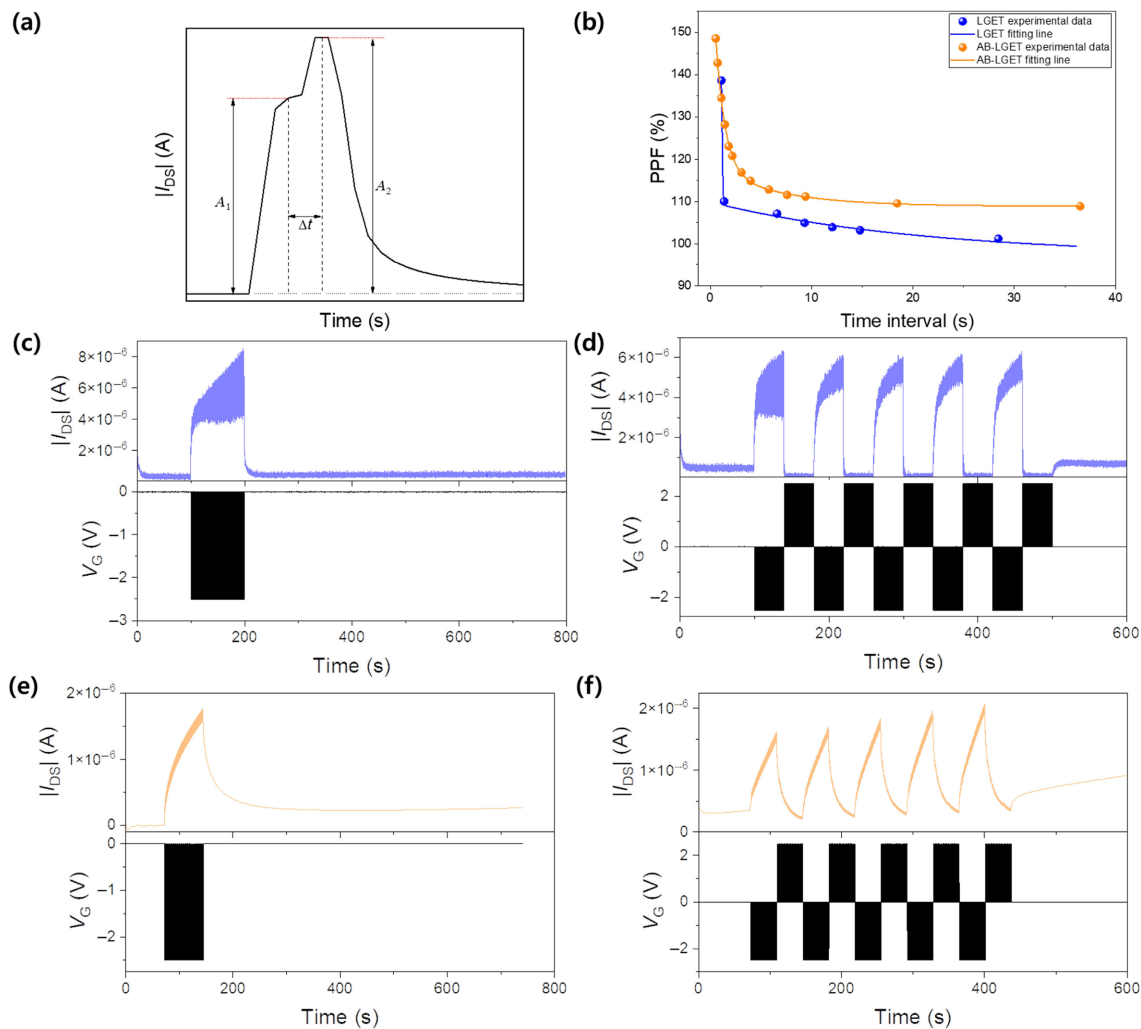


Figure 2 Comparative synaptic characteristics of the LGET and the AB-LGET. (a) The plot of postsynaptic current (PSC) generated by a pair of presynaptic voltage stimuli. (b) PPF index as a function of interval time (Δt), demonstrating STP for the LGET and the AB-LGET. LTP behaviors in response to 100 consecutive voltage pulses for (c) the LGET and (e) the AB-LGET (baseline-normalized). Cyclic potentiation and depression applying 50 potentiating and 50 depressing pulses for (d) the LGET and (f) the AB-LGET.

stimulus is turned off do not exhibit any anomalous delay compared to the typical ion diffusion process. This confirms that the interfacial electron-transfer kinetics of the azobenzene layer are sufficiently fast, ensuring that the redox reaction does not act as a rate-limiting bottleneck for the device's overall operational latency.

