Supplementary Materials for

One-dimensional organic artificial multi-synapses enabling electronic textile neural network for wearable neuromorphic applications

Seonggil Ham, Minji Kang, Seonghoon Jang, Jingon Jang, Sanghyeon Choi, Tae-Wook Kim*, Gunuk Wang*

*Corresponding author. Email: twk@jbnu.ac.kr (T.-W.K.); gunukwang@korea.ac.kr (G.W.)

Published 10 July 2020, Sci. Adv. 6, eaba1178 (2020)
DOI: 10.1126/sciadv.aba1178

This PDF file includes:

Figs. S1 to S18
Tables S1 to S3
Supplementary note
Fig. S1. Surface morphology and capacitance of P(VDF-TrFE) film. (A) Top-view SEM images of the P(VDF-TrFE)-coated Ag wire. (B) Dielectric capacitance with respect to the position of the Ag wire. The capacitance per area was measured at four positions with 10-mm intervals along the Ag wire. The average capacitance per area was measured to be 4.75 ± 0.3 nF/cm². The similar capacitance values indicate that the P(VDF-TrFE) film was uniformly coated on the Ag wire.
Fig. S2. The $I_{DS}$–$V_G$ and $I_{GS}$–$V_G$ characteristics obtained under various $V_G$ double sweep ranges (from ±10 to ±40 V) at $V_{DS} = -10$ V
Fig. S3. Switching characteristics under various tangled conditions. (A) Cross-sectional schematics of the coiled 1D organic multi-synapses on a tube with a radius of $R$. (B) $I_{DS}-V_G$ curves obtained during $V_G$ double sweeps at the range from ±10 to ±40 V at $V_{DS} = -10$ V without the strain. (C–F) $I_{DS}-V_G$ curves of the coiled 1D organic multi-synapses on tubes of various forms with different radius ($R$) of 4 mm (C), 3 mm (D), 1 mm (E), and 0.6 mm (F). Especially, in the case of $D \leq 2$ mm ($R \leq 1$ mm), the ON-OFF ratio at $V_G = 0$ was further decreased as compared with its initial ratio (B). Since the P(VDF-TrFE) is very tolerant to mechanical strain (26), we suspect that the mechanical property of pentacene may cause switching degradation. In fact, thermally evaporated pentacene film has a tensile modulus of ~15 GPa, which is an order of magnitude larger than those of other polymeric materials (27). This could lead to cracks and/or delamination at electrode/pentacene and pentacene/P(VDF-TrFE) interfaces when a large mechanical strain is applied (28). We should note that the device shows a clockwise switching operation, which indicates that the switching is mainly due to the
polarization change of the ferroelectric domain in the P(VDF-TrFE) (29). (Photo credit: Tae-Wook Kim, Jeonbuk National University.)
Fig. S4. Changes in the synaptic weight ($\Delta w$) with respect to the relative timing difference ($\Delta t$) between the single presynaptic ($t_{\text{pre}}$) and postsynaptic pulse ($t_{\text{post}}$). −25 and +25 V were applied to the single presynaptic and postsynaptic pulses, respectively, for 500 ms. When the absolute $\Delta t$ was increased, the $\Delta w$ decreased from ~70% to ~40%. This is because the temporal summation of the input pulse spikes from both neurons was weakened. Note that since the $I_{\text{PSC}}$ can be determined by the relative voltage (or timing) difference between the presynaptic spike $V_G$ and postsynaptic spike $V_{\text{DS}}$, the applied voltage schemes should be different according to the updated and non-updated synaptic cells in the NOR-type array (36). In practice, $\Delta w$ can be modulated by the conductance difference between adjacent synapses that can be changed by the magnitude of the applying effective voltage pulses.
Fig. S5. Retention characteristics of the 1D multi-synapses. (A) Retention characteristics of the completed LTP/LTD state for fabricated 1D multi-synapses during $10^4$ s. (B) Retention characteristics of the selected LTP state during $10^3$ s. Each $I_{PSC}$ corresponding to the LTP state is triggered by different number of repeated potentiating pulses (from 10 to 30 pulses) of $V_G = -30$ V for 500 ms.
Fig. S6. Recognition accuracy for MNIST patterns with respect to the elapsed time after the completion of the training considering the conductance relaxation. The left insets show a reshaped 28 × 28 contour image of the digit “0” from w for 0 s and 31,000 s, respectively. The right inset shows the confusion matrix for a classification test after ~31,000 s. ~90% accuracy had been well maintained at the initial elapsed time, but it abruptly decreased to ~34% if time reached to ~3 × 10^4 s. We suspect that this is because all $G^+$ and $G^-$ matrices in the array converge to $G_{min}$, resulting in the collapse of each synaptic weight.

