

All-solid-state Ion Synaptic Transistor for Wafer-scale Integration with Electrolyte of a Nanoscale Thickness

Ji-Man Yu

Korea Advanced Institute of Science and Technology

Chungryeol Lee

Korea Advanced Institute of Science and Technology

Da-Jin Kim

Korea Advanced Institute of Science and Technology

Hongkeun Park

Korea Advanced Institute of Science and Technology <https://orcid.org/0000-0002-4763-4487>

Joon-Kyu Han

Korea Advanced Institute of Science and Technology

Jae Hur

Georgia Institute of Technology

Jin-Ki Kim

Korea Advanced Institute of Science and Technology

Myung-Su Kim

Korea Advanced Institute of Science and Technology

Myungsoo Seo

Korea Advanced Institute of Science and Technology

Sung Gap Im

Korea Advanced Institute of Science and Technology

Yang-Kyu Choi (✉ ykchoi@ee.kaist.ac.kr)

Korea Advanced Institute of Science and Technology <https://orcid.org/0000-0001-5480-7027>

Article

Keywords: all-solid-state, artificial neural network (ANN), deep neural network (DNN), electrolyte-gated synaptic transistor (EGST), initiated chemical vapor deposition (iCVD), neuromorphic system, polyethylene glycol di-methacrylate (PEGDMA), synapse device, wafer-scale

Posted Date: October 9th, 2020

DOI: <https://doi.org/10.21203/rs.3.rs-73972/v1>

License:  This work is licensed under a Creative Commons Attribution 4.0 International License.

[Read Full License](#)

Version of Record: A version of this preprint was published at Advanced Functional Materials on March 30th, 2021. See the published version at <https://doi.org/10.1002/adfm.202010971>.

1 **All-solid-state Ion Synaptic Transistor**
2 **for Wafer-scale Integration with**
3 **Electrolyte of a Nanoscale Thickness**
4

5
6
7 *Ji-Man Yu,¹ Chungryeol Lee,² Da-Jin Kim,¹ Hongkeun Park², Joon-Kyu Han,¹ Jae Hur,^{1,3} Jin-*
8 *Ki Kim,¹ Myung-Su Kim,¹ Myungsoo Seo,¹ Sung Gap Im,² and Yang-Kyu Choi^{1*}*

9
10
11
12 *¹ School of Electrical Engineering, Korea Advanced Institute of Science and Technology*
13 *(KAIST), 291 Daehak-ro, Yuseong-gu, Daejeon 34141, Republic of Korea*

14 *² Department of Chemical and Biomolecular Engineering, Korea Advanced Institute of*
15 *Science and Technology (KAIST), 291 Daehak-ro, Yuseong-gu, Daejeon 34141, Republic of*
16 *Korea*

17 *³ School of Electrical and Computer Engineering, Georgia Institute of Technology, 791*
18 *Atlantic Dr NW, Atlanta, GA 30332, United States of America*

19
20
21 *Authors to whom correspondence should be addressed.

22 Email address: ykchoi@ee.kaist.ac.kr

23 **Abstract**

24 Neuromorphic hardware computing is a promising alternative to von Neumann computing by
25 virtue of its parallel computation, and low power consumption. To implement neuromorphic
26 hardware based on deep neural network (DNN), a number of synaptic devices should be
27 interconnected with neuron devices. For ideal hardware DNN, not only scalability and low
28 power consumption, but also a linear and symmetric conductance change with the large
29 number of conductance levels are required. Here an all-solid-state polymer electrolyte-gated
30 synaptic transistor (pEGST) was fabricated on an entire silicon wafer with CMOS
31 microfabrication and initiated chemical vapor deposition (iCVD) process. The pEGST showed
32 good linearity as well as symmetry in potentiation and depression, conductance levels up to
33 8,192, and low switching energy smaller than 20 fJ/pulse. Selected 128 levels from 8,192 used
34 to identify handwritten digits in the MNIST database with the aid of a multilayer perceptron,
35 resulting in a recognition rate of 91.7 %.

36

37 **Keywords:** all-solid-state, artificial neural network (ANN), deep neural network (DNN),
38 electrolyte-gated synaptic transistor (EGST), initiated chemical vapor deposition (iCVD),
39 neuromorphic system, polyethylene glycol di-methacrylate (pEGDMA), synapse device,
40 wafer-scale

41

42

43

44

45

46

47 **Introduction**

48 Artificial neural networks (ANN), have been intensively explored with the requirements of
49 many applications¹⁻⁵. Among various kinds of ANN, the deep neural network (DNN) is a well-
50 studied training method, and hardware electronic devices have been actively investigated in
51 efforts to realize DNN with low-power consumption. In the hardware DNN, a number of
52 synaptic devices need to be interconnected with multiple neuron devices, to process cognitive
53 tasks such as image^{6,7} and speech recognition⁸. Therefore, mass production of synaptic device
54 at wafer-scale fabrication is inevitable. Concurrently, to achieve high performance, linear and
55 symmetric conductance changes are required⁹⁻¹¹.

56 Emerging 2-terminal devices are promising candidates due to cost-efficiency, and
57 scalability¹²⁻¹⁵. However, they suffer from non-linear and asymmetric conductance changes. In
58 addition, additional selector components are needed to pick up the target cell, without a sneak-
59 path^{15,16}. Alternatively, 3-terminal synaptic transistors have attracted interests to resolve the
60 aforementioned problems. They have advantages of low power consumption without a sneak-
61 path by gate modulation, and improved controllability of the synaptic weight¹⁷⁻¹⁹. It also
62 provides a parallel ‘write’ and ‘read’ operations²⁰. However, a synaptic transistor that shows
63 excellent linearity, symmetry with a high dynamic range (HDR), and sufficiently low energy
64 consumption, is still missing.

65 The electrolyte-gated transistor (EGT) may be an advanced candidate to address these issues.
66 The EGT is controlled by ions in an electrolyte-gate. The approach has attracted considerable
67 attention because of its similarity to an actual biological system, where the membrane potential
68 is governed by ions such as Ca^{2+} , Na^+ , and K^+ ²¹⁻²³. In EGTs, a gate dielectric in a conventional
69 MOSFET is replaced by electrolytes with movable ions (*e.g.*, H^+ , Li^+ , O^{2-})²⁴⁻³⁰. When gate
70 voltage is applied, movable ions form an electric double layer (EDL). It causes hysteresis, and
71 adjusts the synaptic weight. EGT-based synapses have shown a relatively linear conductance

72 change because of continuous ion migration inside the electrolyte²⁸. However, scalability,
73 wafer-scale microfabrication, and reliability have been issues, because of their reliance on a
74 liquid-based electrolyte and mechanical exfoliated channels. Such Limitation affects the power
75 consumption and switching speed²⁰, and wafer-scale fabrication of EGTs is essential for the
76 architecture of neuromorphic systems where more than 10^{15} synapses may be used to mimic a
77 human brain³¹.

78 In this work, we demonstrate a polymer electrolyte-gated synaptic transistor (pEGST)
79 composed of an all-solid-state polymer electrolyte and a silicon-channel. For the fabrication of
80 the pEGST, an ultra-thin electrolyte was deposited by vapor phase using the initiated chemical
81 vapor deposition (iCVD). This is a solvent-free, one-step polymerization for generating high-
82 performance polymer electrolyte films³². Protons (H^+) incorporated in the polymer electrolyte
83 were selected for CMOS compatibility. Unlike previous EGTs using an ionic liquid or ionic
84 gel, the pEGST is scalable to nanoscale thickness with the aid of iCVD. While previous EGTs
85 have been fabricated in a single device, the pEGST was manufactured in wafer-scale. The
86 pEGST showed high-performance, with linear and symmetric weight update consuming low
87 energy, less than 20 fJ/pulse. Remarkably, the multi-levels of conductance were achieved, up
88 to 13 bits. The attained MNIST recognition rate was 91.7%.

89

90

91

92

93

94

95

96

97 **Results and discussion**

98 **Structure and fabrication of pEGST**

99 **Figure 1a** provides a schematic of the proposed pEGST and depicts the chemical structure of
100 polyethylene glycol di-methacrylate (pEGDMA), which was used as a solid-state gate
101 dielectric to harness the electrolyte's properties. **Figure 1b** shows a transmission electron
102 microscopy (TEM) image of the cross-sectional structure, to identify each layer of the
103 fabricated pEGST. The gate dielectrics are composed of three layers: formed aluminum oxide
104 of 3 nm on top, pEGDMA of 20 nm in the middle, and thermally grown SiO₂ of 2 nm on the
105 bottom, as shown in a cross-sectional TEM image in **Figure 3a**. The gate electrode is made of
106 aluminum and its thickness is 300 nm. The nominal gate length (L_G) of the pEGST was 7 μm
107 and its channel width (W_{ch}) was 50 μm . It was presumed that the 3 nm of aluminum oxide
108 between the Al gate and the pEGDMA was formed by aluminum anodizing during ion
109 migration when gate bias was applied. An interfacial layer of the SiO₂ between the pEGDMA
110 and the silicon channel, was intentionally grown with thermal oxidation to minimize interface
111 trap density, and to suppress gate leakage current during switching to control synaptic weights.

112 **Figure 1c** shows the key fabrication steps for the pEGST, which was fabricated on a p-type
113 (100) 4-inch silicon wafer. For device-to-device isolation, SiO₂ of 1 μm served as a field oxide
114 was thermally grown. For gate-last process, source and drain (S/D) were formed by high dose
115 phosphorous implantation with a dummy gate. Rapid thermal annealing (RTA) in N₂ ambient
116 was employed to activate the dopant. Gate dielectrics composed of the interfacial thermal oxide
117 and the pEGDMA were sequentially stacked after the removal of dummy gate. The Al gate was
118 deposited with the aid of DC magnetron sputter. The gate electrode was patterned by
119 conventional photo-lithography and subsequent etching. Details of the step-by-step fabrication
120 and iCVD processes are described in Supplementary Information 1 and 2.