Beyond STP, the transition to LTP—a prerequisite for memory consolidation—was evaluated by applying consecutive presynaptic pulses. While the conventional LGET exhibited a transient increase in postsynaptic current during the application of 100 pulses, it catastrophically relaxed to its initial baseline immediately after the stimulus ceased, indicating a complete failure in long-term memory retention. In striking contrast, the AB-LGET demonstrated stable, long-lasting LTP that persisted well beyond 800 s post-stimulation. High linearity in conductance modulation is critical for minimizing weight update errors and enhancing the learning accuracy of artificial neural networks (Figs. 2(c) and 2(e)). To systematically evaluate the enhancement of the LTP characteristics and the postsynaptic current retention capabilities of the devices, we adopted a comprehensive model that categorizes the postsynaptic current relaxation into three distinct regimes based on the sites of ion permeation [25]. According to this model, the current decay

following long-term stimulation is governed by three mechanisms: (1) depolarization of the electric double layer (EDL) at the interface between the P3HT channel and the ion gel, (2) spontaneous de-doping of anions from the amorphous regions of the P3HT film, and (3) de-doping of anions from the highly ordered crystalline regions of the P3HT film (Fig. S8 in the ESM). These processes are quantitatively represented by their respective relaxation time constants ($\tau_{p,1}$, $\tau_{p,2}$, and $\tau_{p,3}$), which are extracted by fitting the postsynaptic current decay curves using a tri-exponential decay function (Eq. (6)).

$$I = I_0 + I_1 \exp\left(-\frac{\Delta t}{\tau_{p,1}}\right) + I_2 \exp\left(-\frac{\Delta t}{\tau_{p,2}}\right) + I_3 \exp\left(-\frac{\Delta t}{\tau_{p,3}}\right) \quad (6)$$

$$\tau_{p,1} < \tau_{p,2} < \tau_{p,3}$$

Here, I_0 represents the initial baseline postsynaptic current prior to the application of presynaptic pulses, while I_1 , I_2 , and I_3 are the pre-exponential factors corresponding to each decay regime. The fitting results of the LTP decay profiles explicitly demonstrate that the AB-LGET exhibits significantly prolonged de-doping time constants across these regimes compared to the conventional LGET (Fig. S9

in the ESM). Fundamentally, the amorphous and crystalline domains exhibit disparate de-doping kinetics. Ion permeation into the amorphous regions occurs at relatively low driving voltages, facilitating their facile and spontaneous outward diffusion from the polymer bulk. Conversely, the intercalation of anions into the tightly interdigitated alkyl side chains within the crystalline domains necessitates a higher energetic driving force. Once intercalated, these anions become tightly bound within the crystallites, severely restricting their outward diffusion. This discrepancy in ion migration kinetics directly yields the two distinct de-doping regimes, where τ_{p2} corresponds to the rapid ion de-doping from the amorphous bulk, and τ_{p3} represents the considerably slower de-doping from the crystalline domains. Consequently, the substantially larger τ_{p3} observed in the AB-LGET indicates a significantly higher population of tightly bound, electrochemically doped anions within the P3HT crystalline regions. This sustained molecular doping is a direct manifestation of the azobenzene redox reaction, which successfully anchors the macroscopic electrical neutrality of the electrolyte and intrinsically suppresses spontaneous de-doping.

