

## Organic/inorganic hybrid synaptic transistors gated by proton conducting methylcellulose films (Postprint)

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### Abstract

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### Full Text

#### Organic/Inorganic Hybrid Synaptic Transistors Gated by Proton-Conducting Methylcellulose Films

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### Abstract

The concept of building brain-inspired cognitive systems has been pursued for several decades. Recently, electric-double-layer transistors gated by ion-conducting electrolytes have emerged as promising candidates for synaptic electronics and neuromorphic systems. In this letter, we experimentally demonstrate indium-zinc-oxide transistors gated by proton-conducting methylcellulose electrolyte films that exhibit synaptic plasticity, including paired-pulse facilitation and spatiotemporal-correlated dynamic logic. More importantly, we propose a model based on proton-related electric-double-layer modulation and

a stretched-exponential decay function, and the theoretical results show good agreement with the experimentally measured synaptic behaviors.

## Introduction

Conventional computers based on CMOS technology and von Neumann architecture excel at logic operations and precise scientific calculations, but they cannot compete with biological brains when solving problems involving real-world interaction where inputs and outputs are often imprecisely specified. Inspired by the energy efficiency and cognitive capabilities of biological brains, neuromorphic computing has emerged as an attractive computational paradigm that complements von Neumann architecture. To realize neuromorphic computing, both software- and hardware-based approaches have been proposed. However, software-based approaches face significant challenges in energy dissipation because the algorithms are essentially run on conventional von Neumann computers with limited parallelism. Hardware-based approaches can potentially overcome these challenges by implementing massive parallelism at the physical level. Synaptic electronics have attracted wide attention in recent years, aiming to develop artificial synaptic devices that emulate the memory and computation performed by biological synapses.

To date, a broad spectrum of synaptic devices has been developed with essential synaptic functions. Emerging memory structures exhibit several characteristics of history-dependent analog-like states, making them suitable for neuromorphic systems. For example, amorphous InGaZnO memristors have been proposed to mimic spike-timing-dependent plasticity (STDP), a Hebbian learning rule observed experimentally, which arises primarily from conductance changes induced by oxygen ion migration/diffusion. Carriers in semiconductor channel layers can be strongly accumulated or depleted by electrolyte gating due to the extremely large electric-double-layer (EDL) capacitance formed at the electrolyte/semiconductor interface. The EDL can be regarded as a nanoscale capacitor with a huge specific capacitance exceeding  $1.0 \mu\text{F}/\text{cm}^2$ . Recently, three-terminal EDL transistors gated by ion-conducting electrolytes have also been proposed as promising candidates for synaptic electronics. In previous reports, the gate pulse and channel current (or channel conductance) were defined as the synaptic stimulus and synaptic weight, respectively. The synaptic weight of these transistors can thus be modified by tuning the pattern and timing of the stimuli, enabling plasticity behaviors analogous to biological synapses. However, a physical understanding of such synaptic devices remains lacking.

Indium zinc oxide (IZO) films with high mobility ( $10\text{--}50 \text{ cm}^2/\text{V} \cdot \text{s}$ ) and large-area uniformity can be deposited by radio-frequency sputtering at low temperature or even at room temperature. Thin-film transistors with IZO channel layers are excellent candidates for switching or driving transistors in next-generation and/or flexible flat panel displays, sensors, and other novel applications, including neuromorphic systems. In this letter, we fabricate IZO-based EDL transistors gated by proton-conducting methylcellulose electrolyte films

and demonstrate their use for synaptic function emulation. Synaptic plasticity emulations are theoretically modeled based on proton-related EDL modulation and a stretched-exponential decay function, and the theoretical results show good agreement with experimental data. These findings could provide useful guidelines for designing artificial synapses for neuromorphic computing systems.

## Experimental Methods

Methylcellulose (MC) is a chemical compound derived from cellulose with good thermal, chemical, and mechanical stability. It is synthetically produced through a substitution reaction in which hydroxyl residues (-OH functional groups) are replaced by methoxide (-OCH<sub>3</sub> groups). These functional groups enable MC films to absorb water molecules, contributing to high proton conductivity. MC films with good stability and high proton conductivity are of great interest for EDL transistor fabrication and synaptic electronics applications. First, MC powder (Sinopharm Chemical Reagent Co., Ltd.) was mixed into deionized water at 85 °C for 5 min and then cooled in air to obtain a homogeneous solution with a concentration of 1.0 wt. %. The solution was then spin-coated onto ITO glass substrates and dried in air at 50 °C for 2 h. The thickness (D) of the MC film is estimated to be 9.0 μm. Subsequently, 150-nm-thick IZO patterns (1000 μm × 150 μm) were deposited on the MC film by magnetron sputtering at room temperature using a nickel shadow mask. As shown in Fig. 2 Figure 2: see original paper, when the gap between the shadow mask and substrate is about 50 μm, a thin IZO channel layer with a thinnest part of 30 nm can be self-assembled between the IZO source/drain electrodes due to reflection of IZO nanoparticles at the mask edge and dimensional extension by IZO nanoparticles with a low incident angle. Proton conductivity and specific capacitance of the MC films were measured using a Solartron 1260A impedance analyzer. The electrical performance of the IZO synaptic transistors was characterized using a Keithley 4200 SCS semiconductor parameter characterization system.

