Supplementary Information for

Graphene oxide-DNA/graphene oxide-PDDA sandwiched membranes with neuromorphic function

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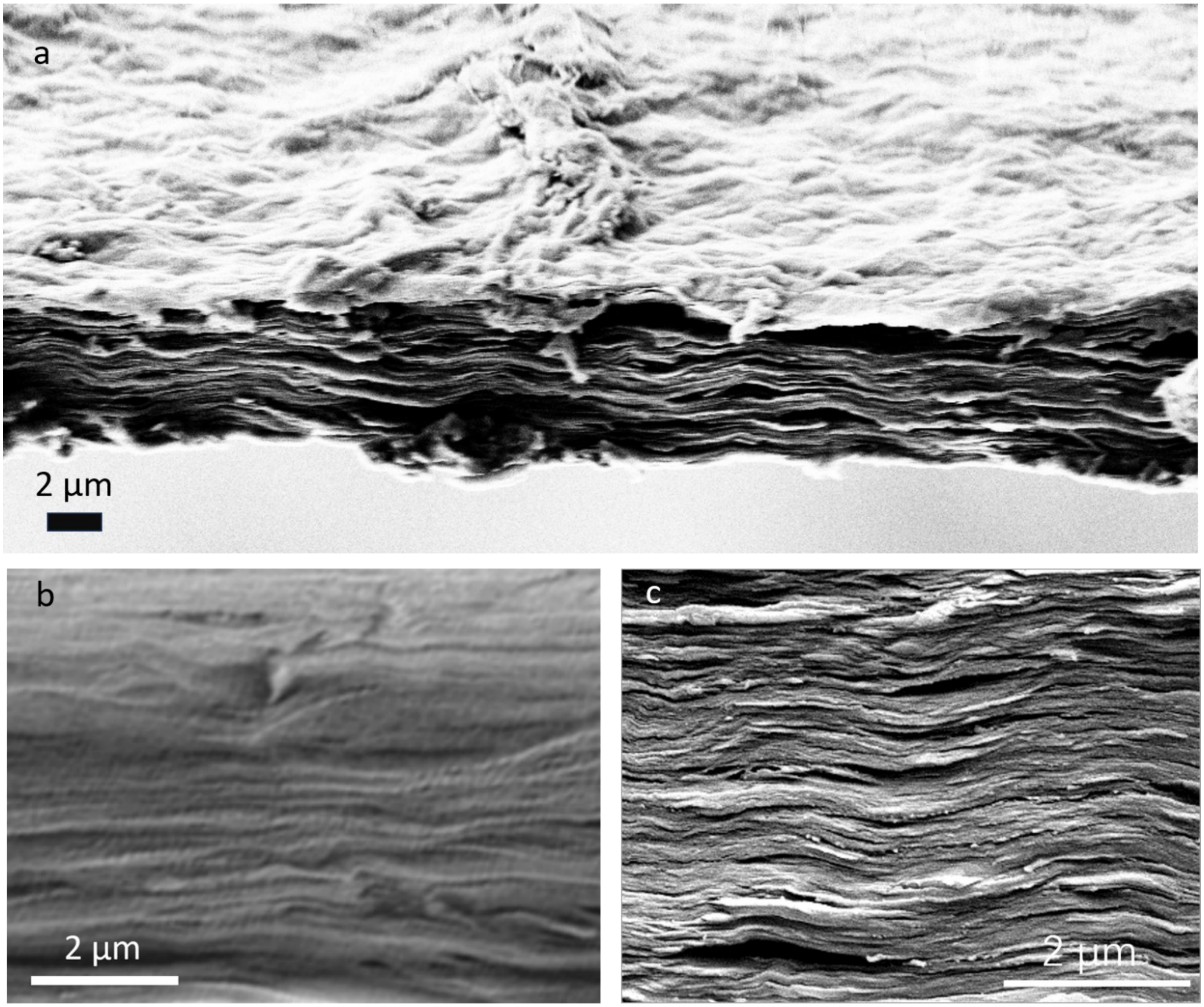

Figure S1. Scanning Electron Microscopy (SEM) image of **(a)** synapse with GO-dsDNA on top and GO-PDDA at the bottom, **(b)** GO-PDDA, and **(c)** GO-dsDNA composite which has nonuniform cross-section morphology with separated phase of dsDNA due to large holes between layers of GO.

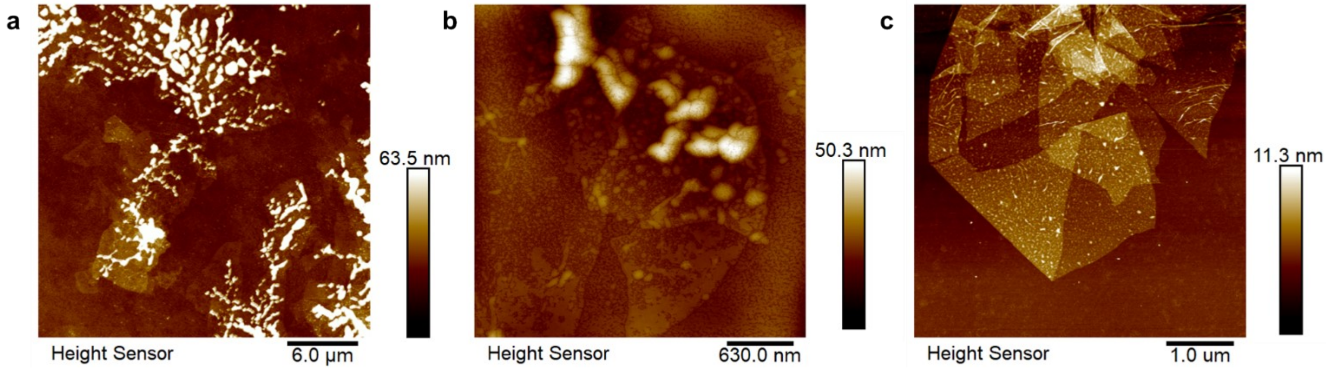

Figure S2. Atomic force microscopy (AFM) images of GO-PDDA (a,b) and GO-dsDNA (c) composite demonstrating uniform distribution of both polymers in GO.