To further substantiate the synaptic biomimicry of the AB-LGET, we investigated the spike number-dependent plasticity (SNDP), which represents the transition from short-term to long-term memory. As shown in Fig. S10(a) in the ESM, the retention level of the AB-LGET progressively increased, and the relaxation rate slowed down as the number of applied pulses increased from 40 to 60 and 100. This behavior closely emulates the memory consolidation process in biological synapses, where repeated electrical stimuli gradually strengthen the synaptic weight and convert temporary excitation into stable long-term memory. Conversely, the LGET (Fig. S10(b) in the ESM) failed to establish stable long-term potentiation even under a high number of consecutive pulses. It exhibited rapid spontaneous relaxation back to the initial state across all tested conditions (20, 60, and 100 pulses). These comparative results clearly demonstrate that the redox-mediated charge compensation provided by the azobenzene layer is essential for bridging the gap between short-term excitation and robust long-term plasticity.

Moreover, the AB-LGET exhibited highly linear and symmetric conductance updates during cyclic potentiation (50 pulses, -2.5 V) and depression (50 pulses, 2.5 V) processes. Unlike the LGET, which failed to maintain intermediate conductance states and fluctuated near the baseline, the AB-LGET produced ideal, triangular-shaped weight update curves with highly linear depression and potentiation responses (Figs. 2(d) and 2(f)). These exceptional properties are direct consequences of the unique redox-mediated charge compensation by the azobenzene layer, which fundamentally suppresses spontaneous ion back-diffusion. Furthermore, evaluating energy consumption is crucial for practical applications. According to the following Eq. (7), the energy consumption (E_c) is defined as the product of gate voltage, peak current, and the time duration of the peak (Fig. S9 in the ESM).

$$E_c = V_G \times I_G \times \Delta t \quad (7)$$

The AB-LGET operates with a low energy consumption of approximately $5.93 \mu\text{J}$ per synaptic event, demonstrating high operational efficiency for low-power artificial synapses.

In summary, we successfully demonstrated a highly reliable, OECD-based artificial synapse by strategically incorporating an azobenzene lithiation layer (AB-LGET). This architecture

fundamentally resolves the intrinsic limitations of conventional OECDs, particularly their inability to retain LTP for neuromorphic memory applications. By exploiting the reversible redox properties of azobenzene, we effectively maintained electrical neutrality within the electrolyte during electrochemical doping. This enhancement was thoroughly validated through comprehensive synaptic performance tests, including PPF and cyclic LTP/long-term depression (LTD) measurements. The AB-LGET exhibited superior synaptic characteristics, including extended STP and LTP retention, as well as highly linear potentiation and depression processes compared to the conventional LGET.

3.3 Structural analysis of P3HT molecular doping via GIWAXS

To explain the structural origin of the enhanced synaptic plasticity and the highly linear weight update characteristics, GIWAXS was conducted. This analysis aimed to verify our hypothesis that the azobenzene-mediated redox reaction effectively promotes and sustains the molecular doping within the P3HT channel. We investigated the crystalline structure and molecular orientation of the P3HT films in both LGET and AB-LGET devices before and after applying 100 presynaptic voltage pulses ($V_G = -2.5$ V). The 2D GIWAXS patterns revealed that all P3HT films exhibited a predominant edge-on molecular orientation, characterized by distinct ($h00$) lamellar diffraction peaks along the out-of-plane (q_z) direction (Figs. 3(a)–3(d)). Following the application of presynaptic pulses, the 1D out-of-plane X-ray profiles exhibited a noticeable shift of the (100) peak toward a lower q_z value, accompanied by a slight degradation in the overall crystallinity of the P3HT films (Figs. 3(e) and 3(f)). Furthermore, the GIWAXS pattern of the P3HT film in the AB-LGET after pulses exhibited a diffuse, isotropic scattering ring, which is attributed to the disordered alkyl side chains of P3HT (Fig. 3(d)) [30]. As quantified in Fig. 3(g), the (100) lamellar d -spacing of the P3HT film in the conventional LGET expanded from 16.76 to 17.12 Å after stimulation, while that in the AB-LGET increased from 16.61 to 16.79 Å. This expansion of the backbone-to-backbone distance provides direct structural evidence of electrochemical doping, confirming that the bulky TFSI⁻ anions successfully intercalated into the alkyl side-chain regions of the P3HT crystallites [47]. Furthermore, to assess the impact of ion intercalation on the crystalline order, the coherence length (D_c), which correlates with the average size of the crystalline domains, was calculated from the (100) diffraction peaks using the Scherrer equation (Eq. (8)).