121 To ensure highly uniform pEGDMA, vapor-phase iCVD was employed^{33,34}. Unlike the liquid

122 or ionic gel-type electrolyte typically used in conventional EGST, the wafer-scale pEGDMA
123 deposited via the iCVD process can be fully realized with matured complementary metal-
124 oxide-semiconductor (CMOS) technology. **Figure 1d** shows a schematic of the iCVD chamber
125 used for the polymerization of the ultra-thin polymer films. The iCVD process is a solvent-free
126 vapor-phase process for depositing various kinds of high-purity polymer films with outstanding
127 uniformity, and conformality. The iCVD polymerization process can be briefly summarized as
128 follows: i) injection of monomer and initiator, ii) thermal decomposition of the initiator to form
129 radicals, iii) collision between radicals and monomers, and iv) free radical formation of the
130 polymer thin films. The effectiveness of surface-growing mechanism was demonstrated on an
131 8-inch wafer via at low temperature ($\sim 40\text{ }^{\circ}\text{C}$)³⁵. A chemical structure of the deposited
132 pEGDMA film was analyzed by Fourier transform infrared (FT-IR) spectroscopy, as described
133 in the Supplementary Information 3³⁶.

134 **Figure 1e** shows the fabricated 4-inch Si wafer demonstrating the wafer-scale
135 manufacturability of the pEGSTs, containing 30 dies. The die size is $1\times 1\text{ cm}^2$, as shown in the
136 inset in **Figure 1e**. CMOS compatibility and the iCVD electrolyte deposition method also leave
137 plenty of room for down-scaling with future structural innovation (See Supplementary
138 Information 4).

139

140 **Electrical characterization of the pEGST**

141 **Figure 2a** shows the measured I_D - V_G curve of the pEGST at a drain voltage (V_D) of 50 mV.
142 The counter-clockwise hysteresis of protons in the pEGDMA was observed with double sweeps
143 of the V_G from -6 V to +6 V forward and from +6 V to -6 V backward. The counter-clockwise
144 direction of the hysteresis, electrolyte characteristics, and ion mass spectrometry supports the
145 conclusion that the closed loop is not produce by electrons tunneling through the gate
146 dielectrics, as is usually observed in a conventional charge-trap memory-based synaptic device,

147 but by positive ions in the electrolyte-based synaptic device.

148 This movement of positive ions in the solid-phase electrolyte of the pEGST, exhibits
149 horizontally wide hysteresis of back-sweep current (BSC) with a vertical ratio of on-state
150 current (I_{ON}) to off-state current (I_{OFF}) of 10^4 at $V_G = 0$ V in the I_D - V_G curve. It should be noted
151 that a wide voltage window of hysteresis is preferred for multiple-level synaptic functionality,
152 because it allows a broad range of channel conductance (G_{DS}) changes across the source (S)
153 and drain (D) with an applied gate electric field.

154 Note that wafer-scale fabrication is essential for chip-based neuromorphic systems where
155 more than a thousand synaptic devices need to be connected to one neuron device. Unlike
156 previously reported EGST that could not be fabricated at wafer-scale owing to liquid processes
157 that use ion gel and ionic liquid electrolytes, the reliable wafer-scale pEGSTs could be
158 fabricated as an all-solid-state thin film using the iCVD and CMOS-compatible
159 microfabrication (See Supplementary Information 5).

160 It is particularly essential that the chip-level system is capable of *in situ* on-chip learning via
161 weight update with on-the-fly training. **Figure 2b** shows the device-to-device variability of the
162 fabricated pEGSTs across the 4-inch silicon wafer. The averaged threshold voltage $V_{T,forward}$ of
163 1.25 V and $V_{T,backward}$ of -1.24 V is uniform. The averaged hysteresis window of 2.49 V, which
164 is defined as $\Delta V_T = V_{T,forward} - V_{T,backward}$, is also uniform. **In both figures, error bars represent**
165 **standard deviations.** These data were extracted from four randomly selected devices from 15
166 chips (for a total 60 pEGSTs) across the entire 4-inch wafer. As shown in **Figure 2c** and **d**, each
167 I_D is modulated by the number of identical pulse widths applied to the gate electrode, which
168 are distinctive for potentiation and depression (P/D), respectively. Each distinguishable I_D
169 corresponds to a weighting state during the P/D in the multi-level states for synaptic operations.

170

171 **Electrolyte characteristics of pEGDMA**

172 To clarify the origin of the counter-clockwise hysteresis in the pEGST, the dynamic behaviors
173 of the pEGDMA used as the electrolyte gate dielectric were analyzed by gate capacitance-
174 frequency ($C-f$) and phase angle-frequency ($\theta-f$) characterization. As shown in **Figure 3b**, a
175 bell-shaped profile of θ was measured³⁷. In general, θ in an ionic relaxation dominant region
176 is known to be larger than -45° , however, it was smaller than that in the pEGST. This was
177 caused by the thermally grown interfacial SiO_2 layer. SiO_2 is an excellent insulator, which
178 causes the capacitive dominance from the dipole relaxation to compete with the resistive
179 dominance from the ionic relaxation. The anodized aluminum oxide can additionally minimize
180 the θ value because of its dielectric characteristics³⁸.

181 **Figure 3c** shows a schematic of the ion distribution under equilibrium without applied gate
182 voltage. When a positive voltage is applied to the gate, H^+ in the pEGDMA move closer to the
183 channel, as shown in **Figure 3d**. When negative voltage is applied to the gate, it moves farther
184 from the channel, as shown in **Figure 3e**. A depth profile of protons from the Si-channel via
185 the interfacial SiO_2 to inside the pEGDMA, was analyzed using time-of-flight secondary ion
186 mass spectrometry (ToF SIMS). It confirmed that the concentration of protons in the doped
187 sample was larger than in the undoped one, and the proton concentration becomes higher
188 toward the gate electrode after applying negative voltage at the gate electrode. Other relevant
189 ion concentrations were also analyzed and compared with the proton concentration, as shown
190 in Supplementary Information 6. The bias dependent mobile ions in the polymer electrolyte
191 properly modulate the G_{DS} by controlling the density of carriers in the silicon channel. In
192 addition, the pEGDMA including protons induce a wide enough hysteresis to realize synaptic
193 characteristics in the hardware-based analog DNN.

194 The $I_{\text{D}}-V_{\text{G}}$ characteristics of the pEGST were also investigated at high temperature (T) over
195 the range of 25°C to 180°C (see Supplementary Information 3). Hysteresis voltage (ΔV_{T})
196 increased as T increased (**Figure S5**). This tendency is caused by accelerated ion migration in

197 the polymer electrolyte at high T . In addition, it was confirmed by FT-IR spectroscopy that the
198 chemical structures in the polymer electrolyte film were not changed even after high
199 temperature treatment at 180 °C (**Figure S6**).

200

201 **Synapse characteristics of pEGST for analog deep neural networks**

202 In the pEGST, the G_{DS} change with each update in synaptic weight was characterized to
203 estimate the linearity and asymmetric ratio (AR), which are important metrics for evaluating
204 the accuracy of a pattern recognition rate in neural network simulations. **Figure 4a** shows the
205 identical pulse scheme used, with constant voltage amplitude and time duration, to control P/D
206 and read operations. An optimal pulse scheme is $V_{pot} = +4$ V with 10 ms for the potentiation
207 and $V_{dep} = -5.3$ V with 10 ms for the depression. The read voltage used to measure the G_{DS}
208 between each P/D pulse was set to 0.1 V, which was small enough to minimize the effect of ion
209 drift in the pEGDMA. Thus, the reading operation did not influence on the G_{DS} determined by
210 the programming and the erasing. Energy consumption for the potentiation and depression was
211 less than 20 fJ/pulse (See Supplementary Information 7). This is comparable to the energy
212 consumption of a biological synapse in the human brain (10 fJ/spike)³⁹, and it is advantageous
213 in terms of energy consumption in on-chip learning.

214 A G_{DS} of 8192 ($=2^{13}$, 13 bits) levels was achieved for each P/D. This is the largest number
215 of multi-levels reported with HDR ever reported, as shown in **Figure 4b**. Here the dynamic
216 range is a conductance window in between the minimum and the maximum conductance. A
217 silicon channel MOSFET with a well-controlled off current can achieve high on/off switching
218 characteristics. Such more than thousands of multi-states conductances are the result of
219 harmonizing the continuous distribution of protons inside the polymer electrolyte with the
220 characteristics of the silicon channel MOSFET. Note that its nominal ratio of on-state I_D to off-
221 state I_D is larger than 10^5 . This feature of the HDR is one of the reasons why the ion

222 incorporated solid-state electrolyte needs to be implemented on the silicon channel MOSFET.

223 This HDR is attractive for improving the recognition rate by selecting highly linearized
224 fraction of conductance changes from the entire P/D region. It is also preferable for customizing
225 energy consumption to be adjustable to a target application and for providing a degree of
226 freedom in circuit design. In this work, conductance levels of 128 ($=2^7$; 7 bits) were selected
227 from those of 8,192 ($=2^{13}$; 13 bits) uniformly distributed in the entire P/D region for evaluating
228 a rate of pattern recognition, because they are high enough to acquire it in MNIST recognition.
229 **Figure 4c** shows the extracted 128 conductance levels obtained by applying an identical
230 training pulse, without the aid of external circuits.