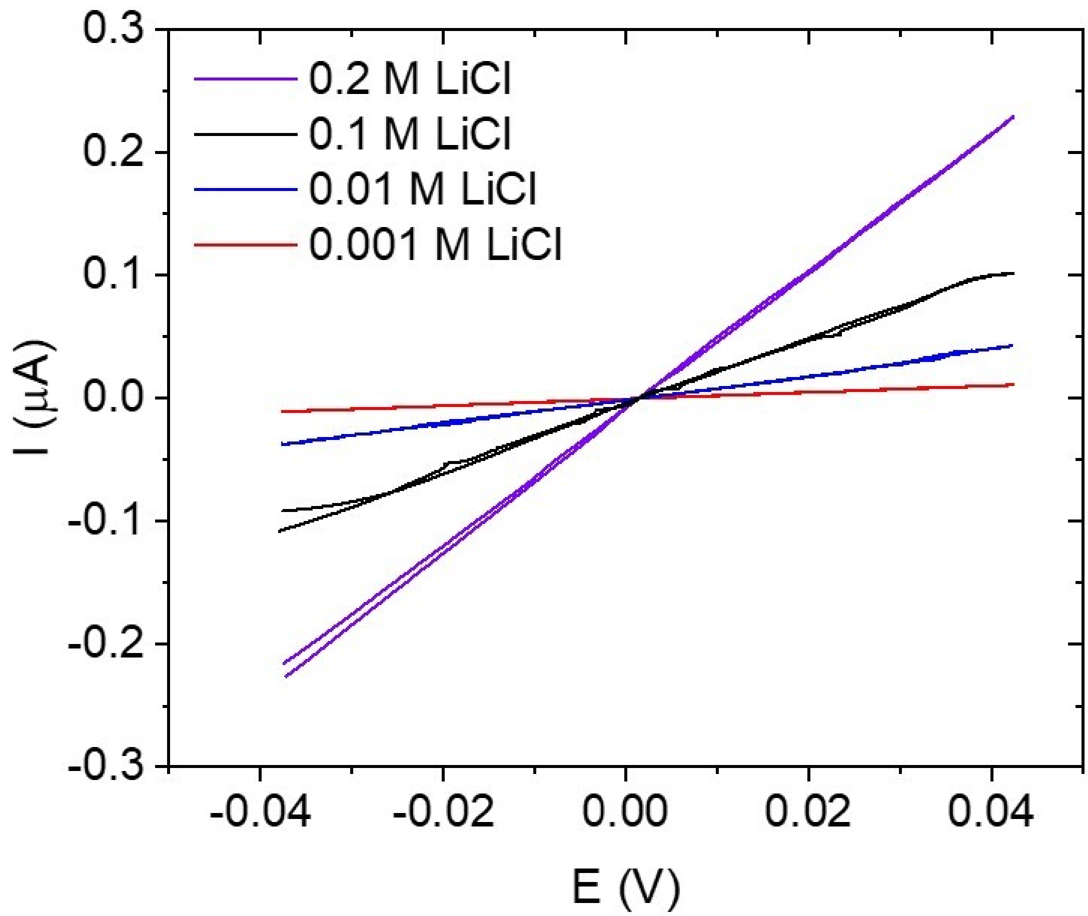

Figure S3. IV curve of GO-PDDA composite with different concentrations of LiCl added. GO-PDDA composite with 0.2 M LiCl showed the highest conductivity, thus most appropriate for iontronic device.

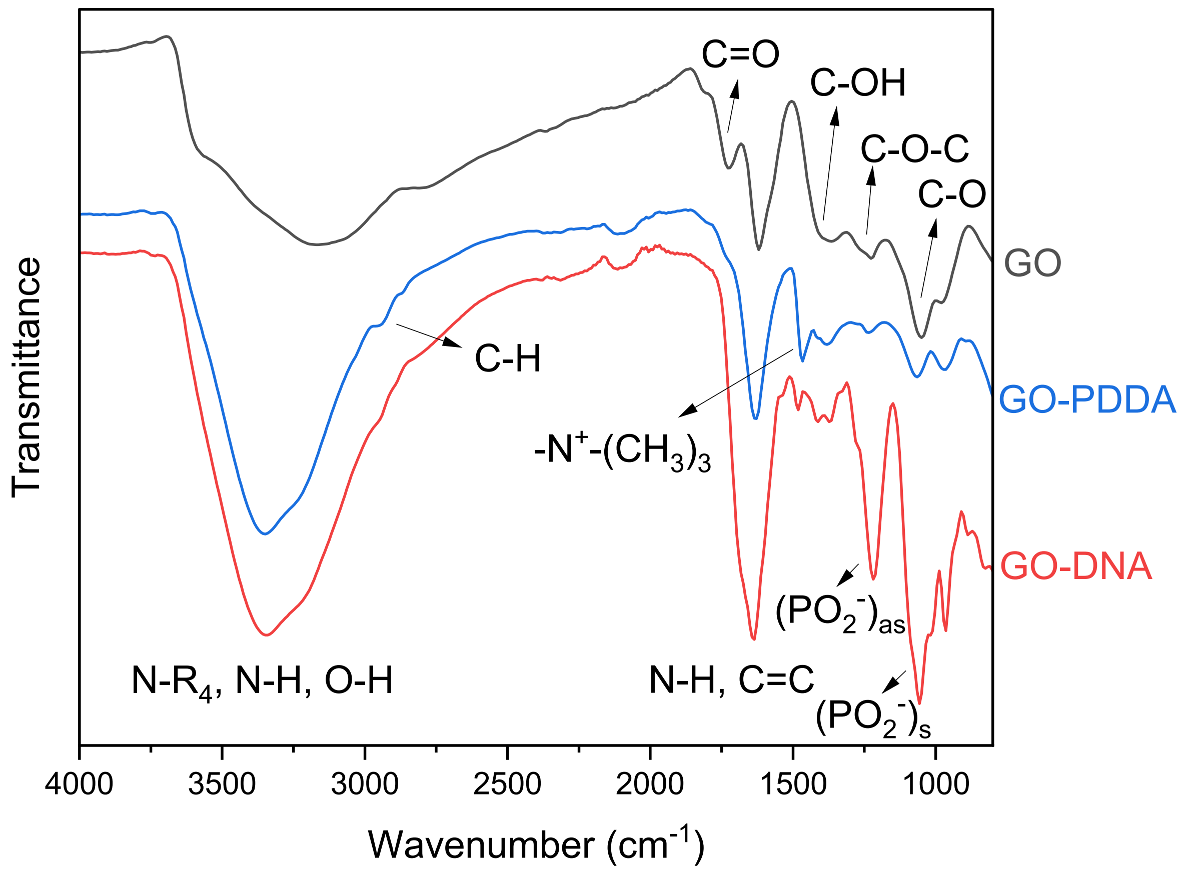

Figure S4. Fourier transform infrared (FTIR) spectra of pure GO, GO-DNA with 0.2 M LiCl and GO-PDDA with 0.2 M LiCl. FTIR was carried out using Shimadzu IRTracer-100 with resolution 0.25 cm^{-1}, equipped with attenuated total reflection accessory. Peaks and respective bond vibrations as indicated.

