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Research Highlight

Polyelectrolyte-confined fluidic memristor for neuromorphic computing in aqueous environment

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Artificial synapse devices with co-location memory and logic functions have attracted enormous attention since they are the indispensable components for neuromorphic computing systems [1]. Extensive efforts have been made to mimic neural electric pulse patterns through solid-state devices like two-terminal memristors and three-terminal transistors. However, it remains highly desired to obtain chemical neuromorphic functions in aqueous environment for brain-inspired computation and biomedical applications [2,3]. Although organic electrochemical transistors could be operated in aqueous solution, their three-terminal structure is unfavorable for reliable integration with biological synapses at high density [4,5]. Fluidic memristor based on simple two-terminal structure and confined ion transport is promising to be well compatible with biological systems, but the neuromorphic function remains an unmet need due to the shielding effect of interionic interaction in aqueous solution [6,7].

Until very recently, inspired by the biological ion channels (Fig. 1a), Mao and co-workers [8] from the Institute of Chemistry, Chinese Academy of Sciences have made a breakthrough by designing a polyelectrolyte-confined fluidic memristor (PFM) to realize neuromorphic functions in the aqueous environment. Such PFM was fabricated through surface-initiated atomic transfer radical polymerization to modify the inner wall of channels with polyimidazolium brushes (PimBs) (Fig. 1b). Based on the spatial confinement and molecular recognition of such modified channels, the resultant PFMs allow for hysteretic ion transport under the stimulation of electric fields and chemicals, thus leading to a typical history-dependent ion memory effect. Besides the capacity of diverse electric pulse patterns to function as typical memristors, the PFMs could also generate chemical-regulated electric pulses and chemical-electric signal transduction benefiting from their intrinsically fluidic properties. With the structural and functional similarity to the natural biological systems, the PFMs are expected to promote the comprehension and interaction of chemical language from the human brain.

The PFM device was composed of PimBs-confined channel immersed in an electrolyte solution, two Ag/AgCl electrodes were respectively placed in the channel and electrolyte for electric measurements. Under the stimulation of external voltage, the PFMs could generate various electric pulse patterns to successfully realize the neuromorphic functions of traditional memristors. With systematical studies, the current-voltage (*I*-*V*) relationship of PFMs exhibited a typical hysteresis loop in contrast to that of bare fluidic memristor (Fig. 1c), indicating the significant influence of surface charge in PimBs-confined channels. Further measurements also demonstrated the change of ion conductivity for PFM was a time-dependent process, ultimately endowing the devices with ion memory and memristive performances. Such ion memory originated from the relatively slow redistribution and diffusion dynamics of ions, which was controlled by Pim–anion interactions due to the spatial confinement effect. To mimic short-term plastic electric pulse patterns, the PFMs were stimulated by voltage pulses to achieve synaptic plasticity like paired-pulse facilitation and paired-pulse depression. These diverse electric pulses operated by the PFMs showed long retention time and low energy consumption similar to biological systems, thus allowing for effective neuromorphic computation under the aqueous environment.

In comparison to the other neuromorphic devices, the key difference offered in this work is that the fluidic-based devices not only exhibit neuromorphic performances comparable to the biological system, but also process advanced chemical-related functions. For instance, the PFMs could mimic chemical-regulated short-term plastic electric pulses according to the changes in external chemical environment (Fig. 1d), including the ionic strength, ion species and electrolyte concentration. This chemical modulation effect of PFMs was obtained by tuning the Pim–anion interactions and the synergism of multiple ion species, which is almost impossible for solid-state systems. More importantly, the fluidic-based ion redistribution dynamics of the PFMs offer possible means to regulate neuroplastic behaviors through the use of bioactive molecules. In a physiological electrolyte, the PFMs could mimic the regulation of memory by adenosine triphosphate, paving the way for the implementation of direct interfaces and communications with biological systems.

Mimicking the neurotransmitter-related functions of chemical synapses has long been a challenge, especially for solid-state memristors. In biological systems, signal transduction among chemical synapses is generally mediated by the release and recognition of