231 In order to quantitatively evaluate the linearity of the G_{DS} change, a non-linearity parameter
232 α was calculated using the following equation¹¹:

$$233 \quad G = \begin{cases} ((G_{max}^\alpha - G_{min}^\alpha) \times w + G_{min}^\alpha)^{1/\alpha} & \text{if } \alpha \neq 0, \\ G_{min}^\alpha \times (G_{max}/G_{min})^w & \text{if } \alpha = 0. \end{cases} \quad (1)$$

234 , where G_{max} and G_{min} are the maximum and minimum channel conductance, α is a parameter
235 that controls potentiation (α_{pot}) or depression (α_{dep}), and w is an internal variable which ranges
236 from 0 to 1. α_{pot} and α_{dep} close to one is ideal for linear and symmetric conductance change, to
237 improve MNIST classification accuracy in the DNN. In the pEGST, the extracted α_{pot} and α_{dep}
238 under the identical gate pulse amplitude and width were 1.51 and 0.38, respectively. They were
239 affected by the magnitude of the gate pulse amplitude and width (see Supplementary
240 Information 8). In addition, the measured G_{DS} was slightly deviated from the linearly fitted line
241 with α_{pot} and α_{dep} . This regression error (e_{reg}) was very small, *i.e.*, $e_{reg,pot} = 0.007$ and $e_{reg,dep} =$
242 0.006 . This feature is attractive not only for avoiding unstable operation induced by device
243 variability, but to improve the accuracy of pattern recognition while saving energy. It is well
244 known that the smaller regression error for the weight update can induce high learning accuracy
245 as the number of training epochs increase⁴⁰⁻⁴².

246 On the other hand, the aforementioned asymmetric ratio (AR) can reflect how much the
247 potentiation slope differs from the depression slope. The AR was also assessed using the
248 following equation²⁵:

$$249 \quad AR = \frac{\max|G_p(n)-G_d(n)|}{G_p(128)-G_d(128)} \quad \text{for } n = 1 \text{ to } 128 \quad (2)$$

250 , where $G_p(n)$ and $G_d(n)$ are the channel conductance values after the n^{th} potentiation and
251 depression pulses, respectively. For ideal symmetry, the AR should be 0. The pEGST in this
252 work showed a small AR of 0.29, which indicates good symmetry. Whereas the AR is in a range
253 of 0.43 to 0.83 for two-terminal based devices with identical pulses, it is in the range of 0.19
254 to 0.31 for other liquid based EGTs²³⁻²⁶ (see Table 1 and Table S1).

255 **Figure 4 d** shows the cyclic endurance after more than 100,000 operations, updating the
256 synaptic weight. The G_{max} decreased to less than 12 % after 10^5 cycles with the same pulse
257 scheme described above. Moreover, the cycle-to-cycle variation induced by the iterative
258 operations was smaller than those in other types of silicon based synaptic devices, such as
259 ferroelectric FETs, and charge-trap based memories⁴³⁻⁴⁶. **Figure 4e** and **f** show the retention
260 characteristics of the pEGST after each step in the conductance update.

261

262 **Schematic of the array and neural network simulation**

263 **Figure 5a** shows a schematic of a feasible 3-terminal based pEGST array for an analog neural
264 network system. In the neural network system, the crossbar array architecture has two key
265 kernels: for parallel weight updates and the vector-matrix multiply-accumulate operation (*e.g.*,
266 MAC)³¹. Reflecting a 3-terminal MOSFET structure, these can be realized because the weight
267 update is enabled via the gate, the simultaneous reading of conductance is allowed via the drain,
268 and the accumulation of signals is accomplished via the source.

269 A 3-terminal synaptic device was previously developed to demonstrate the potential of

270 parallel programming, which can update synaptic weight during supervised learning (*e.g.*,
271 backpropagation)^{47,48}. In a feasible synapse array, a ‘write’ pulse denoted by the green line on
272 the left side of **Fig. 5a**, is independently applied to each synaptic device to carry out the weight
273 updates, and a ‘read’ pulse, marked with a yellow line on the right side of **Fig. 5a**, is applied to
274 the drain electrode to read out the level of weighted analog conductance. As a result, the writing
275 and reading operations can be concurrently performed by the 3-terminal structure, which is
276 superior to a conventional 2-terminal based synaptic device for efficient neural network. The
277 accumulated voltage, which is multiplied by the read-out current and synaptic weight
278 conductance, is transmitted to a postsynaptic neuron via a source in order to perform the vector-
279 matrix MAC operations. A feasible procedure for fabricating a high-density pEGST array with
280 a vertical pillar structure is shown in the Supplementary Information (see Supplementary
281 Information 4).

282 To evaluate the functionality of the fabricated pEGST for pattern recognition, a hand-written
283 dataset (Modified National Institute of Standards and Technology (MNIST)) was used, and
284 supervised learning with backpropagation was carried out. Multi-layer perceptron (MLP) was
285 composed of 24×22 input neurons cropped from 28×28 pixels, 250 and 125 neurons for
286 the 1st and 2nd hidden layers, and 10 output neurons for the neural network simulations, as
287 shown in **Figure 5b** (see Supplementary Information 9 for details pertaining to the neural
288 network system)⁴⁹. Recognition accuracy after each training epoch is shown in **Figure 5c**. In
289 the simulations, the fabricated pEGST achieved a recognition accuracy of 91.7 % after 30
290 training epochs, which is comparable to the 92.7 % that was obtained from an ideal synapse.
291 Here the ideal synapse is a device which has 128-levels of multi-states, perfect linearity ($\alpha=1$),
292 and complete symmetry (AR=0) in conductance change. In this study, the high level of the
293 recognition accuracy is attributed to the number of multi-states more than 128-levels, good
294 linearity ($\alpha_{\text{pot}}=1.51$ and $\alpha_{\text{dep}}=-0.38$), and reasonable symmetry (AR=0.29) in conductance

295 change.

296 **Figure 5d** shows the recognition accuracy for the MNIST dataset according to the number of
297 conductance levels. To exclude side effects arising from other factors that can influence the
298 accuracy of the recognition, other parameters such as G_{\max} , G_{\min} and linearity were fixed. As
299 previously mentioned, the 128 extracted conductance levels were enough to achieve highly
300 accurate the pattern recognition. Note that there was no notable increase in the recognition
301 accuracy with more than 128 extracted conductance levels. **Table 1** compares the intraspecific
302 structures of various electrolyte-gated FETs and their synaptic properties with the pEGST. In
303 addition, other interspecific synaptic devices are compared with the pEGST in Supplementary
304 Information 10.

305

306 **Conclusion**

307 An all-solid-state polymer electrolyte-gated synaptic transistor (pEGST) was implemented on
308 a silicon channel, and demonstrated high performance in an analog deep neural network. As
309 the solid-state electrolyte, pEGDMA including protons (H^+) was used for the first time. Unlike
310 liquid or ionic gel-type electrolytes that have been used in a conventional EGSTs, the pEGDMA
311 deposited via the iCVD process at a wafer-scale allows the full utilization of mature CMOS
312 microfabrication technology. This reduces process-induced variability across the wafer. The
313 pEGST showed conductance control with bidirectional analog, linear and symmetric synapse
314 behavior. It achieved notable performance as a synapse device with weight updates of more
315 than 8,192-levels with fine conductance control. The pEGST also demonstrated an excellent
316 recognition accuracy of 91.7 % for the MNIST dataset. The pEGST provides a feasible pathway
317 to realize a bio-inspired electrolyte synapse for chip-level integration thanks to its wafer-scale
318 CMOS-compatibility.

319

320 **Methods**

321 **Device fabrication:** See Supplementary Information 2 for details of the fabrication process.

322 **iCVD process:** To deposit the conformal and uniform thin polymer electrolyte film, vaporized
323 ethylene glycol dimethacrylate (EGDMA) monomer was flowed into a pressure-controlled
324 vacuum chamber with a tert-butyl peroxide (TBPO) initiator. The ratio of the flow rate between
325 the monomer and initiator was 1:1, and a proportional-integral-derivative (PID) controller kept
326 the chamber pressure at approximately 60 mTorr. In the vacuum chamber, a heated filament
327 thermally decomposes the initiator and produces the radical, which activates the vinyl group in
328 the monomer. The polymerization reaction and adsorption occur simultaneously on the surface
329 of the samples, with a substrate temperature of 40 °C.

330 **Device characterization:** Electrical characteristics of the fabricated pEGST were measured
331 using a B1500 semiconductor parameter analyzer with PMU module (Agilent Technologies)
332 using optimized pulse schemes. Each I_D-V_G curve was obtained by sweeping the applied
333 voltage from 0 V to less than ± 6 V with a 0.05 V step. '+' indicates forward sweep and '-' is
334 backward sweep. Capacitance-frequency ($C-f$) and phase angle-frequency ($\theta-f$) measurements
335 were also carried out using a E4980A LCR meter (Agilent Technologies) at a frequency range
336 of 1 kHz to 1 MHz with a small AC signal voltage of 30 mV. And DC 2 V were was
337 superimposed to the AC signal to make channel inversion. To get measurable capacitance, a
338 large-sized pEGST with an L_G of 50 μm and W_{ch} of 50 μm was selected.

339 **TEM analysis:** TEM images were taken using high-resolution corrected scanning transmission
340 electron microscopy (JEM-ARM200F) with EDS mapping (Bruker Quantax 400).

341 **FT-IR analysis:** To confirm the chemical structure and heat resistance of the pEGDMA thin
342 film, FT-IR spectroscopy (Nicolet iN10MX) was used.

343 **ToF-SIMS analysis:** To confirm the ion mass in the pEGDMA film, time of flight secondary
344 ion mass spectrometry (ToF-SIMS) from Ion-ToF GmbH was used. Each 400 $\mu\text{m} \times 400 \mu\text{m}$

345 sample was comprised of a silicon channel, interfacial silicon dioxide and pEGDMA.

346 **Acknowledgements**

347 We acknowledge inspiring discussions about polymer electrolyte with M. -L. Seol, H. Bae,
348 and H. Moon. This work was supported by National Research Foundation (NRF) of Korea,
349 under Grant 2018R1A2A3075302, 2019M3F3A1A03079603 and 2017R1A2B3007806, in
350 part by the IC Design Education Center (EDA Tool and MPW).

351 **Author Contributions**

352 J.-M. Yu, J. Hur and Y.-K. Choi conceived the idea and designed the experiments. J.-M. Yu, C.
353 Lee, D.-J. Kim, H.-G. Park, M.-S. Kim, and M. Seo fabricated the devices. J.-M. Yu, D.-J. Kim
354 and J.-K. Han performed the experiments and data analysis. J.-K. Kim performed the neural
355 network simulation. J.-M. Yu wrote the manuscript. S.G. Im and Y.-K. Choi supervised the
356 project on the relevant portions of the research. All authors discussed the results and
357 commented on the manuscript.