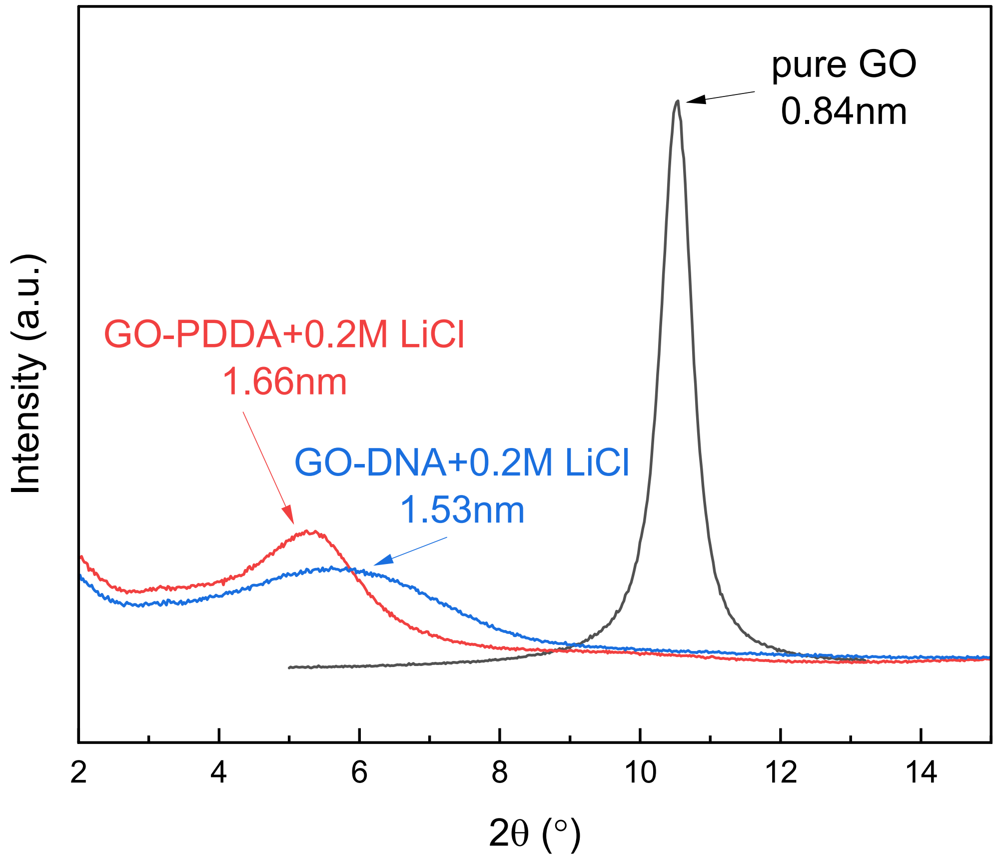

Figure S5. X-ray Diffraction (XRD) spectra of pure GO, GO-DNA with 0.2 M LiCl and GO-PDDA with 0.2 M LiCl. XRD was carried out using Bruker D8 ADVANCE Diffractometer with Cu Kα radiation (λ = 0.1514 nm) using LYNXEYE_XE_T detector in 1D mode. Peaks and respective interlayer distances as indicated.

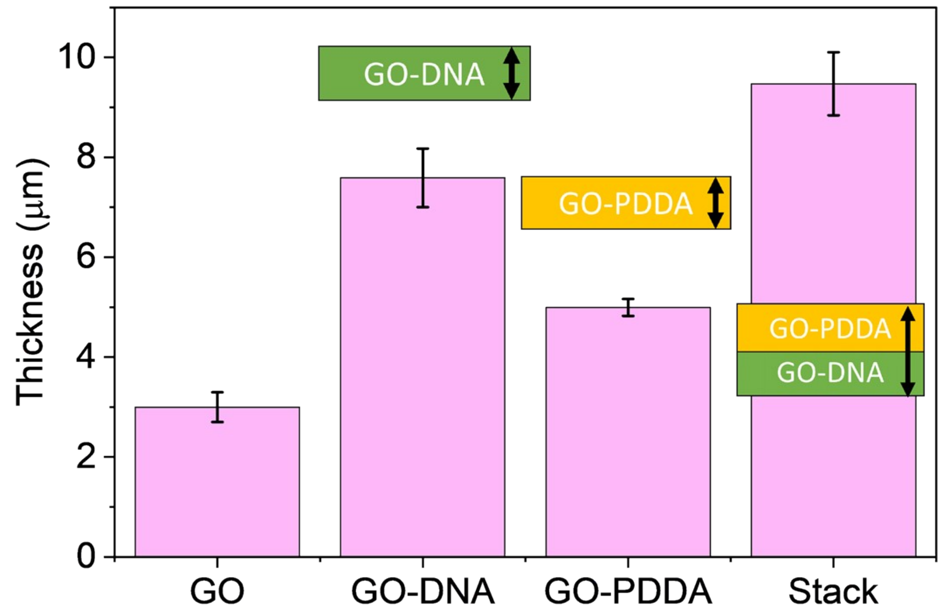

Figure S6 Measured thickness of membranes. KLA TencorAlpha Step IQ Surface Profiler was used with stylus force 8.62 mg and resolution 1 μm. At NTP, membranes of pure GO (without salts), GO-DNA, GO-PDDA and stack with 0.2 M LiCl were measured (schematics illustrated).

Table S1 Measured mass of membrane components. "Added" indicates the mass of each component added for vacuum filtration. "Measured" indicates the final mass of each component, assuming the mass of GO remained constant. Each membraned was left at 30% RH for 24 hours before measurement to exclude water mass.

Component	Added (mg)	Measured (mg)
GO	5	5
PDDA	10	1.3
DNA	10	7

Table S2 Inductively Coupled Plasma (ICP) results of Cl^-, Li^+ and Na^+ concentrations of pure 0.2 M LiCl, filtrate of GO-PDDA after vacuum filtration, filtrate of 0.2 M LiCl through GO-PDDA after vacuum filtration, and similarly for GO-DNA membrane. Excess counterions Cl^- from GO-PDDA filtrate and Na^+ from GO-DNA filtrate indicated the excess PDDA and DNA respectively which were filtrated (values corroborates with measured mass in **Table S1**).

Sample/ Concentration in Molar	Cl^-	Li^+	Na^+
0.2 M LiCl	0.229	0.239	-
GO-PDDA filtrate	2.28 mM	-	-

GO-PDDA+LiCl filtrate	0.156	0.156	-
GO-DNA filtrate	-	-	0.0526 mM
GO-DNA+LiCl filtrate	Not Detected	<0.0146 mM	0.0176 mM

Table S3 Molar concentrations of components. Demonstrating high absorption of LiCl with no selection for cation or anion in the charged GO-PDDA or GO-DNA membranes.

Component	**Calculations for molar concentrations**
GO	From titration, $C_{COO^-}=C_{H^+}$ 2.135×10^{-6} mol of H^+ / 1mg of GO 1.068×10^{-5} mol of H^+ / 5mg of GO
PDDA	M_w of repeating unit = 126 g/mol From mass, C_{PDDA}=1.3mg/126gmol^{-1}= 1.03×10^{-5} mol Each unit has 1 N^+. (**1:1 GO:PDDA** by charges)
DNA	M_w of 1 Base pair of dsDNA = 660 g/mol From mass, C_{DNA}= 7mg/660gmol^{-1}=1.06×10^{-5} mol C_{POO^-}= $2C_{DNA}$= 2.12×10^{-5} mol (**1:2 GO:DNA** by charges)
LiCl in GO-PDDA	From ICP, $C_{Li^+}=C_{Cl^-}=2.2\times10^{-4}$ mol
LiCl in GO-DNA	From ICP, $C_{Li^+}=C_{Cl^-}=1\times10^{-3}$ mol