$$D_c = \frac{K\lambda}{\beta \cdot \cos \theta} \quad (8)$$

As shown in Fig. 3(h), the calculated D_c for the P3HT film in the LGET anomalously increased from 124.0 to 140.5 Å after the pulses. Conversely, the AB-LGET exhibited a distinct decrease in D_c from 106.2 to 102.0 Å. This reduction in the average crystalline domain size in the AB-LGET implies that a significant number of TFSI⁻ anions remained stably trapped within the P3HT side-chains even after the electrical stimulus was removed. The sustained presence of these anions slightly disrupts the highly ordered edge-on packing, thereby decreasing the coherence length (Fig. S10 in the ESM). Consequently, this persistent structural alteration strongly corroborates our proposed mechanism: The azobenzene layer effectively stabilizes the doped state by maintaining macroscopic electrical neutrality, which directly translates into the superior long-term memory retention observed in the AB-LGET. Details of

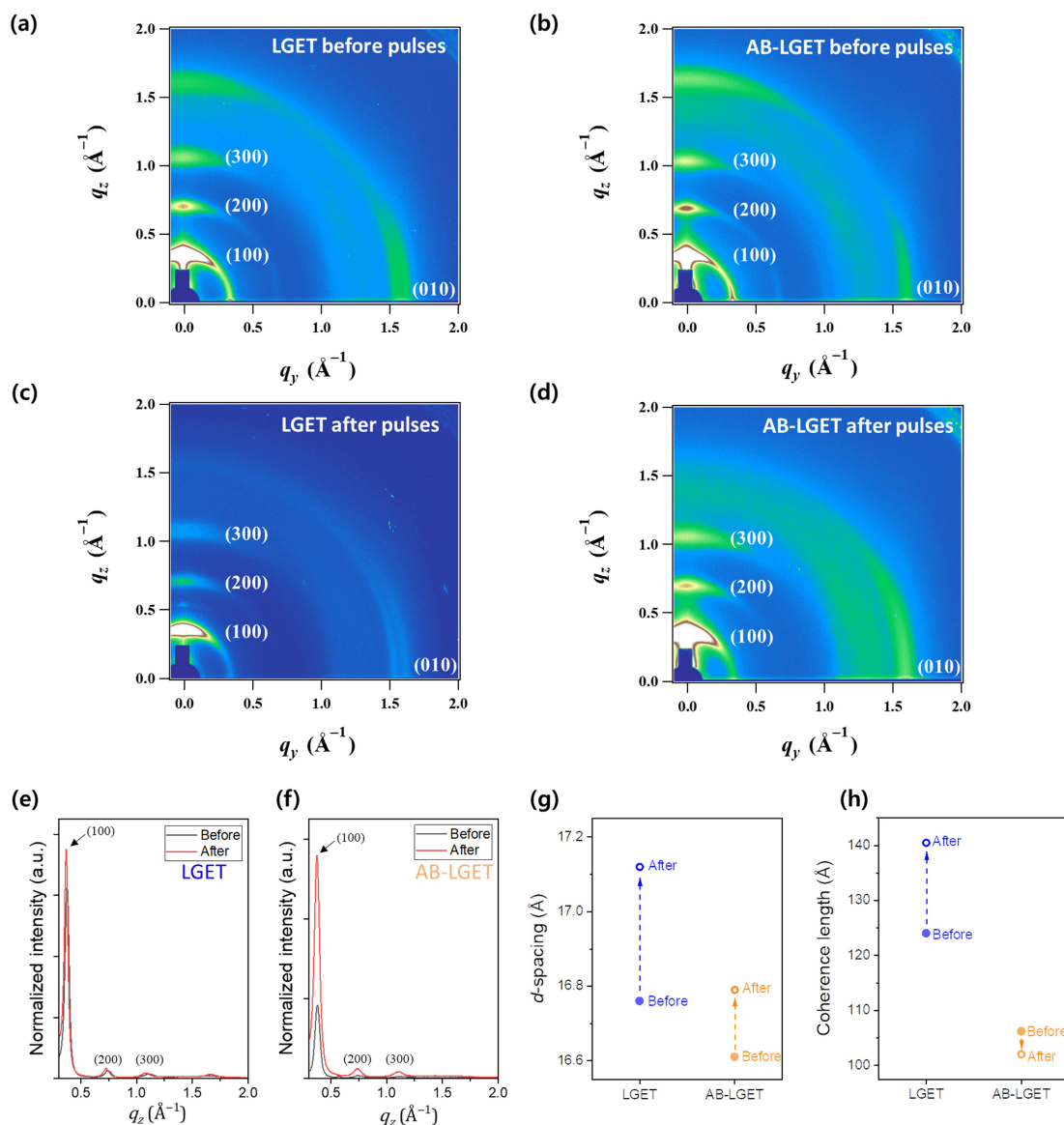


Figure 3 GIWAXS characterization of the P3HT thin films before and after applying 100 presynaptic pulses. GIWAXS patterns of the P3HT thin films before the pulses in (a) the LGET and (b) the AB-LGET. GIWAXS patterns of the P3HT thin films after 100 presynaptic pulses in (c) the LGET and (d) the AB-LGET. 1D out-of-plane X-ray scattering profiles extracted along the q_z axis for (e) the LGET and (f) the AB-LGET. Plots comparing (g) the d -spacing and (h) the coherence length calculated from the corresponding GIWAXS profiles.

crystallographic parameters in the LGET and AB-LGET are summarized in Table S2 in the ESM.

3.4 Learning/recognition test of LGET and AB-LGET