358 **Data availability**

359 The data that support the findings of this study are available from the corresponding author
360 upon reasonable request.

361 **Code availability**

362 The codes used for plotting the deep neural network simulation data are available from the
363 corresponding author on reasonable request.

364

365 **References**

- 366 [1] Mead, C. Neuromorphic electronic systems. *Proc. IEEE* **78**, 1629–1636 (1990).
367 [2] Mahowald, M. & Douglas, R. A silicon neuron. *Nature* **354**, 515–518 (1991).
368 [3] Zidan, M. A., Strachan, J. P. & Lu, W. D. The future of electronics based on memristive systems. *Nat.*
369 *Electron.* **1**, 22–29 (2018).

- 370 [4] Yang, J. J., Strukov, D. B. & Stewart, D. R. Memristive devices for computing. *Nat. Nanotech.* **8**,
371 13–24 (2013).
- 372 [5] Indiveri, G. et al. Neuromorphic silicon neuron circuits. *Front. Neurosci.* **5**, 73 (2011).
- 373 [6] LeCun, Y., Bengio, Y. & Hinton, G. Deep learning. *Nature* **521**, 436–444 (2015).
- 374 [7] Krizhevsky, A., Sutskever, I. & Hinton, G. E. ImageNet classification with deep convolutional neural
375 networks. in *Advances in Neural Information Processing Systems 25* (eds Pereira, F., Burges, C. J. C.,
376 Bottou, L. & Weinberger, K. Q.) 1097–1105 (Curran Associates, Red Hook, NY, 2012).
- 377 [8] Hinton, G. et al. Deep neural networks for acoustic modeling in speech recognition. *IEEE Signal*
378 *Processing Magazine* **29**, 82–97 (2012).
- 379 [9] van de Burgt, Y. et al. A non-volatile organic electrochemical device as a low-voltage artificial
380 synapse for neuromorphic computing. *Nat. Mater.* **16**, 414–418 (2017).
- 381 [10] Burr, G. W. et al. Experimental demonstration and tolerancing of a large-scale neural network (165
382 000 synapses) using phase-change memory as the synaptic weight element. *IEEE Trans. Electron*
383 *Devices* **62**, 3498–3507 (2015).
- 384 [11] Jang, J.-W., Park, S., Burr, G. W., Hwang, H. & Jeong, Y.-H. Optimization of conductance change
385 in $\text{Pr}_{(1-x)}\text{Ca}_x\text{MnO}_3$ -based synaptic devices for neuromorphic systems. *IEEE Electron Device Lett.* **36**,
386 457–459 (2015).
- 387 [12] J. Park et al., TiO_x-Based RRAM synapse with 64-Levels of conductance and symmetric
388 conductance change by adopting a hybrid pulse scheme for neuromorphic computing. *IEEE Electron*
389 *Device Lett.* **37**, 1559–1562 (2016)
- 390 [13] H. Bae et al., Bioinspired polydopamine-based resistive switching memory on cotton fabric for
391 wearable neuromorphic device applications. *Adv. Mater. Technol* **4**, 1900151 (2019).
- 392 [14] S. H. Jo et al., Nanoscale Memristor Device as Synapse in Neuromorphic Systems, *Nano Lett.* **10**,
393 1297–1301 (2010).
- 394 [15] S. Lashkare et al., PCMO-based RRAM and NPN bipolar selector as synapse for energy efficient
395 STDP. *IEEE Electron Device Lett.* **38**, 1212–1215 (2017).
- 396 [16] J. Tao, K.-H. Kim & W. Lu, Crossbar RRAM arrays: Selector device requirements during read

397 operation. *IEEE Trans. Electron Devices* **61**, 1369–1376 (2014)

398 [17] Diorio, C., Hasler, P., Minch, B. A., & Mead, C. A. Single transistor silicon synapse. *IEEE Trans.*
399 *Electron Devices* **43**, 1972–1980 (1996).

400 [18] J. Sun et al., Optoelectronic synapse based on IGZO-alkylated graphene oxide hybrid structure.
401 *Adv. Funct. Mater.* **28**, 1804397 (2018).

402 [19] M.-K. Kim et al., Ferroelectric analog synaptic transistor. *Nano Lett.* **19**, 2044–2050 (2019)

403 [20] Elliot J. Fuller et al., Parallel programming of an ionic floating-gate memory array for scalable
404 neuromorphic computing. *Science* **364**, 570–574 (2019).

405 [21] Bi, G. Q. & Poo, M. M. Synaptic modifications in cultured hippocampal neurons: dependence on
406 spike timing, synaptic strength, and postsynaptic cell type. *J. Neurosci.* **2**, 10464–10472 (1998).

407 [22] Zucker, R. S. & Regehr, W. G. Short-term synaptic plasticity. *Annu. Rev. Physiol.* **64**, 355–405
408 (2002)

409 [23] Voglis, G. & Tavernarakis, N. The role of synaptic ion channels in synaptic plasticity. *EMBO Rep.*
410 **7**, 1104–1110 (2006).

411 [24] Zhu et al., Ion gated synaptic transistors based on 2D van der Waals crystals with tunable diffusive
412 dynamics. *Adv. Mater.* **30**, 1800195 (2018).

413 [25] C.-S. Yang et al., All-solid-state synaptic transistor with ultralow conductance for neuromorphic
414 computing. *Adv. Funct. Mater.* **28**, 1804170 (2018).

415 [26] K. Kim et al., A carbon nanotube synapse with dynamic logic and learning. *Adv. Mater.* **25**, 1693–
416 1698 (2013).

417 [27] C. S. Yang et al., A synaptic transistor based on quasi-2D molybdenum oxide. *Adv. Mater.* **29**,
418 1700906 (2017).

419 [28] J. Shi, S. D. Ha, Y. Zhou, F. Schoofs, & S. Ramanathan, A correlated nickelate synaptic transistor.
420 *Nat. Commun.* **4**, 2676 (2013).

421 [29] J. Go et al., W/WO_{3-x} based three-terminal synapse device with linear conductance change and
422 high on/off ratio for neuromorphic application. *Appl. Phys. Express* **12**, 026503 (2019).

423 [30] E. J. Fuller et al., Li-ion synaptic transistor for low power analog computing. *Adv. Mater.* **29**,

424 1604310 (2017).

425 [31] L. Q. Zhu et al., Artificial synapse network on inorganic proton conductor for neuromorphic
426 systems. *Nature Commun.* **5**, 3158, (2014).

427 [32] W. Li, L. C. Bradley & J. J. Watkins, Copolymer solid-state electrolytes for 3D microbatteries via
428 initiated chemical vapor deposition. *ACS Appl. Mater. Interfaces*, **11**, 5668–5674, (2019).

429 [33] H. Moon, H. Seong, W. C. Shin, W.-T. Park, M. Kim, S. Lee, J. H. Bong, Y.-Y. Noh, B. J. Cho, S.
430 Yoo & S. G. Im, Synthesis of ultrathin polymer insulating layers by initiated chemical vapour deposition
431 for low-power soft electronics. *Nat. Mater.* **14**, 628–635 (2015).

432 [34] H. Seong, J. Baek, K. Pak & S. G. Im, A surface tailoring method of ultrathin polymer gate
433 dielectrics for organic transistors: improved device performance and the thermal stability thereof. *Adv.*
434 *Funct. Mater.* **25**, 4462–4469 (2015).

435 [35] W. E. Tenhaeff & K. K. Gleason, Initiated and oxidative chemical vapor deposition of polymeric
436 thin films: iCVD and oCVD. *Adv. Funct. Mater.* **18**, 979–992 (2008).

437 [36] H. Seong, K. Pak, M. Joo, J. Choi, & S. G. Im, Vapor-phase deposited ultrathin polymer gate
438 dielectric for high-performance organic thin film transistors. *Adv. Electron. Mater.* **2**, 1500209 (2016).

439 [37] F. Bordini, C. Cametti & R. H. Colby, Dielectric spectroscopy and conductivity of polyelectrolyte
440 solutions. *J. Phys.: Condens. Matter* **16**, R1423 (2004).

441 [38] O. Larsson, E. Said, M. Berggren & X. Cripin, Insulator polarization mechanisms in
442 polyelectrolyte-gated organic field-effect transistors. *Adv. Funct. Mater.* **19**, 3334–3341 (2009).

443 [39] D. Kuzum, S. Yu & H. -S. P. Wong, Synaptic electronics: materials, devices and applications.
444 *Nanotechnology*, **24**, 382001 (2013).

445 [40] J. Chen et al., LiSiOx-based analog memristive synapse for neuromorphic computing. *IEEE*
446 *Electron Device Lett.* **40**, 542-545 (2019).

447 [41] H. Wu et al., Device and circuit optimization of RRAM for neuromorphic computing. *IEEE*
448 *International Electron Devices Meeting (IEDM) (Invited)*, (2017).

449 [42] C. -C. Chang et al., Challenges and opportunities toward online training acceleration using RRAM-
450 based hardware neural network. *IEEE International Electron Devices Meeting (IEDM)*, (2017).

451 [43] Jerry, M. et al., Ferroelectric FET Analog Synapse for Acceleration of Deep Neural Network

452 Training. *2017 IEEE Int. Electron Devices Meeting (IEDM) 6-2* (IEEE, 2017).

453 [44] Y. -J. Park et al., 3-D Stacked Synapse Array Based on Charge-Trap Flash Memory for
454 Implementation of Deep Neural Networks, *IEEE Trans. on Electron Device.* **66**, 420-427 (2019)

455 [45] J. Hur et al., A Recoverable Synapse Device Using a Three-Dimensional Silicon Transistor, *Adv.*
456 *Funct. Mater.* **28**, 1804844 (2018).