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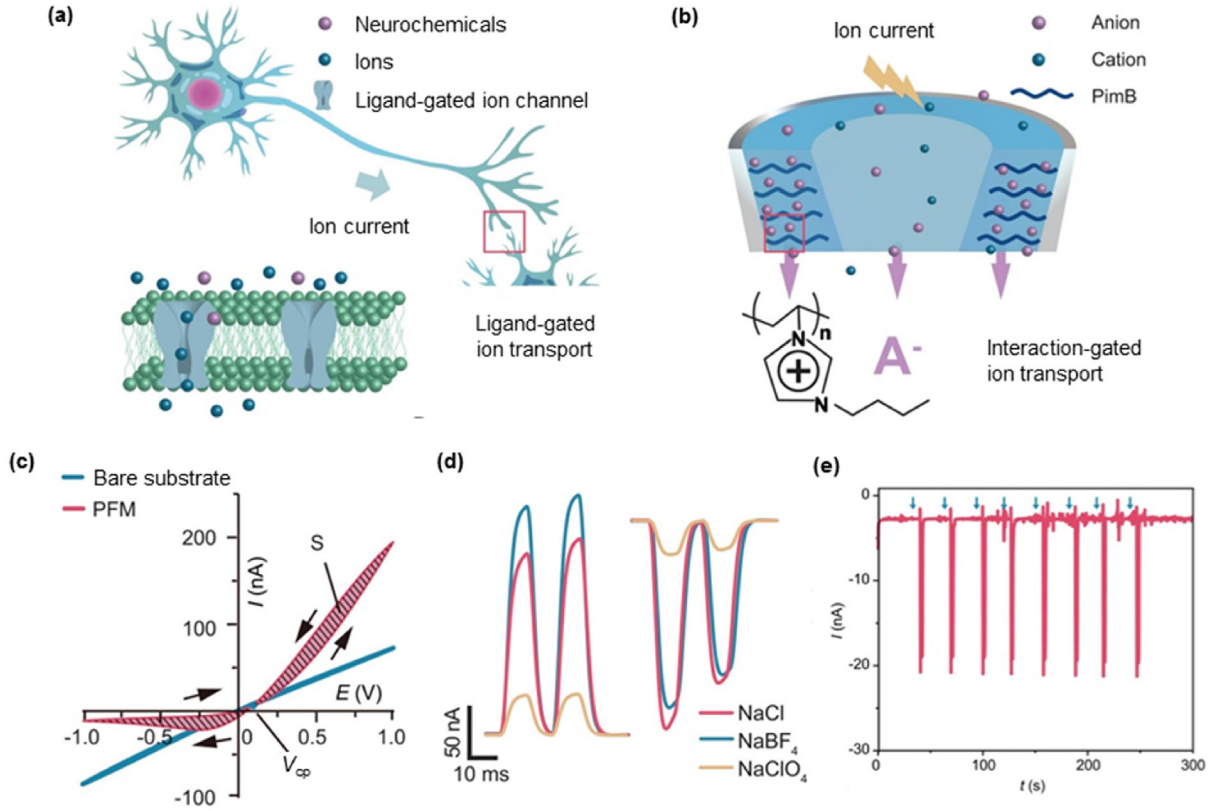

Fig. 1. Schematic illustration of the neuromorphic functions realized by interaction-gated ion current in biological neurons (a) and a PimBs-confined fluidic system (b). (c) *I-V* curves of PFM (red) and memristor with bare channels (blue) in 10 mmol/L electrolyte solution under a triangle wave with a scan rate of 50 mV/s. The hysteresis loop area is shaded in purple. (d) Paired-pulse facilitation and paired-pulse depression of PFM in different electrolyte solutions. (e) Electric pulse response of PFM under chemical stimulation. The blue arrows indicate the delivery of artificial neurotransmitters. Reprinted with permission from Ref. [8], Copyright © 2023, American Association for the Advancement of Science.

neurotransmitters. Like the transport of neurotransmitters among biological synapses, the chemical-electric signal transduction could be accomplished with PFMs by tuning the interaction between polyelectrolytes and multiple ion species in the channels. When the chemical stimuli (e.g., ClO_4^-) as artificial neurotransmitters were injected, the PFM showed an electric pulse response (Fig. 1e), demonstrating its capability of transferring the chemical stimuli of certain species into electric pulse signals. That is to say, the PFMs with easily adaptable configurations can work as artificial chemical synapses for implantable devices and neuroprosthetics.

In summary, this study paves the avenue to connect chemistry with neuromorphic devices for brain-inspired computing, intelligence sensing, human–machine interfaces and neurosensory prosthetics, representing a remarkable landmark for constructing multi-functional artificial synapse devices by interface chemistry. The fabricated PFM is versatile and features with high biocompatibility, considerable memristive performances and neuromorphic functions, which will make an important impact on the future of artificial intelligence. For practical applications, given the ion-based fluidic systems in nature, more research could be explored to improve the stability and switching speed of devices. Also, in a crossbar array, the design of device packaging, soft circuit and large-scale integration is desired to fulfill the requirement for brain-inspired computing and biomedical applications.

Conflict of interest

The authors declare that they have no conflict of interest.

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