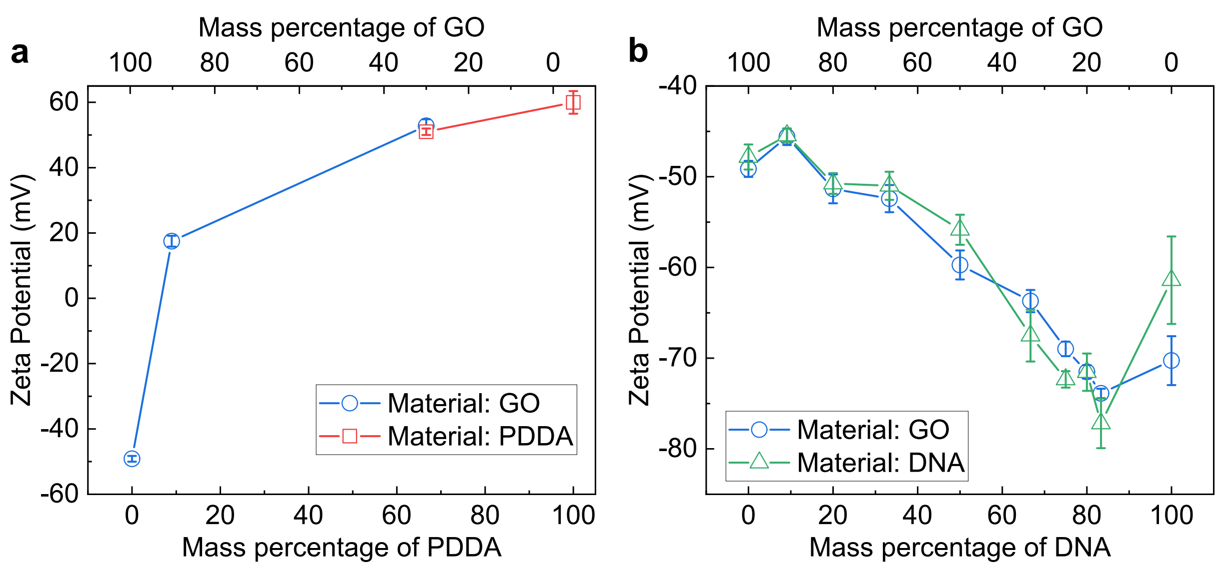

Figure S7 Zeta potential of GO dispersions with varying mass concentrations of **(a)** PDDA and **(b)** DNA, measured using Malvern Panalytical Zetasizer Ultra. **a**, Same samples were measured using both GO and PDDA optical characteristics with high corroboration. GO (blue circles) has refractive index 1.5 and absorption 1; PDDA (red squares) refractive index 1.42 and absorption 0.001. **b,** DNA (green triangles) has refractive index 1.58 and absorption 0.001.

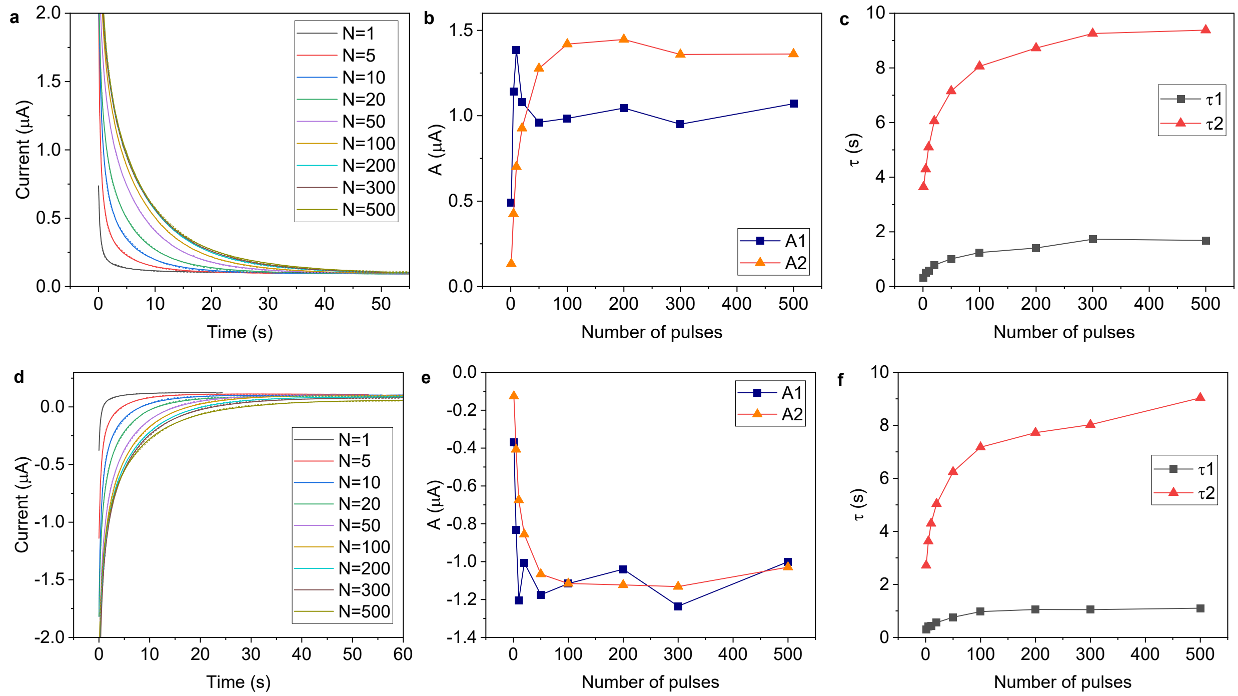

Figure S8 Synaptic behaviour of Synapse 2, a second device made from the same membrane indicating uniformity of device characteristics, also used in demonstration of artificial synapse in computation. **a-c**, Results of excitatory post-synaptic decay curves from 1 to 500 pulses of +1 V with pulse duration of 0.2 s and rest duration of 0.1 s. (**a**) Results of EPSC (in solid lines) and fitted with exponential decay equation (in dotted lines): $I = I_0 + A_1\exp(-t/\tau_1) + A_2\exp(-t/\tau_2)$, where $A_{1,2}$ are plotted in (**b**) and $\tau_{1,2}$ are plotted in (**c**). A showed a continuously increasing amplitude of the decay curve and t showed a continuously increasing time constant of decay as number of pulses increased. **d-f**, Results of IPSC from 1 to 500 pulses of -1 V with the same input parameters and fitting, as plotted in **(e)** and **(f)**.