In order to investigate the impact of the $I_{PSC}$ relaxation on the learning accuracy, each decay degree corresponding to each LTP state (Fig. S5) was obtained from the exponential fitting using a simple equation of $G(t) = A_0 + B_0 e^{-t/t_0}$, then we converted each parameter as a function of $G$ of the synaptic cell. Note that we assumed that this parameter affects linearly to all conductance range from $G_{min}$ to $G_{max}$. Considering this assumption, all the synaptic weight values after the completion of the training will start to be differently decreased depending on the decay degree of $G$. In the simulation, we modulated all the synaptic weight values after training epoch in the function of elapsed time considering by the decay degree. Then, we calculated the recognition accuracy for MNIST test patterns. The details of simulation for pattern recognition are described in Fig. S11, Fig. S12, and Fig. S15.
Fig. S7. Nonlinearity ($N_L$) and dynamic range. (A–E) $N_L$ and dynamic range with respect to $V_G$ ranges. $N_L$ and dynamic range in w updating based on LTP (black dots) and LTD (red dots) at different $V_G$ values ranging from $\pm 20$ to $\pm 40$ V. The $N_L$ can be calculated using the following equation (41):

$$N_L = \frac{\max |G_p(n) - G_d(n)|}{G_{\text{max}} - G_{\text{min}}}, \quad n = 1, 2, 3, \ldots, \text{or 80},$$

where $G(n)$ represents the conductance value after the $n^{\text{th}}$ programming pulse; the subscripts $p$ and $d$ represent potentiation and depression, respectively; $G_{\text{max}}$ and $G_{\text{min}}$ represent the maximum and minimum conductance, respectively; and the ratio $G_{\text{max}}/G_{\text{min}}$ represents the dynamic range. The $N_L$ ranged from 0 to 1. $N_L = 0$ indicates that the $w$ updates for LTP and LTD are identical and have perfect linear relationships. (F) Ratio of the dynamic range to $N_L$ for different $V_G$ values ranging from $\pm 20$ to $\pm 40$ V. At $V_G = \pm 30$ V, the maximum ratio was observed; thus, this was selected as the optimal presynaptic pulse for a high learning accuracy.
Fig. S8. (A–E) LTP and LTD of the $I_{PSC}$ for $V_G = \pm35$ V and $\pm30$ V of 500 ms based on 5 different synaptic cells.
Fig. S9. Calculated bending strain at different bending radii. (A) Schematic of the 1D multi-synapses placed on the PET substrate with a bending radius $R$. (B) Plot of the bending strain $\varepsilon$ versus the bending radius $R$ for the 1D artificial multi-synapses on the PET substrate. The bending strain $\varepsilon$ can be estimated using the equation in (B) (44), where $d_{\text{substrate}} \approx 200 \, \mu\text{m}$, $d_{\text{Ag wire}} \approx 100 \, \mu\text{m}$, and $R = \infty$, 5, or 2.5 mm.
Fig. S10. Switching characteristics of 60 cells in 10 1D multi-synapses. (A) Schematic of all 60 cells in 10 multi-synapses. (B) $I_{DS}-V_G$ curves of the 60 cells obtained during $V_G$ double sweep ranging from $+30$ V to $-30$ V at $V_{DS} = -10$ V.
Fig. S11. Analog potentiation and depression of the conductance at $V_G = \pm 30$ V for 500 ms. The solid red line shows the fitting results of $\Delta G = \Delta I_{\text{PSC}}/V_{DS}$ for LTP and LTD obtained using the fitting parameters (in Table S1).