457 [46] C. Diorio, Paul Hasler, A. Minch, & C. A. Mead, A Single-Transistor Silicon Synapse. *IEEE Trans.*
458 *on Electron Device.* **43**, 1972–1980 (1996)

459 [47] S. Agarwal et al., Energy scaling advantages of resistive memory crossbar based computation and
460 its application to sparse coding, *Front. Neurosci.* **9**, 484 (2016)

461 [48] H. Han et al., Recent progress in three-terminal artificial synapses: from device to system. *Small.*
462 **15**, 1900695 (2019)

463 [49] M. Seo et al., First demonstration of a logic-process compatible junctionless ferroelectric FinFET
464 synapse for neuromorphic applications. *IEEE Electron Device Lett.* **39**, 1445–1448 (2018)

465

466

467

468

469

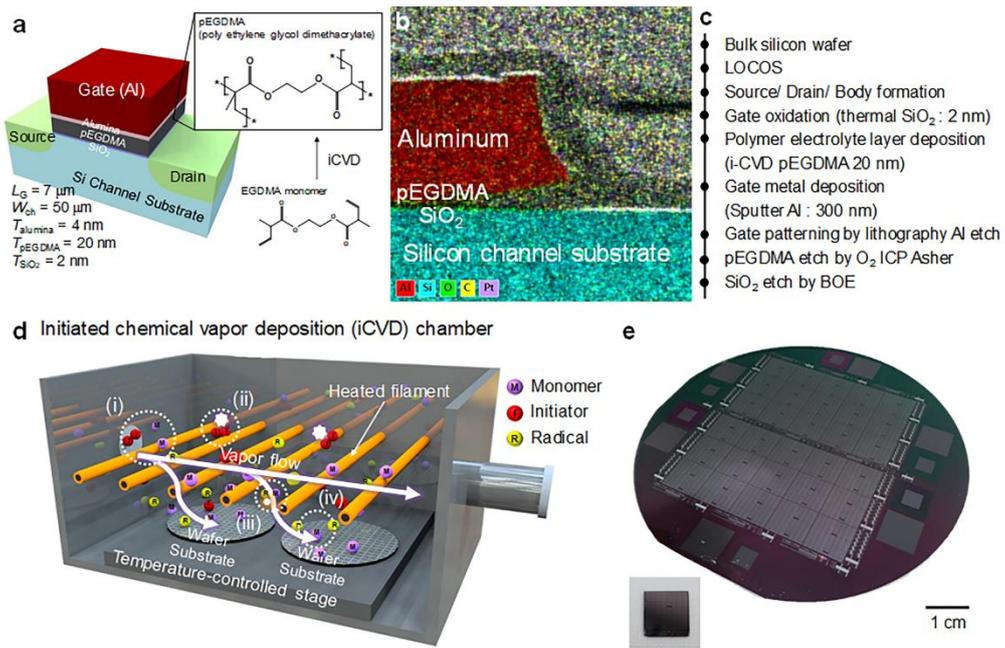


Fig. 1| Schematic and TEM image of pEGST with microfabrication procedures, iCVD process, and photographic image of a fabricated wafer. a, Schematic illustration of the pEGST and chemical structure of the EGDMA and pEGDMA. **b,** Cross-sectional TEM image of the gate stack. The iCVD method permits the conformal formation of thin polymer film across the wafer **c,** Key fabrication processes of the pEGST. **d,** Schematic of the iCVD chamber for the pEGDMA electrolyte deposition **e,** Optical photograph of a 4-inch silicon wafer and a single chip after the fabrication of the pEGST.

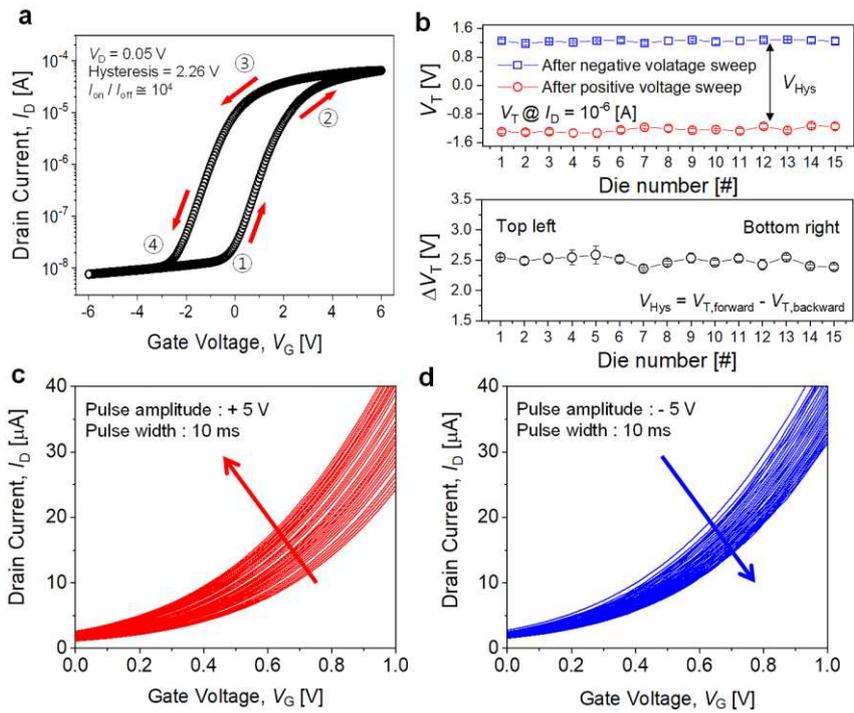


Fig. 2] Electrical measurement characteristics of pEGST. a, Transfer characteristic (I_D - V_G) curve of the pEGST with hysteresis of 2.26 V and I_{ON}/I_{OFF} ratio of 10^4 . **b,** Wafer-scale uniformity of the threshold voltage (V_T) and hysteresis ($\Delta V_T = V_{T,forward} - V_{T,backward}$) in the entire 4-inch wafer level. **c,** I_D is changed by each number of gate pulse for potentiation. **d,** I_D is changed by each number of gate pulse for depression. These graphs represent the analog memory characteristics of the pEGST.

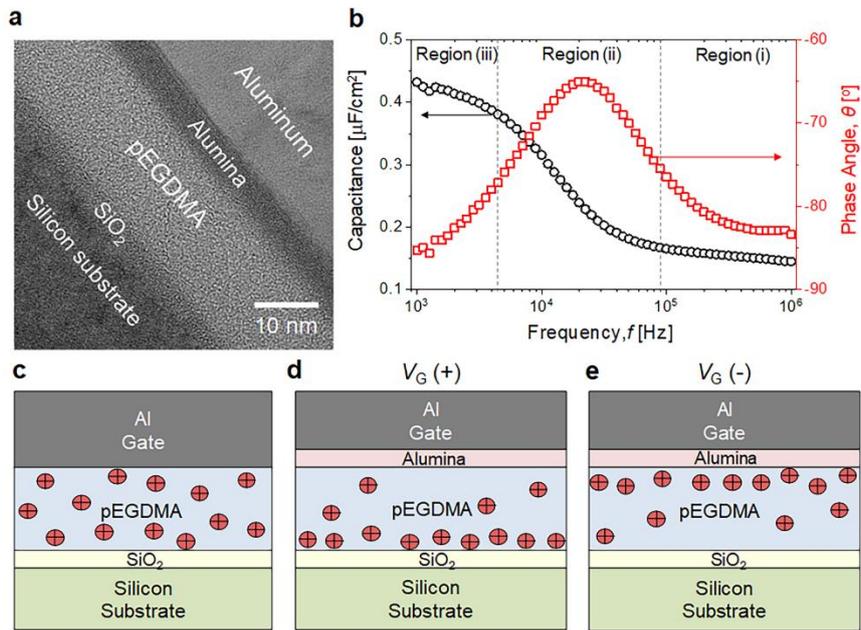


Fig. 3 | TEM image of solid-state gate dielectrics and their ultra-thin electrolyte characteristics. **a**, TEM image of the gate dielectrics composed of SiO₂, pEGDMA and Al₂O₃. **b**, Measured capacitance per unit area and phase angle as a function of applied frequency. They are categorized as three regions: (i) dipole relaxation, (ii) ionic relaxation, and (iii) electric double layer formation. **c**, Distribution profile of H⁺ (ρ_{H+}) under equilibrium without V_G. **d**, ρ_{H+} with positive V_G(+). and **e**, ρ_{H+} with negative V_G(-). The ρ_{H+} is redistributed according to the polarity of the V_G that causes hysteresis in the pEGST.