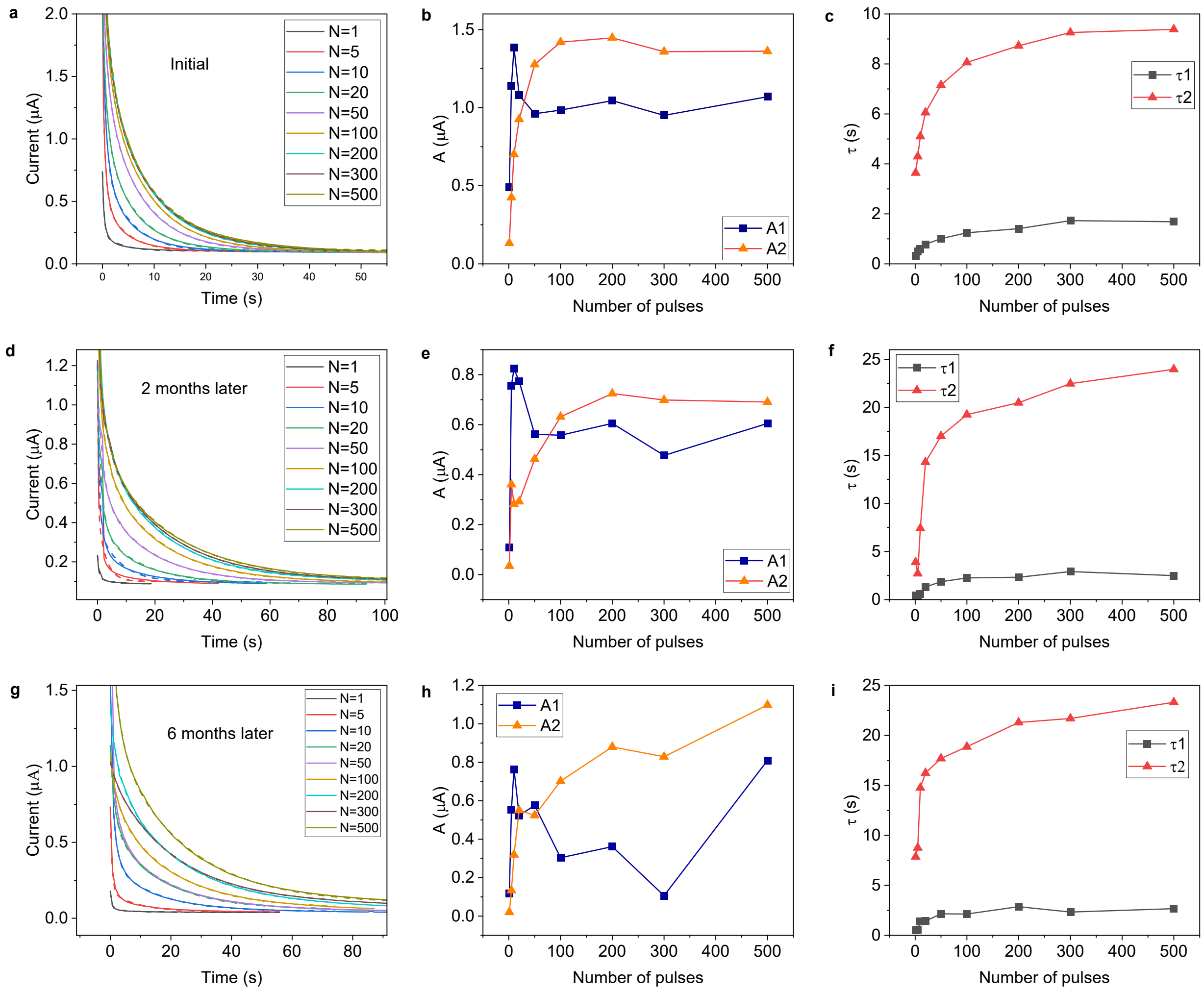

Figure S9 Performance of synapse overtime, at day 0 **(a-c)**, 2 months later **(d-f)** and 6 months later (g-h). There was minimal change in performance after 2 months. These are results of excitatory post-synaptic decay curves from 1 to 500 pulses of +1 V with pulse duration of 0.2 s and rest duration of 0.1 s. (**a,d,g**) show their respective EPSC (in solid lines) and fitted with exponential decay equation (in dotted lines): $I = I_0 + A_1\exp(-t/\tau_1) + A_2\exp(-t/\tau_2)$, where $A_{1,2}$ are plotted in (**b,e,h**) and $\tau_{1,2}$ are plotted in (**c,f,l**) respectively.

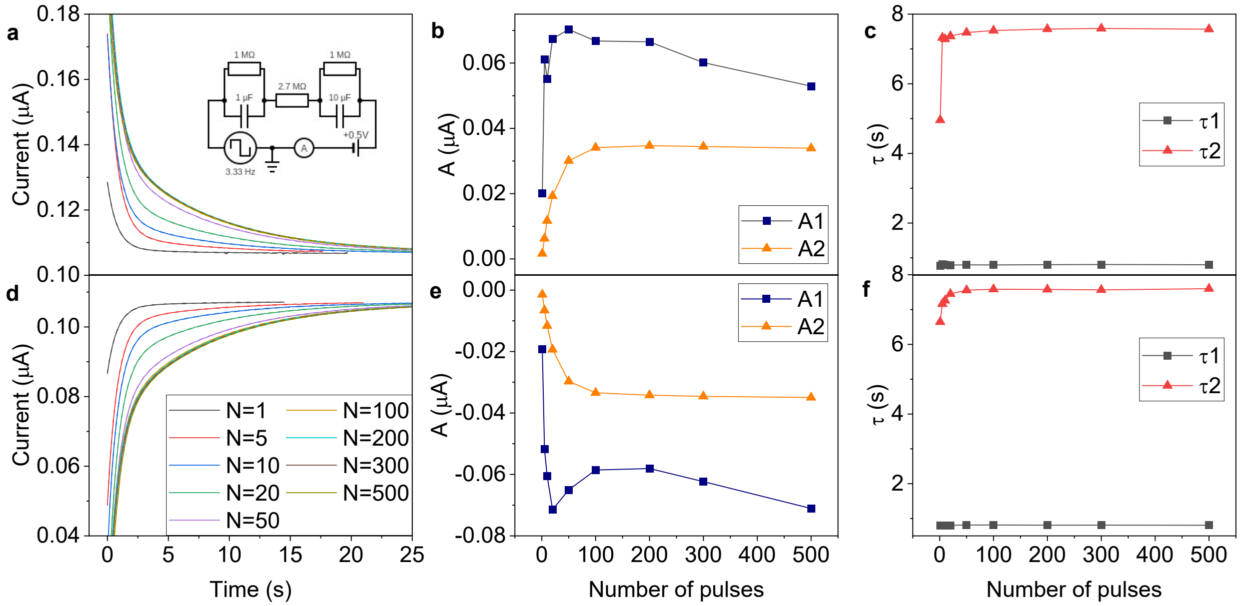

Figure S10 Analog circuit representing synapse using electrical circuit schematic in (**a**). Formation of double layer can be represented as a capacitor with parallel resistance. Two different capacitors and resistances imitate charge/discharge of anions and cations which would give two various time decay constants. 2.7 MΩ resistor in series mimic the resistance of membrane. Signal generator applied pulses on the source electrode with pulse duration of 0.2 s and rest duration of 0.1 s. Sourcemeter was used as ammeter (V_{SD}=0.5 V) to measure source-drain current. **a-c**, Results of EPSC observed with +1 V pulses applied, (**a**) shows decay curves (in solid lines) and individual fitted curves (in dotted lines) according to the exponential decay equation: $I = I_0 + A_1\exp(-t/\tau_1) + A_2\exp(-t/\tau_2)$, where A were plotted in (**b**) and τ in (**c**). **d-f**, Results of IPSC observed with -1 V pulses applied, (**d**) shows decay curves (in solid lines) and individual fitted curves (in dotted lines) according to the exponential decay equation: $I = I_0 + A_1\exp(-t/\tau_1) + A_2\exp(-t/\tau_2)$, where A were plotted in (**e**) and τ in (**f**).