Because the $\Delta G$ can determine the degree of the $w$ update, each $\Delta G$ value for LTP and LTD can be fitted by the following equation (41):

$$
\Delta G_p = a_p + b_p e^{-c_p (G_{\text{max}} - G_{\text{min}})} \quad \text{and} \quad \Delta G_d = a_d + b_d e^{-c_d (G_{\text{max}} - G_{\text{min}})},
$$

where $\Delta G_p$ and $\Delta G_d$ represent the $\Delta G$ for LTP and LTD, respectively; $a_p$, $b_p$, $c_p$, $a_d$, $b_d$, and $c_d$ are the fitting parameters used to estimate $\Delta G_p$ and $\Delta G_d$; and the subscripts $p$ and $d$ represent potentiation and depression, respectively. The available range of $\Delta w$ is constrained in the range of the $\Delta G$ (i.e., from $G_{\text{min}}$ to $G_{\text{max}}$). The fitting parameters are presented in Table S1. Using these parameters, we performed the MNIST pattern-recognition simulation.
**Fig. S12.** Recognition accuracy for the MNIST patterns at $V_G = \pm 30$ V as a function of the learning phase. After 10 epochs, ~90% accuracy was achieved. Insets show reshaped $28 \times 28$ contour images of the final $w$ corresponding to “0”, “1”, “3”, and “8” according to the number of epochs (0, 1, and 10).

It is known that the maximum recognition accuracy of single-layer neural networks is ~88% (46). In this case, they assumed that the network has the limited 7,850 free parameters including 10 biases. However, the main difference between our simulation and the literature is that one weight in our case is defined as the difference in conductance of two synaptic cells. In our case, each $G_{ij}^+$ and $G_{ij}^-$ corresponds to the free parameter, hence the network can have 15,680 ($2 \times 7,840$) free parameters that is twice than the previous literature. Since increasing the number of free parameters or hidden layers in the network can lead to improving recognition accuracy (47), we think that the increased number of free parameters is the main cause of higher accuracy. Note that ~92% accuracy was reported based on the same definition of synaptic weight (48).
Fig. S13. Cell-to-cell variation for LTP/LTD functions based on 8 different synaptic cells. (A–H) LTP and LTD of the $I_{PSC}$ for ±30 V of 500 ms based on 8 different synaptic cells. (I) Statistical LTP and LTD of the $I_{PSC}$ for ±30 V of 500 ms.
Fig. S14. Comparison of the recognition accuracy for the MNIST patterns with and without cell-to-cell variation. We assumed that selected five cells were randomly positioned on the neural network array (Fig. S13).
Fig. S15. Single-layer neural network for MNIST pattern recognition. (A) Constituents of a single-layer neural network for the MNIST pattern-recognition process. (B) Diagram of the crossbar array mapped to the single-layer neural network.