Before learning/recognition test of synaptic devices, it is important to check the stability over potentiation and depression process cycle of OECT-based synaptic devices. In order to observe long-term potentiation and depression cycle stability, we applied 100 voltage pulse of -2.5 V for potentiation and 2.5 V for depression, respectively, for 90 cycles (Fig. 4(a)). Even after 90 cycles, the shape of graph for each cycle remained triangular in AB-LGET synaptic device. Additionally, the dynamic range ($G_{\max}/G_{\min}$), which represents the maximum conductance of the cycle relative to the minimum of conductance of cycle, increased. An enlarged dynamic range can enhance the learning and recognition process by making each step of conductance easier to distinguish, thereby resulting in higher recognition accuracy. With these data, we calculated

nonlinearity of long-term potentiation (NL_{LTP}) and depression (NL_{LTD}) by fitting the measured data to the normalized one (Fig. 4(b)) [40]. Typically, OECT-based synaptic devices exhibited a larger value in NL_{LTD} compared to NL_{LTP} , because injecting anions into the P3HT film requires additional external forces like electrical stimuli, whereas the ions can diffuse back without such forces. Thereby, anions trapped by external forces in the crystalline structure of the P3HT film have a depression rate proportional to the doping concentration, causing them to revert to their original state quickly once the external force is removed. As a result, the long-term depression exhibits worse nonlinearity compared to the potentiation. The calculated NL_{LTP} and NL_{LTD} were approximately 0.43 and 6.08, respectively.

In order to fundamentally explain the origin of this performance disparity, the cyclic long-term potentiation and depression characteristics of the conventional device were evaluated under identical pulse conditions. In stark contrast to the AB-LGET, the