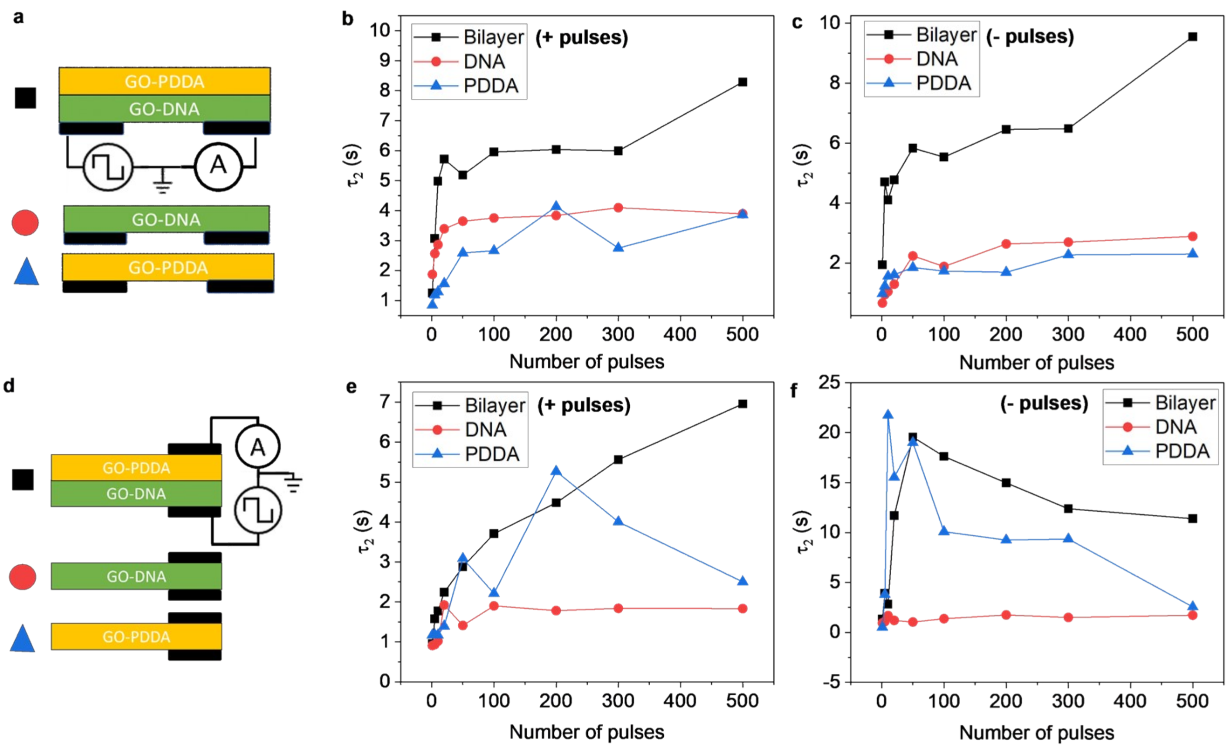

Figure S11 EPSC/ IPSC of single layer membranes. **a**, Schematic of electrical circuit used for (**b**)-(**c**) along the membrane. **d**, Schematic of electrical circuit used for (**e**)-(**f**) for across the membranes. +1 V pulses were applied in **(b,e)** (-1V in **(c,f)**) on the source electrode with pulse duration of 0.2 s and rest duration of 0.1 s. Sourcemeter was used as ammeter (V_{SD}=0 V) to measure source-drain current. EPSC (or IPSC) decay curves were fitted with the exponential decay equation: $I = I_0 + A_1\exp(-t/\tau_1) + A_2\exp(-t/\tau_2)$, where τ_2 were plotted as the higher time decay constant and major contribution on synaptic behaviour.

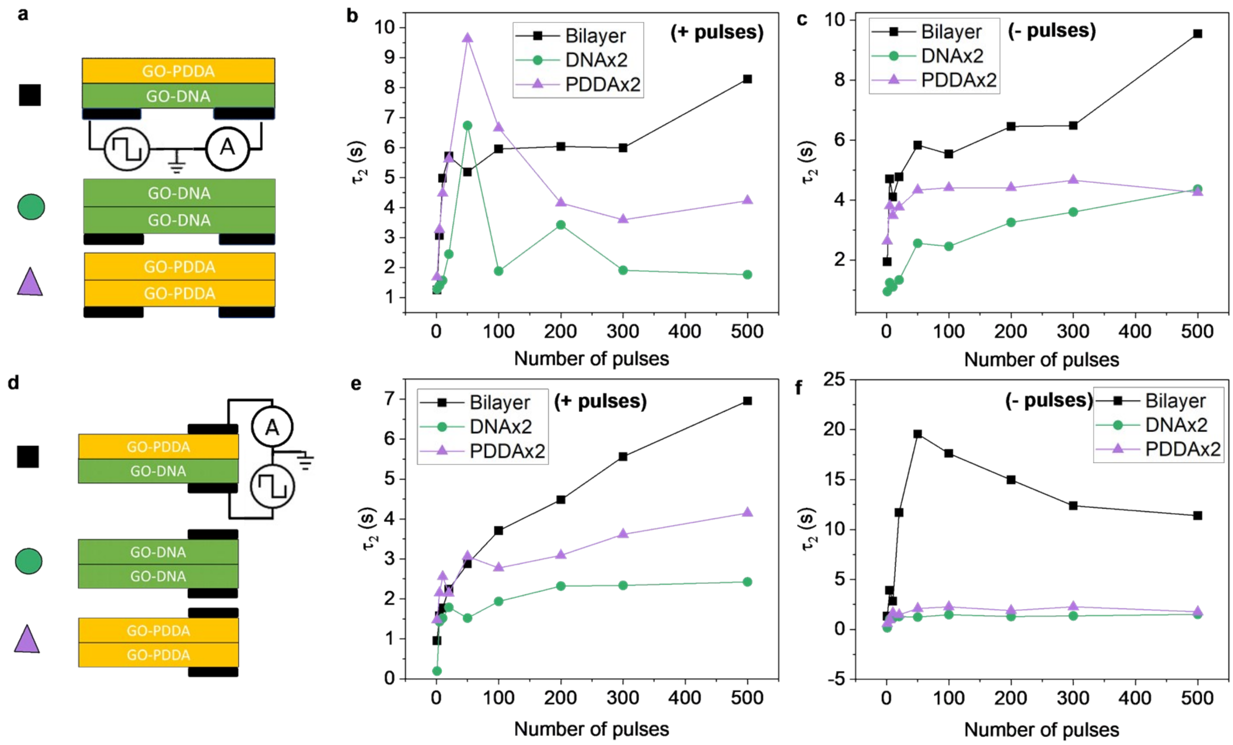

Figure S12 EPSC/ IPSC of double layer membranes. **a**, Schematic of electrical circuit used for (**b**)-(**c**) along the membrane. **d**, Schematic of electrical circuit used for (**e**)-(**f**) for across the membranes. +1 V pulses were applied in **(b,e)** (-1V in **(c,f)**) on the source electrode with pulse duration of 0.2 s and rest duration of 0.1 s. Sourcemeter was used as ammeter (V_{SD}=0 V) to measure source-drain current. EPSC (or IPSC) decay curves were fitted with the exponential decay equation: $I = I_0 + A_1 exp(-t/\tau_1) + A_2 exp(-t/\tau_2)$, where τ_2 were plotted as the higher time decay constant and major contribution on synaptic behaviour. The combination of GO-DNA and GO-PDDA membranes is essential to synaptic behaviour as behaviour is not replicated in double GO-DNA and double GO-PDDA membranes.

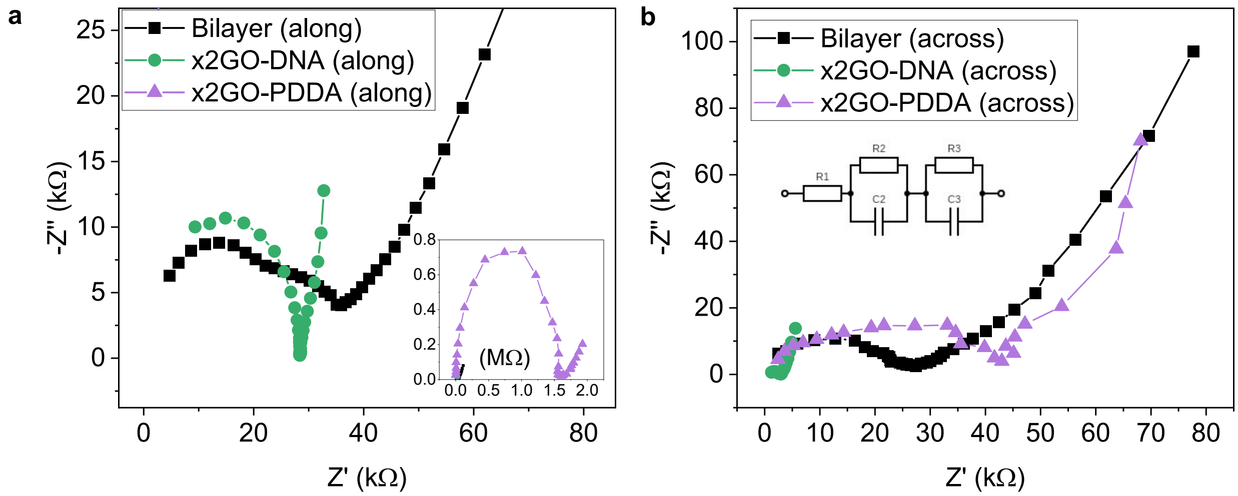

Figure S13 Nyquist plot of double layer membranes. Schematics of membrane stacking are found in **S12(a)** for along and **S12(d)** for across. The inset in (**a**) shows high impedance region. These double membranes were fitted to second-order RC model as in **(b)**. AC impedance measurements estimated frequency-dependent resistances of membranes. Fitted resistance values in **Figure S14**.