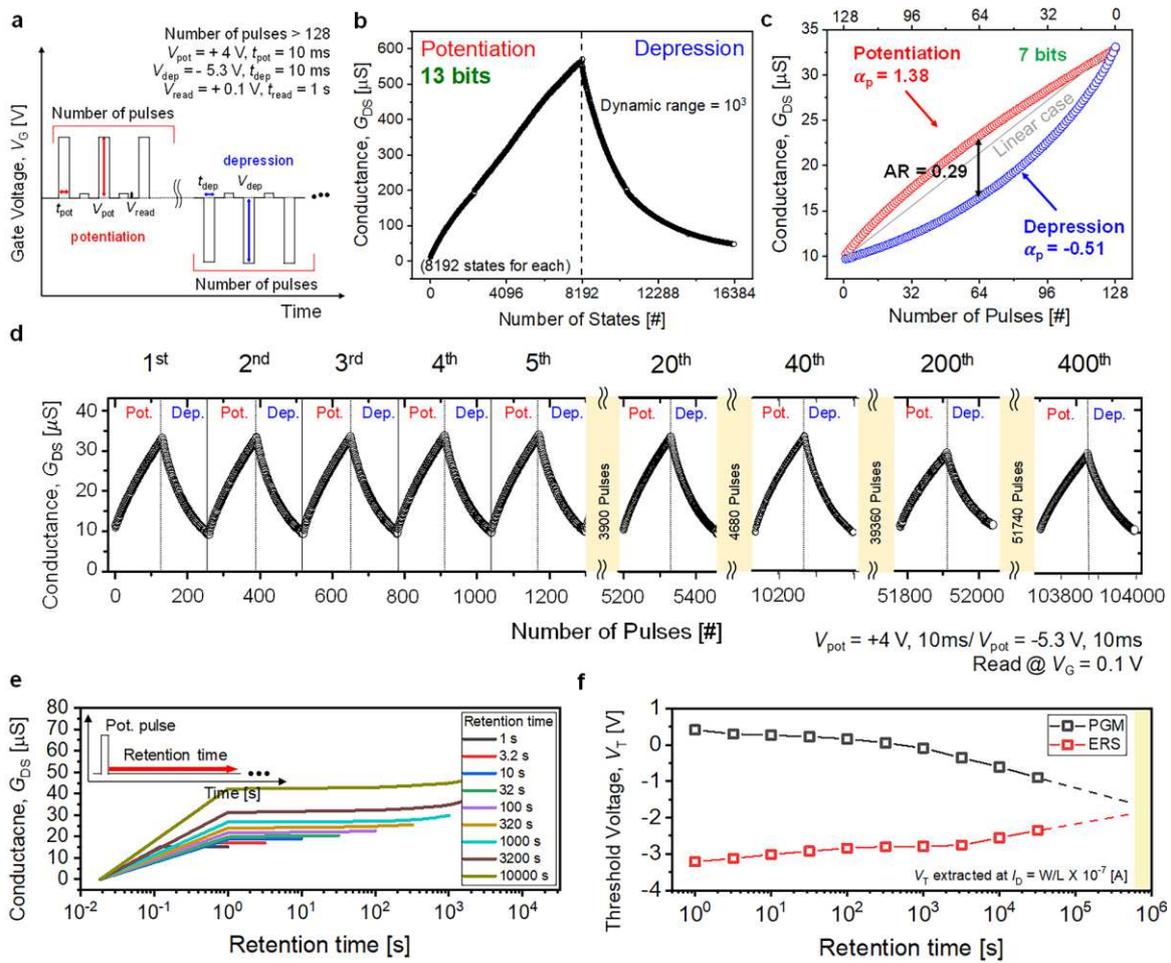


Fig. 4| Synapse characteristics for analog deep neural network of pEGST. a, Specific pulse scheme of potentiation (+4 V, 10 ms), depression (-5.3 V, 10 ms), and read operation (0.1 V). **b**, Measured multi-level channel conductance (G_{DS}) of 8,192 (13 bits) according to the number of pulses with a high current ratio of I_{ON} to I_{OFF} ($>10^3$). **c**, Good linearity ($\alpha_{pot} = 1.38$ and $\alpha_{dep} = -0.51$) with asymmetric ratio of 0.29 in the selected range of conductance updating. **d**, Endurance characteristic of the pEGST more than 100,000 cycles. **e**, G_{DS} change by conductance retention time after applying potentiation pulses. **f**, Non-volatile retention characteristic of the pEGST by evaluation of a threshold voltage (V_T) shift after the pulse application.

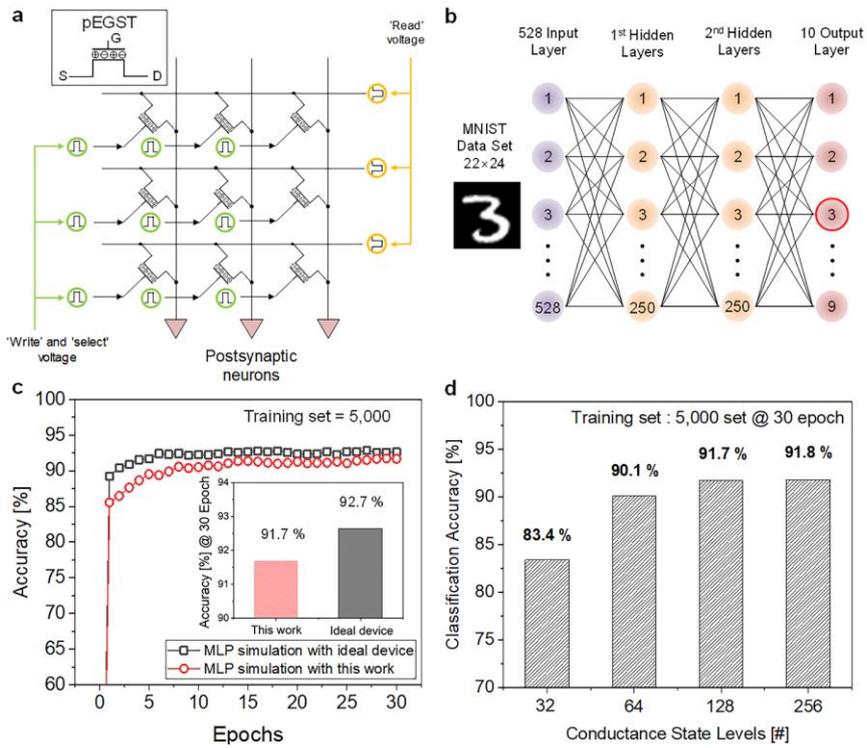


Fig. 5| Schematic of pEGST array and simulated pattern recognition accuracy by use of multi-layer perceptron (MLP) for deep neural network a, Schematic of the pEGST array. b, Configuration of MLP for the simulation to evaluate the pattern recognition accuracy in the MNIST dataset. c, Simulated pattern recognition accuracy from the pEGST compared to that from ideal synaptic device. d, Recognition accuracy according to the number of the G_{DS} .

Table. 1| Comparison of electrolyte-gated FET based artificial synaptic devices

Electrolyte-gated FET based artificial synaptic devices							
Reference number	[24]	[25]	[26]	[27]	[28]	[29]	This work
Constitution	1T	1T	1T	1T	1T	1T	1T
Fabrication scale	Device-scale	Device-scale	Device-scale	Device-scale	Device-scale	Device-scale or Wafer-scale	Wafer-scale
CMOS compatible	No	No	No	No	No	Yes	Yes
Electrolyte	Ion gel (LiClO ₄ /PEO)	Ion gel (LiClO ₄ /PEO)	All-solid-state polymer (PEG)	Ionic liquid	Ionic liquid	Metallic oxide (WO _{3-x})	All-solid-state polymer (PEGDMA)
Deposition method of electrolyte	Mixture drop	Dip-coating	Spin-coating	Liquid drop	Liquid drop	Sputtering	Initiated CVD
Thickness of electrolyte	-	-	90 nm	-	-	80 nm	20 nm
Active ion	Li ⁺	Li ⁺	H ⁺	H ⁺	O ²⁻	O ²⁻	H ⁺
Channel	WSe ₂	α -MoO ₃	Single-walled CNTs	α -MoO ₃	SmNiO ₃	W	Silicon
Channel formation	Mechanical exfoliation	Mechanical exfoliation	CNT solution dipping	Mechanical exfoliation	Sputtering	Sputtering	Commercial silicon wafer
# of bits	6 (60)	5 (50)	6 (100)	5 (50)	>7	6	>13
Conductance change linearity ($\epsilon_{pot}/\epsilon_{dep}$)	1.9/0.5	2.6/-0.4	Poor	1.7/0.4	1.3/0.9	1.3/-0.3	1.51/-0.38
Asymmetry ratio	0.19	0.31	-	0.2	-	-	0.29
Pulse width	50 ms	10 ms	1 ms	1 ms	10 ms	10 ms	10 ms
Switching energy	~30 fJ	0.16 pJ	~7.5 pJ	500 fJ	-	-	< 20 fJ