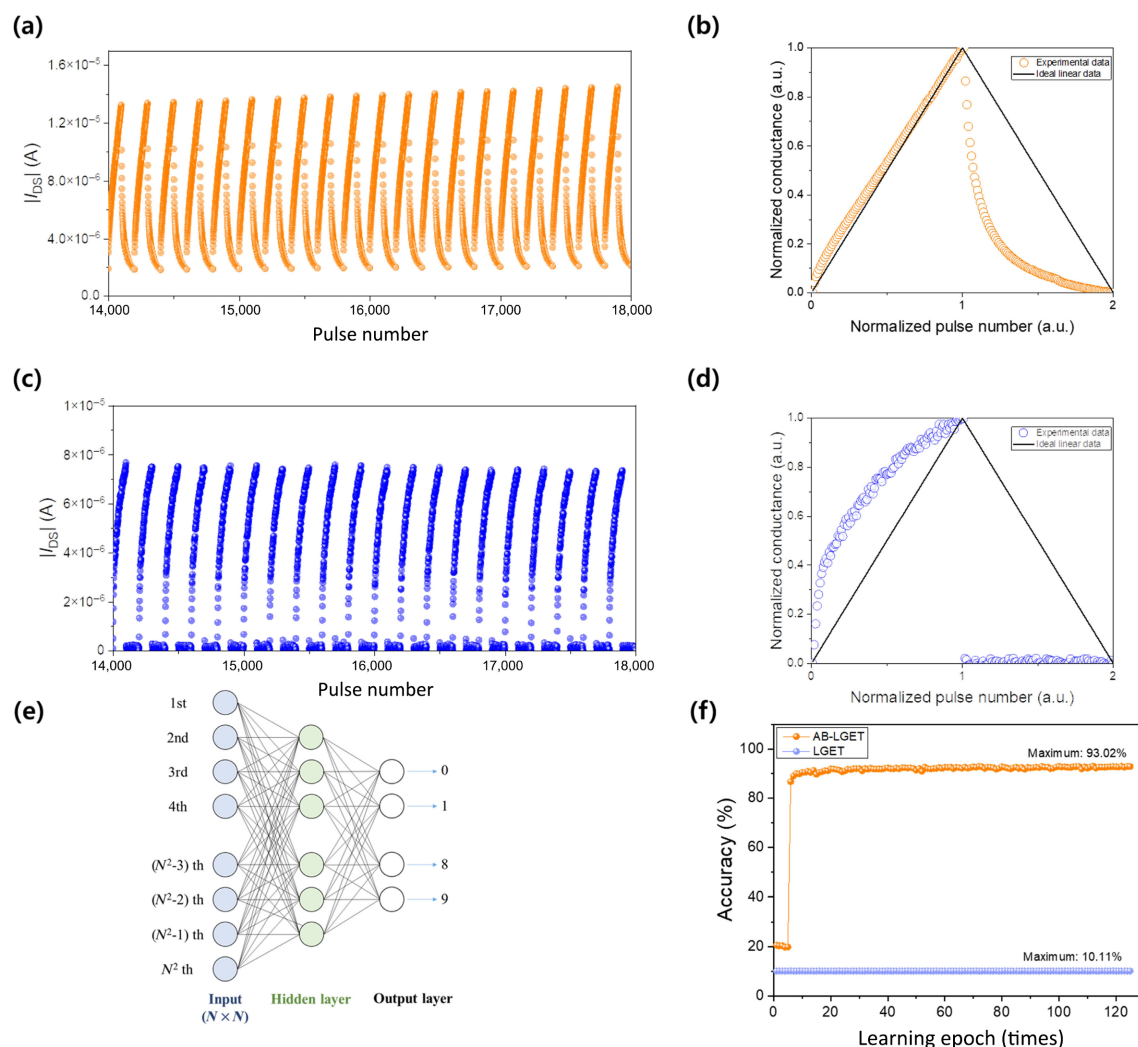


Figure 4 Pattern training and recognition performance of the LGET and the AB-LGET. LTP and LTD curves between 70th and 90th cycles, along with the enlarged view of the 90th cycle, for (a) and (b) the AB-LGET and (c) and (d) the LGET. (e) Schematic illustration of the multilayer perceptron (MLP)-based ANN with a $400 \times 300 \times 10$ configuration. (f) Recognition accuracy as a function of the number of training epochs for the AB-LGET and the LGET.

conventional architecture failed to exhibit the triangular cycle and displayed a significantly restricted dynamic range (Fig. 4(c)). During the potentiation phase, the conductance increase suffered from severe degradation in linearity. More critically, in the depression regime, the postsynaptic current precipitously plummeted to near-zero immediately upon stimulation and merely fluctuated around the baseline. This abrupt current decay explicitly indicates the immediate spontaneous de-doping of the P3HT channel, which is thermodynamically driven to maintain the electrical neutrality of Li-ion gel (Fig. 4(d)). Therefore, the calculated NL_{LTP} and NL_{LTD} values for the conventional device were 2.71 and 9.68, respectively, demonstrating profoundly higher nonlinearity in both potentiation and depression regimes when compared to the AB-LGET neuromorphic device.