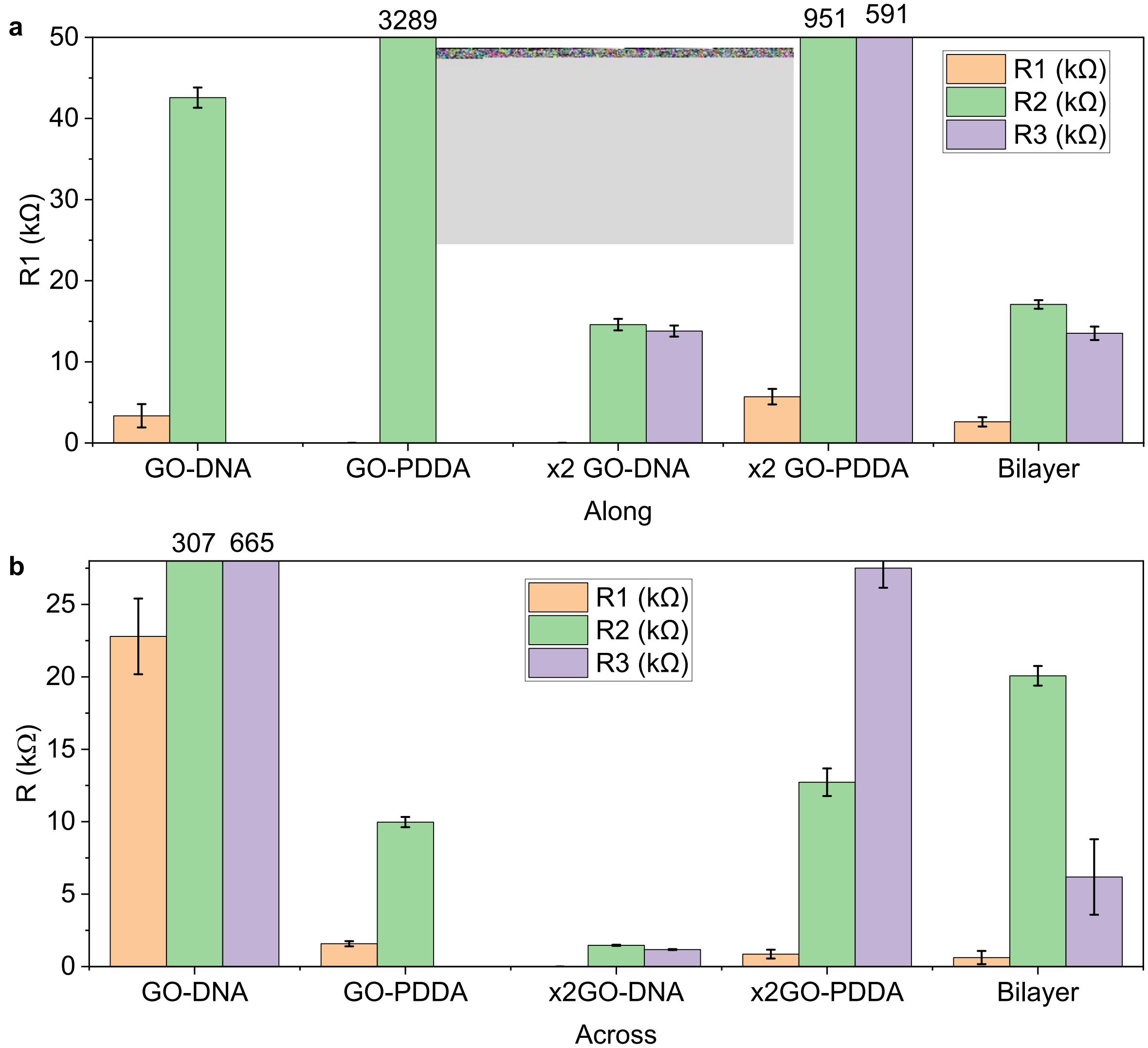

Figure S14 Fitted resistance of Electrochemical Impedance Spectroscopy (EIS) measurements in various configurations for along **(a)** and across **(b)** the membranes. Circuit model drawn in **(a)** for two RC circuit, which was used for all double layer membranes and across GO-DNA membrane due to its phase separation between GO and DNA. While single RC circuit model was used for the rest of the single layer membranes. It is expected that resistance along the membrane is higher than across the membrane is the distance between electrodes along the membrane is 1.7 cm while the thickness across the membrane is roughly ~10 µm on average.