* 1T = Single transistor

Figures

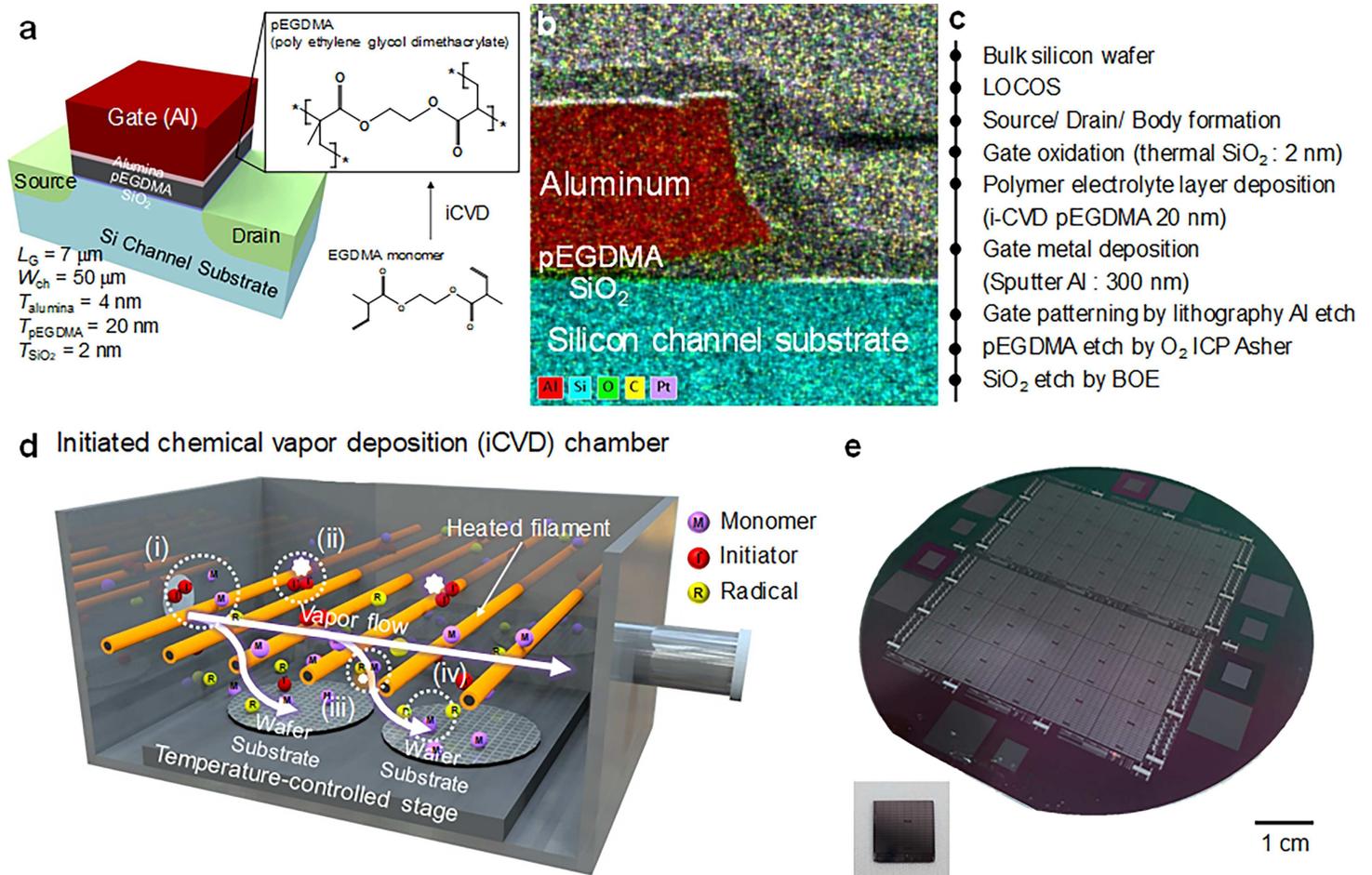


Figure 1

Schematic and TEM image of pEGST with microfabrication procedures, iCVD process, and photographic image of a fabricated wafer. a, Schematic illustration of the pEGST and chemical structure of the EGDMA and pEGDMA. b, Cross-sectional TEM image of the gate stack. The iCVD method permits the conformal formation of thin polymer film across the wafer c, Key fabrication processes of the pEGST. d, Schematic of the iCVD chamber for the pEGDMA electrolyte deposition e, Optical photograph of a 4-inch silicon wafer and a single chip after the fabrication of the pEGST.

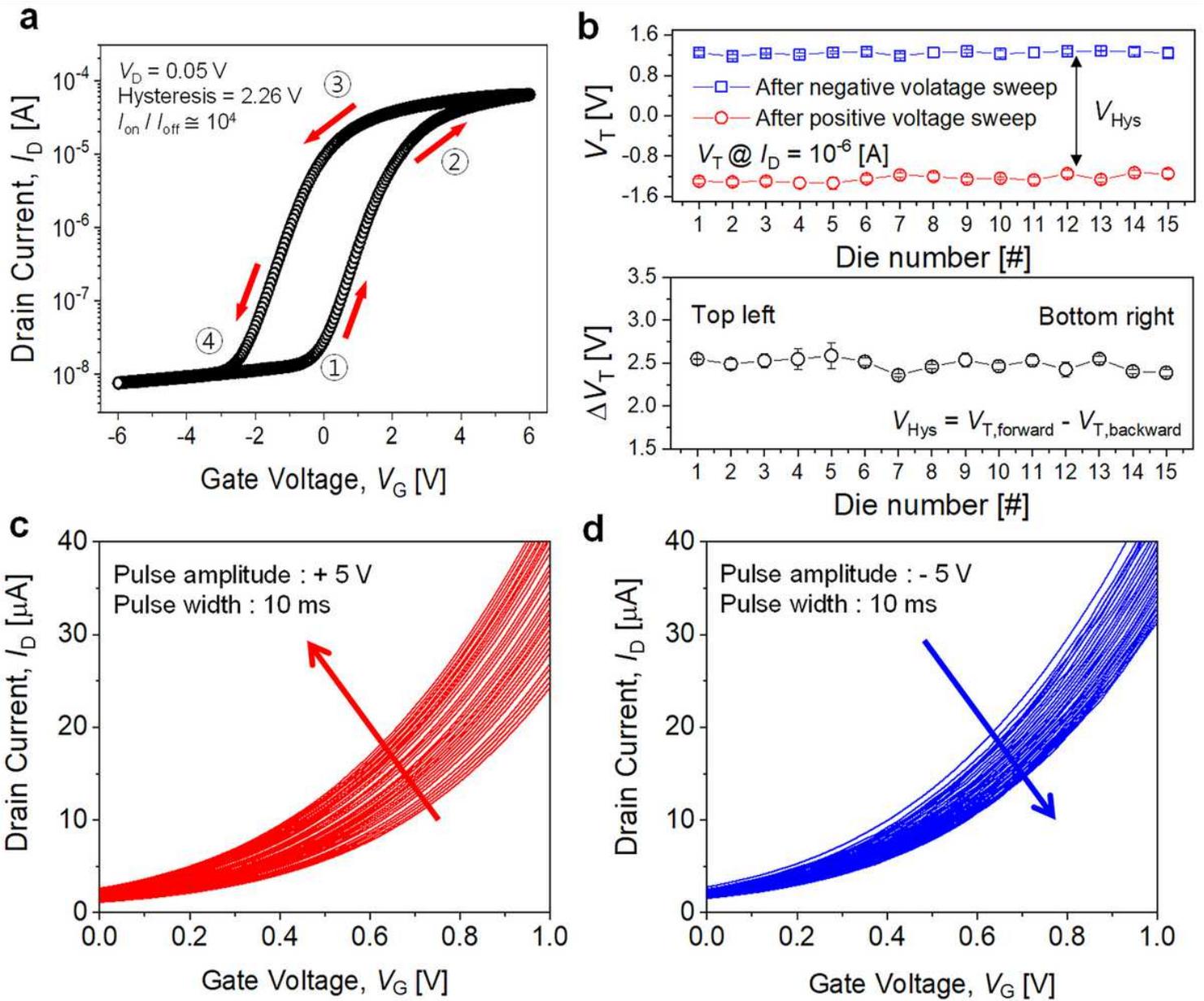


Figure 2

Electrical measurement characteristics of pEGST. a, Transfer characteristic (I_D - V_G) curve of the pEGST with hysteresis of 2.26 V and I_{ON}/I_{OFF} ratio of 104. b, Wafer-scale uniformity of the threshold voltage (V_T) and hysteresis ($\Delta V_T = V_{T,forward} - V_{T,backward}$) in the entire 4-inch wafer level. c, I_D is changed by each number of gate pulse for potentiation. d, I_D is changed by each number of gate pulse for depression. These graphs represent the analog memory characteristics of the pEGST.

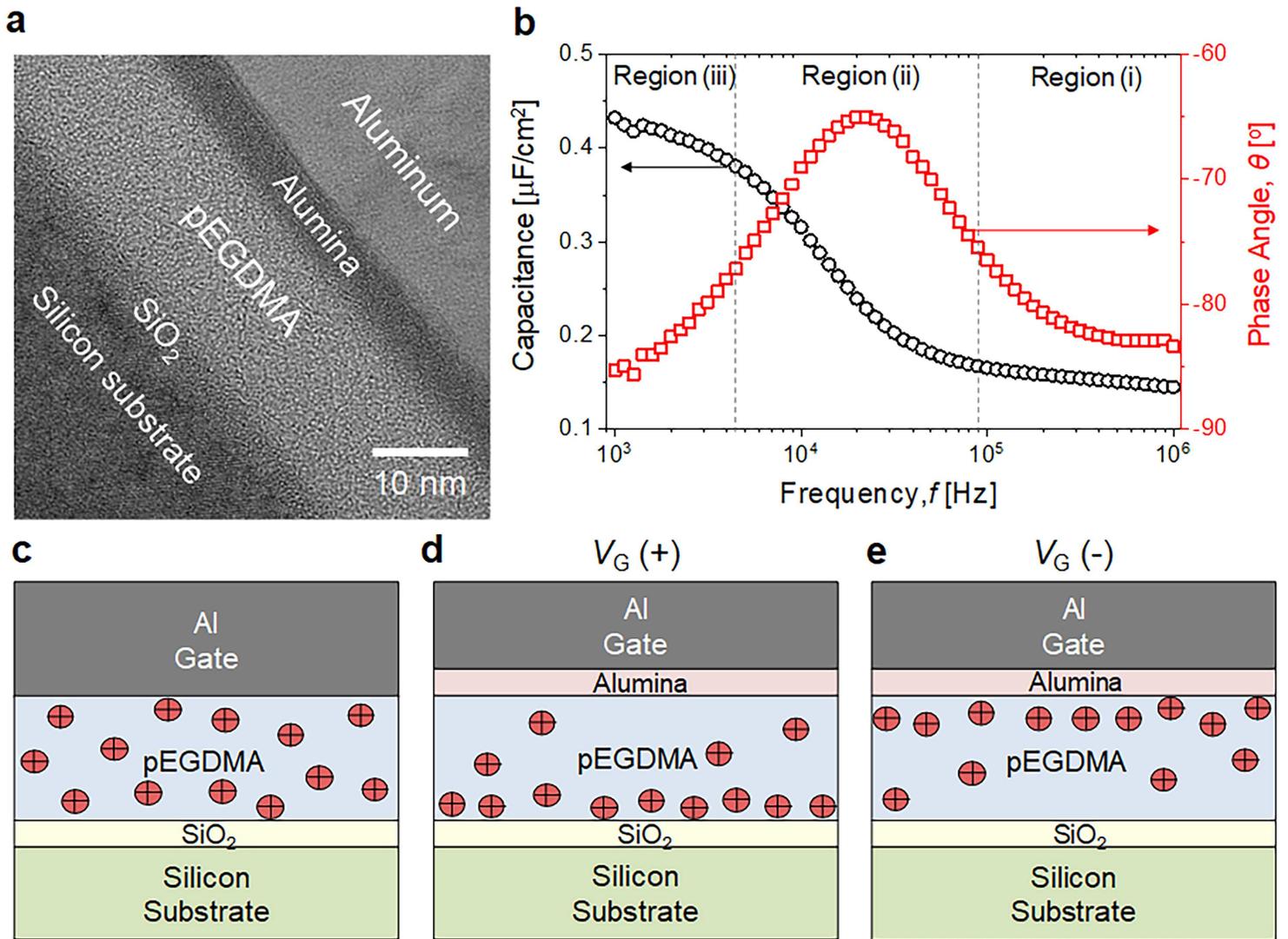


Figure 3

TEM image of solid-state gate dielectrics and their ultra-thin electrolyte characteristics. a, TEM image of the gate dielectrics composed of SiO_2 , pEGDMA and Al_2O_3 . b, Measured capacitance per unit area and phase angle as a function of applied frequency. They are categorized as three regions: (i) dipole relaxation, (ii) ionic relaxation, and (iii) electric double layer formation. c, Distribution profile of H^+ (pH^+) under equilibrium without V_G . d, pH^+ with positive $V_G(+)$. and e, pH^+ with negative $V_G(-)$. The pH^+ is redistributed according to the polarity of the V_G that causes hysteresis in the pEGST.