To evaluate the potential of a neuromorphic computing system based on the AB-LGET in comparison to the LGET synaptic device, we simulated ANNs using a multi-layer perceptron (MLP) model with dimensions of $400 \times 300 \times 10$, incorporating the measured LTP/LTD characteristics. This model, designed to recognize 20×20 -pixel images, uses 400 input neurons, 300 hidden neurons, and 10 output neurons corresponding to 10-digit

classes (Fig. 4(e)). Each pair of neurons is connected through a synapse, with the connection strength reflecting the synaptic weights. We performed training and recognition tasks using MNIST handwritten digit patterns, and tracked the recognition rate of each device across epochs. During each epoch, the model was trained on 60,000 randomly selected images from the training dataset, classified 10,000 images from the testing dataset, and assessed the recognition accuracy of these classified images. In this way, learning/recognition accuracy of LGET and AB-LGET was calculated and plotted as a graph (Fig. 4(f)). This graph demonstrated significantly improved accuracy of learning/recognition through the introduction of azobenzene. The LGET synaptic device maintained a steady accuracy of 10.11% over 125 epochs without any improvement. In contrast, the AB-LGET synaptic device initially had an accuracy of approximately 20%, which increased to over 80% by the 6th epoch and reached up to 93.02% during the same epoch as the LGET device. Through this MNIST handwritten digit patterns learning and recognition task, we validated the feasibility of utilizing the AB-LGET for ANN-based neuromorphic computing.

4 Conclusion

In summary, we successfully demonstrated a highly reliable, OECT-based artificial synapse by strategically incorporating an azobenzene lithiation layer (AB-LGET). This architecture fundamentally resolves the intrinsic limitations of conventional OECTs, particularly their inability to retain LTP for neuromorphic memory applications. By exploiting the reversible redox properties of azobenzene, we effectively maintained electrical neutrality within the electrolyte during electrochemical doping. This enhancement was thoroughly validated through comprehensive synaptic performance tests, including PPF and cyclic LTP/LTD measurements. The AB-LGET exhibited superior synaptic characteristics, including extended STP and LTP retention, as well as highly linear potentiation and depression processes compared to the conventional LGET. GIWAXS analysis provided definitive physical evidence that this improved plasticity stems from enhanced molecular doping retention within the P3HT crystalline domains, facilitated by the azobenzene redox reaction. Furthermore, evaluating energy consumption is crucial for practical applications; the AB-LGET operates with a remarkably low energy consumption of approximately 5.93 μJ per synaptic event, demonstrating high operational efficiency for low-power artificial synapses. Leveraging these ideal synaptic behaviors, an ANN simulation based on the MNIST handwritten digit dataset was conducted. The AB-LGET achieved a remarkable learning and recognition accuracy of 93.02%, drastically outperforming the conventional LGET (10.11% over 125 epochs). Ultimately, the simple yet highly effective integration of an azobenzene lithiation layer provides a robust design principle for engineering the interfacial electrical neutrality in organic neuromorphic devices, paving the way for next-generation, high-performance computing hardware.

Electronic Supplementary Material: Supplementary material (including additional device fabrication details, morphological characterization, cyclic voltammetry, multistate conductance modulation, spike duration- and spike number-dependent plasticity measurements, relaxation analysis, energy-consumption analysis, a nonlinearity table, and additional structural characterization) is available in the online version of this article at <https://doi.org/10.26599/NR.2026.94909008>.

Data availability

All data needed to support the conclusions in the paper are presented in the manuscript and the Electronic Supplementary Material. Additional data related to this paper are available from the corresponding author upon reasonable request.

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Declaration of competing interest

All the contributing authors report no conflict of interests in this work.

Author contribution statement

H. K.: Idea conceptualization, device fabrication and characterization, numerical simulations and analysis, writing original draft, reviewing and editing manuscript. Y. W.: Idea conceptualization, numerical simulation and analysis. J. H. O.: Idea conceptualization, supervision, writing original draft, reviewing and editing manuscript. All authors discussed the results and commented on the manuscript.

Use of AI statement

None.

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