## Results and Discussion

The proton conductivity of the MC film was determined from the Cole-Cole plot, as shown in Fig. 1 Figure 1: see original paper. Proton conductivity ( $\sigma$ ) is calculated using the relation  $\sigma = D/(R - R_0)A$ , where D, R, R<sub>0</sub>, and A represent the thickness of the MC film, the intersecting point on the real axis of the Cole-Cole plot, the resistance of the electrodes, and the electrode area, respectively. Here, A is  $1.5 \times 10^{-3}$  cm<sup>2</sup>, and R<sub>0</sub> is 30 Ω. Therefore, the  $\sigma$  value is estimated to be  $6.5 \times 10^{-4}$  S/cm. Figure 1(b) shows the frequency-dependent specific capacitance of the proton-conducting MC film measured with an in-plane test structure (inset in Fig. 1(b)). The specific capacitance increases with decreasing frequency and gradually reaches a maximum value (C<sub>{DL}</sub>) of 3.1 μF/cm<sup>2</sup> at 1.0 Hz, indicating EDL formation at the electrode/electrolyte interface. Figure 2(a) shows a schematic of the IZO-based synaptic transistor gated by MC film

on a glass substrate with two in-plane gates. The voltage applied to the in-plane gates can be capacitively coupled to the IZO channel. Figure 2(b) shows the transfer curve of the IZO-based synaptic transistor with a constant  $V_{DS}$  of 1.5 V. The gate voltage was swept from -1.0 V to 0.8 V and then back. A clear anticlockwise hysteresis window of 0.25 V was observed, likely due to mobile protons within the MC electrolyte film. The subthreshold swing (SS), current on/off ratio ( $I_{on}/I_{off}$ ), and threshold voltage ( $V_{TH}$ ) are estimated to be 85 mV/decade,  $2.5 \times 10^6$ , and 0.14 V, respectively. In addition, the field-effect electron mobility ( $\mu_{FE}$ ) in the saturation region ( $V_{DS} > V_{GS} - V_{TH}$ ) is estimated to be  $21.2 \text{ cm}^2/\text{V} \cdot \text{s}$  using the following equation:

$$I_{DS} = \frac{WC_{DL}\mu_{FE}}{2L}(V_{GS} - V_{TH})^2$$

where  $W$ ,  $L$ , and  $C_{DL}$  are the channel width, channel length, and specific capacitance of the EDL, respectively.

Excitatory postsynaptic currents (EPSCs) or inhibitory PSCs (IPSCs) can be triggered by potential spikes from presynaptic neurons, which are collectively processed by postsynaptic neurons to establish spatial and temporal correlated functions. As previously reported, the gate pulse and channel current (or channel conductance) were defined as the synaptic stimulus and synaptic weight, respectively. The synaptic weight of the devices can be modified by tuning the pattern and timing of the stimuli, similar to biological synapses. In our experiments, presynaptic spikes were applied to the gate electrodes to emulate synaptic behaviors. The channel current measured at  $V_{DS} = 0.1 \text{ V}$  was regarded as EPSC. Figure 3 Figure 3: see original paper shows the EPSC response triggered by a presynaptic spike (40 ms, 0.1 V) applied to Gate1 (G1). The inset of Fig. 3(a) shows the schematic diagram for EPSC measurement. As shown in Fig. 3(b), during a presynaptic spike, protons are electrically triggered and migrate to the IZO channel/methylcellulose electrolyte interface. The transient accumulation of protons then induces a sharp increase in channel current through strong EDL electrostatic coupling. When the spike ends, protons diffuse back to equilibrium positions due to concentration gradients. The decay of proton concentration at the IZO/methylcellulose interface thus results in a gradual decay of channel current.