Figure S15A shows the components of a single-layer network for the typical MNIST pattern-recognition process. In the simple single-layer network, 784 input neurons are fully connected to 10 output neurons by 7,840 synaptic weights each. As shown in Fig. S15A, the 784 pixels (28 \times 28 pixels) of the MNIST handwritten digit image are connected to the input neurons in order. Then, the input signal \( x_i \) corresponding to each pixel is multiplied by the corresponding synaptic weight \( w_{i,j} \) and summed with the other weighted inputs at the \( j^{th} \) output neurons \( \sum w_{i,j} x_i \), where the subscripts \( i \) and \( j \) indicate that the corresponding constituent is connected to the \( i^{th} \) input neuron and \( j^{th} \) output neuron, respectively. Note that \( w_{i,j} \) is determined by the conductance difference of neighboring synapses; i.e., \( w_{i,j} = G_{ij}^+ - G_{ij}^- \), where \( G_{ij}^+ \) and \( G_{ij}^- \) represent the conductance values for a pair of adjacent synapses. After the multiply–accumulate operation, a sigmoid activation function is applied, resulting in the output signal \( y = f(\sum w_{i,j} x_i) \), where \( f = (1 + e^{-x})^{-1} \). The neural network can be implemented easily by a 1D multi-synapse array, which allows neural processing, as shown in Fig. S15B. The learning model of the single-layer neural network is based on a simple supervised learning algorithm, i.e., stochastic gradient ascent/descent learning algorithm, which was described in detail in our previous reports (24, 53). In the learning process, the hand-written digit images ranging from “0” to “9” are fed to the input neurons in random order. Then, the error value \( e_j \) is calculated according to the difference between the output \( y_j \) and the target value \( t_j \). Finally, the weight is updated according to the delta rule, i.e., \( \Delta w_{i,j} = \eta \cdot e_j x_i \), where \( \eta \) represents the learning rate. The recognition accuracy is estimated by employing 10,000 inference patterns that are not used in the learning process. The entire learning process is described in the flowchart of Fig. S16.
Fig. S16. Flowchart for one epoch of the learning process based on the stochastic gradient ascent/descent learning algorithm.
Fig. S17. (A–C) Three waveforms for "N" class. Note that they show different periods (1T) of heart rates and normalized amplitudes. For 1T, (A), (B), and (C) show 848, 1168, and 696 ms, respectively. For the normalized amplitudes, 0.256, 0.367, and 0.057 are set for each class.
Fig. S18. Recognition accuracy for MNIST (A) and ECG (B) patterns with respect to the number of learning epochs. The insets show the confusion matrix for a classification test involving 10,000 MNIST after 10 epochs and 800 ECG patterns after 50 epochs. In the recognition simulation for MNIST and ECG patterns, we excluded all the synaptic characteristics of the device and used only arithmetic operation. The maximum accuracies are ~91% and ~71% for MNIST and ECG patterns, respectively, which are slightly higher than the simulation result reflecting the device information (~90% for MNIST and ~70% for ECG).
Potentiation

\[ 6.44 \times 10^{-12} - 6.04 \times 10^{-12} + 6.10 \]

\[ 1.32 \times 10^{-10} \]

Depression

\[ 1.41 \times 10^{-10} - 1.68 \times 10^{-10} + 0.17 \]

\[ 1.12 \times 10^{-12} \]

Table S1. Fitting parameters for 30 LTP and 30 LTD at \( V_G = \pm 30 \text{ V} \).
| Category (AAMI EC57 standard) | Annotations |
|-------------------------------|-------------|
| **N** *(Any heartbeat not in the S, V, F, or Q class)* | • Normal  
• Left/right bundle branch block  
• Atrial escape  
• Nodal escape |
| **S** *(Supraventricular ectopic beat)* | • Atrial premature  
• Aberrant atrial premature  
• Nodal premature  
• Supra-ventricular premature |
| **V** *(Ventricular ectopic beat)* | • Premature ventricular contraction  
• Ventricular escape |
| **F** *(Fusion beat)* | • Fusion of ventricular and normal |
| **Q** *(Unknown beat)* | • Paced  
• Fusion of paced and normal  
• Unclassifiable |