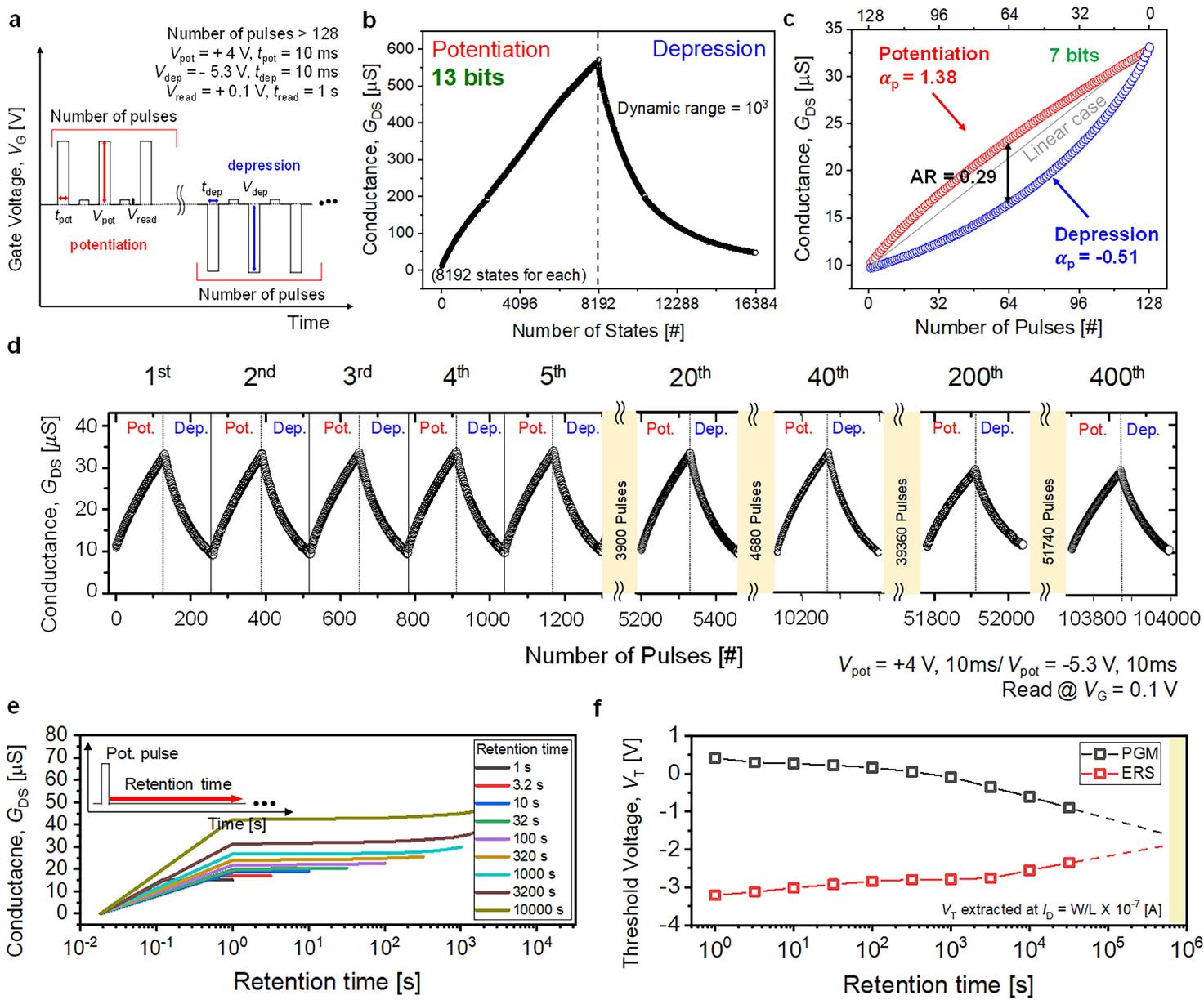


Figure 4

Synapse characteristics for analog deep neural network of pEGST. a, Specific pulse scheme of potentiation (+4 V, 10 ms), depression (-5.3 V, 10 ms), and read operation (0.1 V). b, Measured multi-level channel conductance (G_{DS}) of 8,192 (13 bits) according to the number of pulses with a high current ratio of I_{ON} to I_{OFF} (>103). c, Good linearity ($\alpha_{pot} = 1.38$ and $\alpha_{dep} = -0.51$) with asymmetric ratio of 0.29 in the selected range of conductance updating. d, Endurance characteristic of the pEGST more than 100,000 cycles. e, G_{DS} change by conductance retention time after applying potentiation pulses. f, Non-volatile retention characteristic of the pEGST by evaluation of a threshold voltage (V_T) shift after the pulse application.

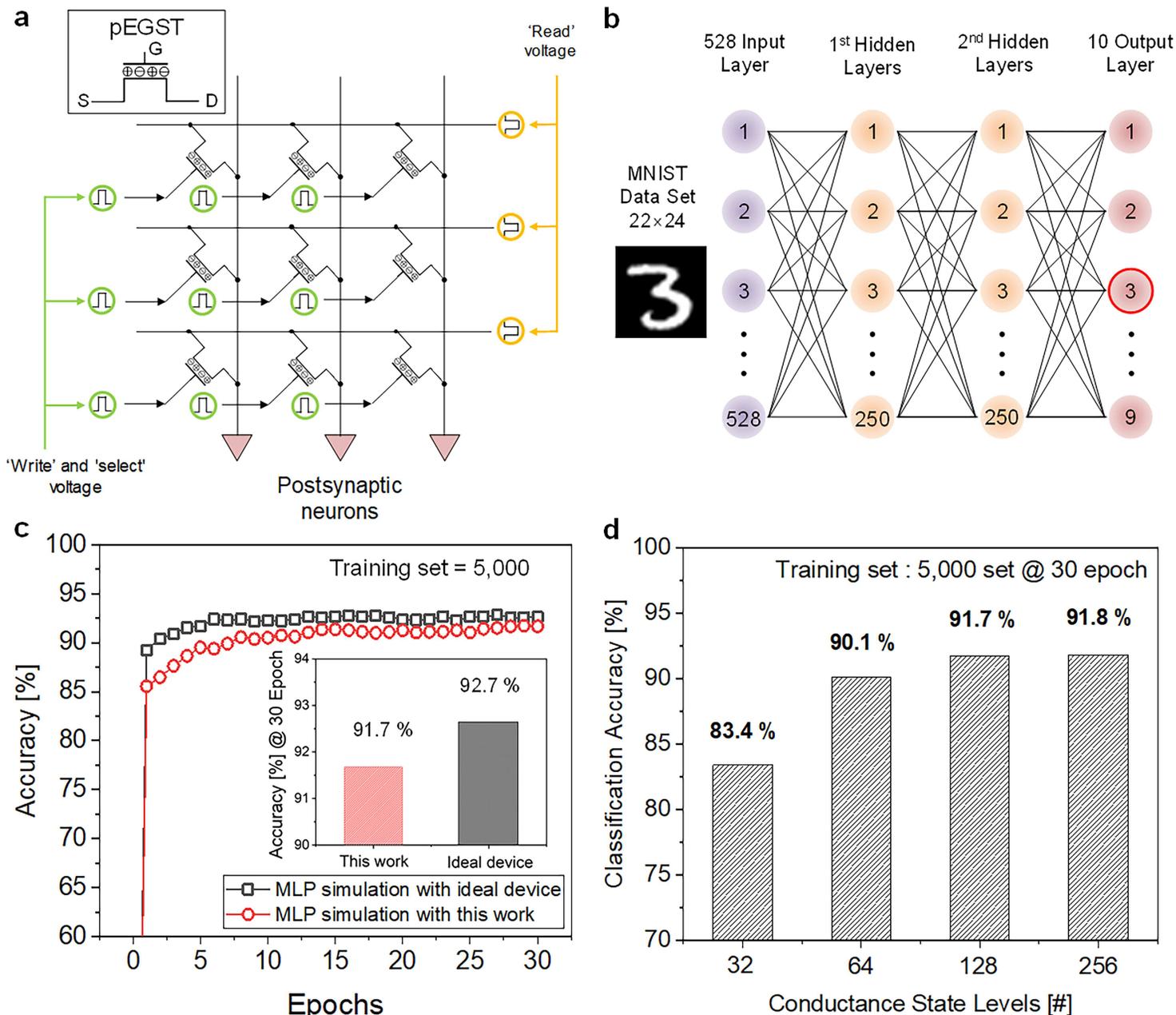


Figure 5

Schematic of pEGST array and simulated pattern recognition accuracy by use of multi-layer perceptron (MLP) for deep neural network a, Schematic of the pEGST array. b, Configuration of MLP for the simulation to evaluate the pattern recognition accuracy in the MNIST dataset. c, Simulated pattern recognition accuracy from the pEGST compared to that from ideal synaptic device. d, Recognition accuracy according to the number of the GDS.

Supplementary Files

This is a list of supplementary files associated with this preprint. Click to download.

- [SupplementaryInformation.pdf](#)

- [Table1.tif](#)