As mentioned above, the increment in channel current is due to proton accumulation at the MC/IZO interface. We assumed no interfacial electrochemical reaction occurs at low gate voltage ( $V_{GS} \leq 1.5 \text{ V}$ ). Thus, the electrolyte film can be simply modeled as a resistor-capacitor (RC) circuit, as shown in Fig. 3(b). The voltage across the electrolyte film yields:

$$\Delta V_{DL}(t) = V_A \left[ 1 - \exp\left(-\frac{t}{\tau_{DL}}\right) \right]$$

where  $V_A$  and  $\tau_{DL}$  are the applied voltage and time constant of the RC circuit, respectively. During the charging process, the induced carriers in the IZO channel layer increase the channel conductance. If the duration of a positive spike is  $T$ , the proton concentration ( $t \leq T$ ) will increase with the charging voltage. According to Eq. (2), the proton concentration ( $\Delta N(t)$ ,  $t \leq T$ ) can be expressed as:

$$\Delta N(t)_{t \leq T} = \frac{C_{DL}(V_{GS} - V_{TH})}{ZqW L d_E} \left[ 1 - \exp\left(-\frac{t}{\tau_{DL}}\right) \right]$$

where  $Z$ ,  $q$ , and  $d_E$  are the ionic valence ( $Z = 1$  for proton), elementary charge, and depth of the ion-accumulation layer in the electrolyte, respectively. When the spike ends, protons gradually diffuse back to equilibrium positions. Stretched-exponential decay has been widely applied to model relaxation processes such as hydrogen diffusion in a-Si:H and oxygen vacancy diffusion in WO<sub>x</sub>. In our case, the stretched-exponential function can also model proton relaxation processes in the proton-conducting MC film. Thus, the proton concentration after the presynaptic spike ( $t > T$ ) can be determined by:

$$\Delta N(t)_{t > T} = \Delta N(T)_{t \leq T} \exp\left[-\left(\frac{t-T}{\tau}\right)^\beta\right]$$

where  $\tau$  and  $\beta$  are the time constant of ion relaxation and the stretch index (ranging between 0 and 1), respectively. It should be noted that the total EPSC ( $I_{DS}$ ) is determined by the relation  $I_{DS} = I_0 + \Delta I_{DS}$ , where  $I_0$  is the base channel current when no presynaptic spike is triggered ( $V_{GS} = 0$  V), and  $\Delta I_{DS}$  is the change in channel current (EPSC) induced by a presynaptic spike. The change in conductivity ( $\Delta\sigma$ ) of the semiconductor layer can be determined by:

$$\Delta I_{DS} = \Delta n q \mu \frac{V_{DS} W d_S}{L}$$

where  $\Delta J$ ,  $E$ ,  $d_S$ , and  $\Delta n$  are the changes in current density, electric-field intensity, depth of the carrier-accumulation layer in the semiconductor, and induced electron concentration, respectively. Therefore,  $\Delta I_{DS}$  can be written as:

$$\Delta I_{DS} = \Delta n q \mu \frac{V_{DS} W d_S}{L}$$

Due to the huge EDL capacitance, the induced electron concentration should be approximately equal to the induced proton concentration ( $\Delta n = \Delta N$ ). Thus, the EPSC amplitude ( $A(t)$ ) can be deduced by substituting  $\Delta N(t)\{t \leq T\}$  and  $\Delta N(t)\{t > T\}$  into Eq. (7):

$$A(t) = \Delta I_{DS} = \begin{cases} \gamma \left[ 1 - \exp\left(-\frac{t}{\tau_{DL}}\right) \right] & t \leq T \\ \gamma \left[ 1 - \exp\left(-\frac{T}{\tau_{DL}}\right) \right] \exp\left[-\left(\frac{t-T}{\tau}\right)^\beta\right] & t > T \end{cases}$$

where  $\gamma = (C_{DL} L_{dS} / Z_{qWLdE}) (V_{GS} - V_{TH}) V_{DS}$ .  $\gamma$  can be regarded as the EPSC amplitude triggered by a continuous presynaptic spike ( $T \rightarrow \infty$ ).

The measured EPSC is plotted as open circles in Fig. 3(a). The fitted result based on Eq. (8) is plotted as a solid line in Fig. 3(a). The fitting parameters  $\gamma$ ,  $\tau_{DL}$ ,  $\tau$ , and  $\beta$  are estimated to be 766 nA, 257 ms, 9.0 ms, and 0.74, respectively. The fitting curve matches the experimental results very well, indicating the validity of our proposed model.