Table S2. Mappings between the beat annotations of the PhysioNet MIT-BIH Arrhythmia dataset and the Advancement of Medical Instrumentation (AAMI) EC57 standard. The original MIT-BIH dataset comprises ECG recordings from 47 respective subjects, which are annotated by at least two cardiologists. Each annotated beat was classified to create five different beat categories in accordance with the Association for the Advancement of Medical Instrumentation (AAMI) EC57 standard (55). Note that for higher accuracy for ECG patterns, the deeper neural network equipped with adequate feature extractors, not a single-layer neural network array, can be considered. Recently, the cardiologist-level detection and classification performance for ECG patterns had been achieved using the software-based deep neural networks such as a recurrent neural network (RNN) or/and convolutional neural network (CNN) with multiple convolution layers acting as feature extractors (56–57).
### Table S3

Fitting parameters for the LTP and LTD of the 1D devices under different bending conditions ($R = \infty$, 5, and 2.5 mm).

| $R = \infty$ | a          | b        | c  | $G_{\text{max}}$ (S) | $G_{\text{min}}$ (S) |
|-------------|------------|----------|----|----------------------|----------------------|
| Potentiation | $1.19 \times 10^{-12}$ | 0.27141  | 122 | $4.80 \times 10^{-11}$ | $4.41 \times 10^{-12}$ |
| Depression  | $-1.68 \times 10^{-12}$ | $-2.48 \times 10^{-11}$ | 2.68 |                      |                      |

| $R = 5 \text{ mm}$ | a          | b        | c  | $G_{\text{max}}$ (S) | $G_{\text{min}}$ (S) |
|-------------------|------------|----------|----|----------------------|----------------------|
| Potentiation      | $1.11 \times 10^{-12}$ | $5.39 \times 10^{-12}$ | 7.98 | $4.79 \times 10^{-11}$ | $3.64 \times 10^{-11}$ |
| Depression        | $2.24 \times 10^{-12}$ | $-2.40 \times 10^{-11}$ | 2.38 |                      |                      |

| $R = 2.5 \text{ mm}$ | a          | b        | c  | $G_{\text{max}}$ (S) | $G_{\text{min}}$ (S) |
|---------------------|------------|----------|----|----------------------|----------------------|
| Potentiation        | $1.30 \times 10^{-12}$ | $6.15 \times 10^{-12}$ | 8.38 | $5.37 \times 10^{-11}$ | $2.61 \times 10^{-12}$ |
| Depression          | $5.86 \times 10^{-12}$ | $-3.50 \times 10^{-11}$ | 1.81 |                      |                      |
Supplementary note

Challenges for realizing the circuit level of ANNs.

There are many challenges and issues for realizing the circuit level of ANNs if the 1D synaptic cell having quite low $I_{PSC}$ (~nA) is used. One of the main obstacles is to directly detect the synaptic weight itself through the peripheral circuitry. If the maximum conductance level of the active cell is ~nS, it is quite difficult to distinguish from the circuit-noise signal naturally generated from surroundings, which can limit the maximum array size. Furthermore, a general circuit board functioning artificial neurons itself cannot precisely detect this conductance level. In this sense, it is much hard to detect the differences between the intermediated states, so that the degree of the weight update can be limited. These all could limit the implementation of learning process. In such a case, sense amplifiers need to be connected to all post-neurons in order to convert the output current into voltage signal using a voltage-mode ADC, but this would make the neuromorphic electronic system more complex at the same time. The low conductance of the synaptic cell might affect the reading process (e.x., addressing process) of the state because the sense amplifier is operated only when the input signal transmits over the threshold. In addition, we think that the low-conducting charge transfer would be easily disturbed by the electrode word/bit lines and surroundings, leading to slow addressing. Lastly, when some of the cells in the array become the electrical short circuit, the output signals in the array are severely affected and disturbed because some integrated $I_{PSC}$ is mainly determined by the electrical-short synaptic cell. Therefore, we should note that it is essential to increase the conductance level of our 1D multi-synapses. In fact, it can be improved by the change of the junction dimension, the use of thinner ferroelectric and high-mobility semiconductor layers.