Paired-pulse facilitation (PPF) is a short-term form of synaptic plasticity representing an amplitude increase in the second synaptic response when it follows shortly after the first. Figure 4: see original paper shows the EPSC response triggered by two successive presynaptic spikes (0.1 V, 10 ms) with a time interval ( $\Delta T$ ) of 40 ms. The second EPSC peak ( $A_2$ ) is 156 nA, larger than the first EPSC peak ( $A_1$ ) of 148 nA. The inset in Fig. 4(a) schematically illustrates the PPF measurement. The facilitation ratios ( $A_2/A_1$ ) are plotted as a function of  $\Delta T$  in Fig. 4(b). The obtained PPF emulation results are in good agreement with observations in biological systems. According to Eq. (8), the EPSC triggered by the first spike reaches its maximum value ( $A_1$ ) of  $\gamma [1 - \exp(-T/\tau_{DL})]$  at the end of the spike ( $t = T$ ). After that, protons diffuse back to equilibrium positions following the stretched exponential function. During the second pulse, residual protons from the first spike add to the total proton concentration, inducing an increment in EPSC. Therefore, the amplitude of the second EPSC peak can be estimated by:

$$A_2 = \gamma \left[ 1 - \exp\left(-\frac{T}{\tau_{DL}}\right) \right] + \kappa \gamma \left[ 1 - \exp\left(-\frac{T}{\tau_{DL}}\right) \right] \exp\left[-\left(\frac{\Delta T}{\tau}\right)^\beta\right]$$

where  $\kappa$  is a factor related to the effective residual proton concentration contributing to the second EPSC. The facilitation ratio ( $A_2/A_1$ ) between the two EPSC amplitudes can then be determined by:

$$F(\Delta T) = 1 + \kappa \exp\left[-\left(\frac{\Delta T}{\tau}\right)^\beta\right]$$

The facilitation ratio ( $A_2/A_1$ ) is plotted as a function of  $\Delta T$  in Fig. 4(b). The blue solid line is fitted using Eq. (10) with  $\kappa = 2.2$ . We find that the fitting curve approaches the experimental results very well.

Dynamic logics established by spatiotemporal correlated spikes are also demonstrated in the proposed synaptic transistor with two in-plane gates (G1 and G2) as presynaptic terminals. Figure 5 Figure 5: see original paper shows the schematic diagram of this emulation. Two presynaptic spikes  $P_1$  and  $P_2$  (0.1 V, 10 ms) with a time interval ( $\Delta T$ ) are applied to presynaptic terminals G1 and G2, respectively. Zero time ( $t = 0$ ) is defined as when  $P_2$  ends. The EPSC amplitudes measured at  $t = 0$  are plotted as a function of  $\Delta T$  in Fig. 5(b), decreasing asymmetrically with increasing  $|\Delta T|$ . The EPSC amplitudes gradually decrease from 520 nA to 217 nA when  $\Delta T$  changes from 0 to -150 ms, while no significant change is observed when  $\Delta T > 0$ .

When  $\Delta T > 0$  (i.e., spike  $P_1$  triggers after spike  $P_2$ ), the EPSC amplitude measured at  $t = 0$  equals the EPSC individually triggered by  $P_2$ . When  $\Delta T < 0$  (i.e., spike  $P_1$  triggers before spike  $P_2$ ), the EPSC amplitude measured at  $t = 0$  is the summed current of EPSC<sub>1</sub> triggered by  $P_1$  and EPSC<sub>2</sub> triggered by  $P_2$ . Therefore, the summed current can be written as  $\kappa' A_1 \exp[-(-\Delta T/\tau)^\beta] + A_2$ , where  $\kappa'$  is a factor related to the effective residual proton concentration (triggered by  $P_1$ ) contributing to the summed current, and  $A_1 = \gamma_1 [1 - \exp(-T/\tau_{DL})]$  and  $A_2 = \gamma_2 [1 - \exp(-T/\tau_{DL})]$  are the EPSC amplitudes individually triggered by  $P_1$  and  $P_2$ , respectively.

In conclusion, the EPSC amplitude at  $t = 0$  can be determined by:

$$A(\Delta T)_{t=0} = \begin{cases} \kappa' A_1 \exp \left[ - \left( -\frac{\Delta T}{\tau} \right)^\beta \right] + A_2 & \Delta T \leq 0 \\ A_2 & \Delta T > 0 \end{cases}$$

The EPSC amplitudes measured at  $t = 0$  are plotted as a function of time interval ( $\Delta T$ ) in Fig. 5(b). Furthermore, the measured EPSC amplitudes are fitted using Eq. (11) and plotted as a blue solid line in Fig. 5(b). The parameters  $\kappa'$ ,  $A_1$ , and  $A_2$  are estimated to be 0.92, 332 nA, and 217 nA, respectively. Again, we find that the fitting curve matches the experimental results very well, indicating the validity of the proposed model.

## Conclusion

In summary, we have experimentally demonstrated synaptic plasticity including PPF and spatiotemporal-correlated dynamic logic in an organic/inorganic hybrid artificial synapse based on an IZO-based EDL transistor gated by methylcellulose electrolyte film. A model based on proton-related electric-double-layer modulation and a stretched-exponential decay function was proposed, and the theoretical simulation results are consistent with the measured synaptic behaviors. Our results may provide useful guidelines for artificial synapses and brain-inspired neuromorphic systems.

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