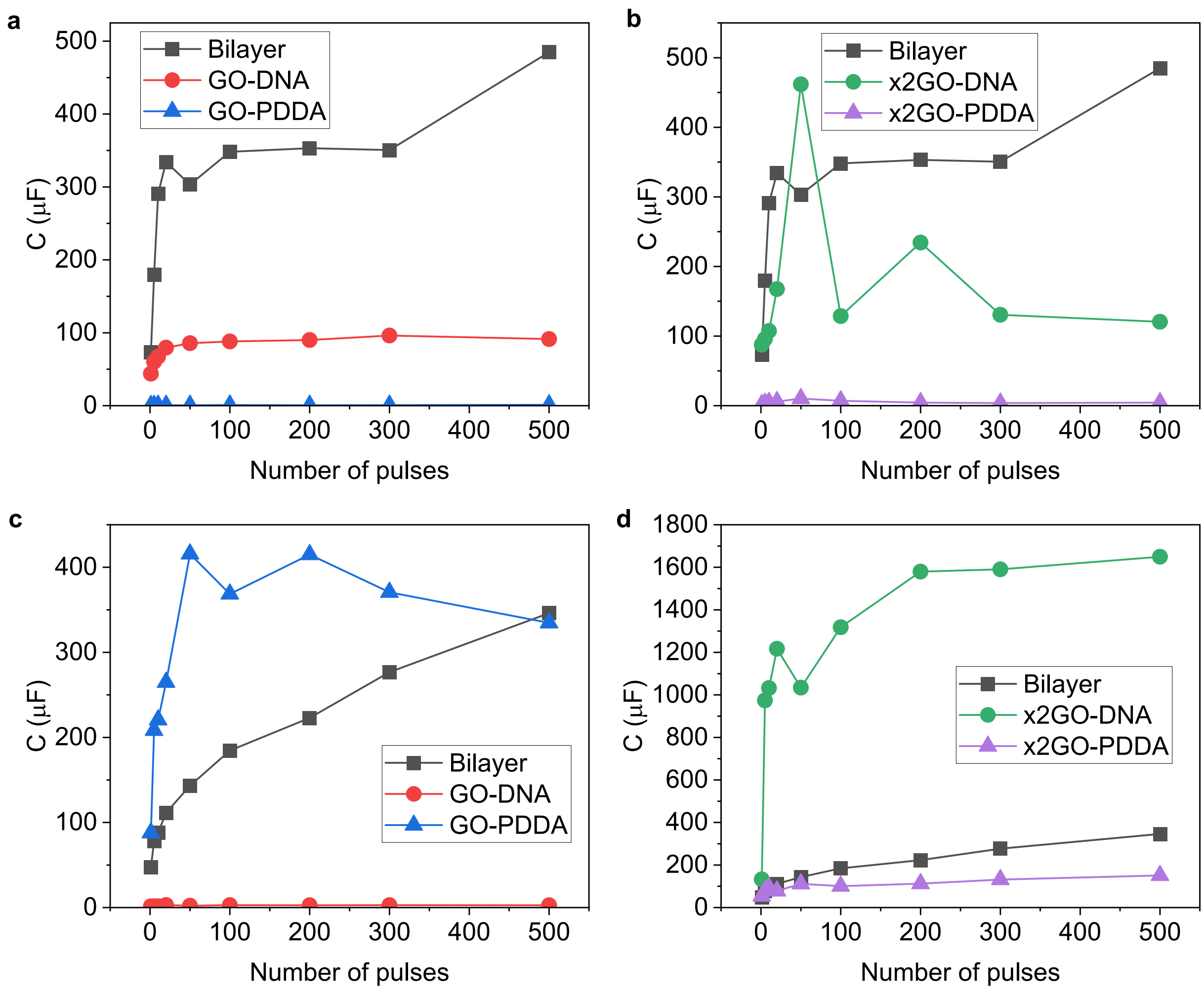

Figure S15 Calculated capacitances of participating ions. Capacitance was calculated from τ_2=RC, where τ_2 was taken from **S11, S12** and the higher resistance R was taken from **S14**, both selected as major contribution to synaptic behaviour. **a,b** for along membrane and **c,d,** for across membrane. See **S11** and **S12** for schematics of respective membranes.

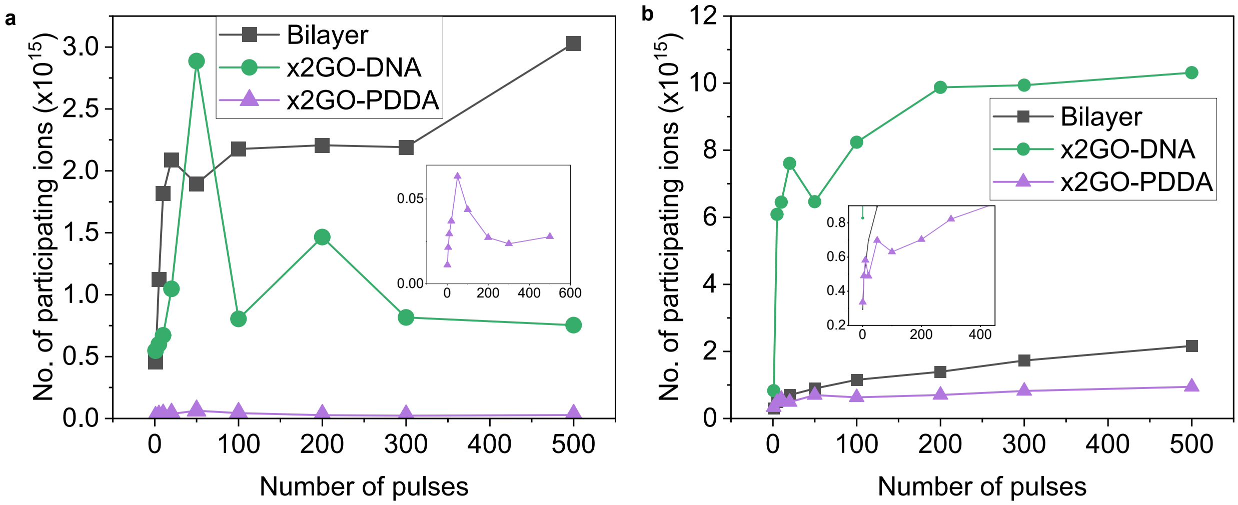

Figure S16 Calculated number of participating ions for double layer membranes. Charge transferred, Q=CV from **S15** determines the number of Li^{+} and Cl^{-} ions participating in charge transfer, where V=1V of applied pulses. **a,** for the along membrane and **b,** for the across membrane. Onset is an enlarged figure showing x2 GO-PDDA behavior. See **S12a,d** for schematics of respective membranes.

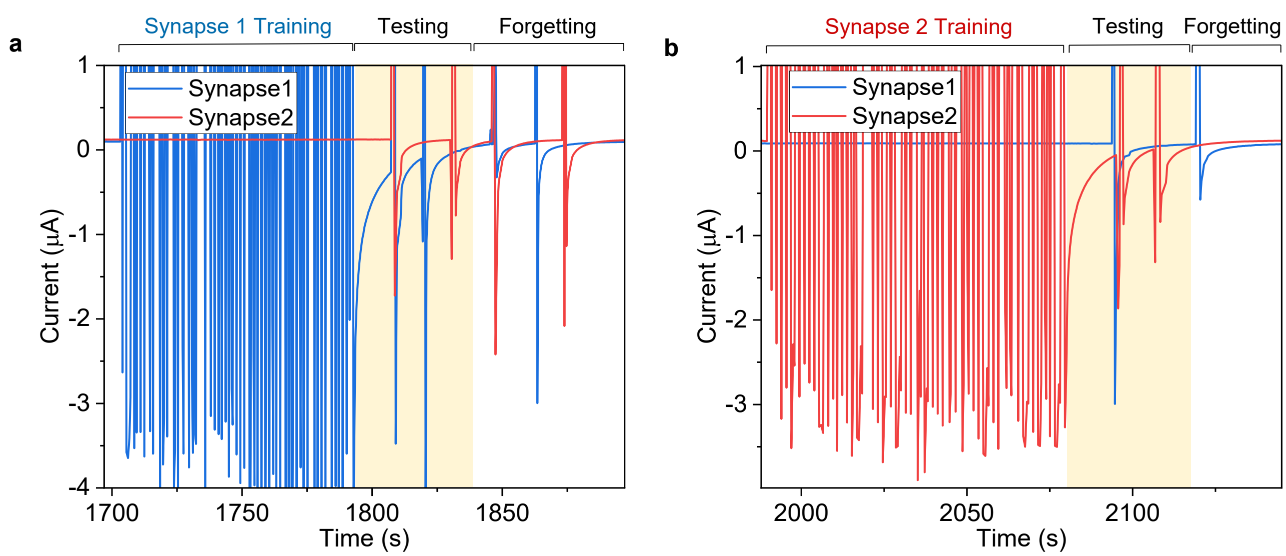

Figure S17 Binary logic functions with trained and untrained synapses. **a**-**b,** IPSC of two synapses measured as one of the synapses was trained with 300 pulses of -1 V with pulse duration of 0.2 s and rest duration of 0.1 s. Either or both synapses were tested with 5 pulses of -1 V with pulse duration of 0.2 s and rest duration of 0.1 s, but trained synapse constantly remain of higher current for approximately 30 s. (**a**) Synapse 1 (in blue) was trained. (**b**) Synapse 2 (in red) was trained.

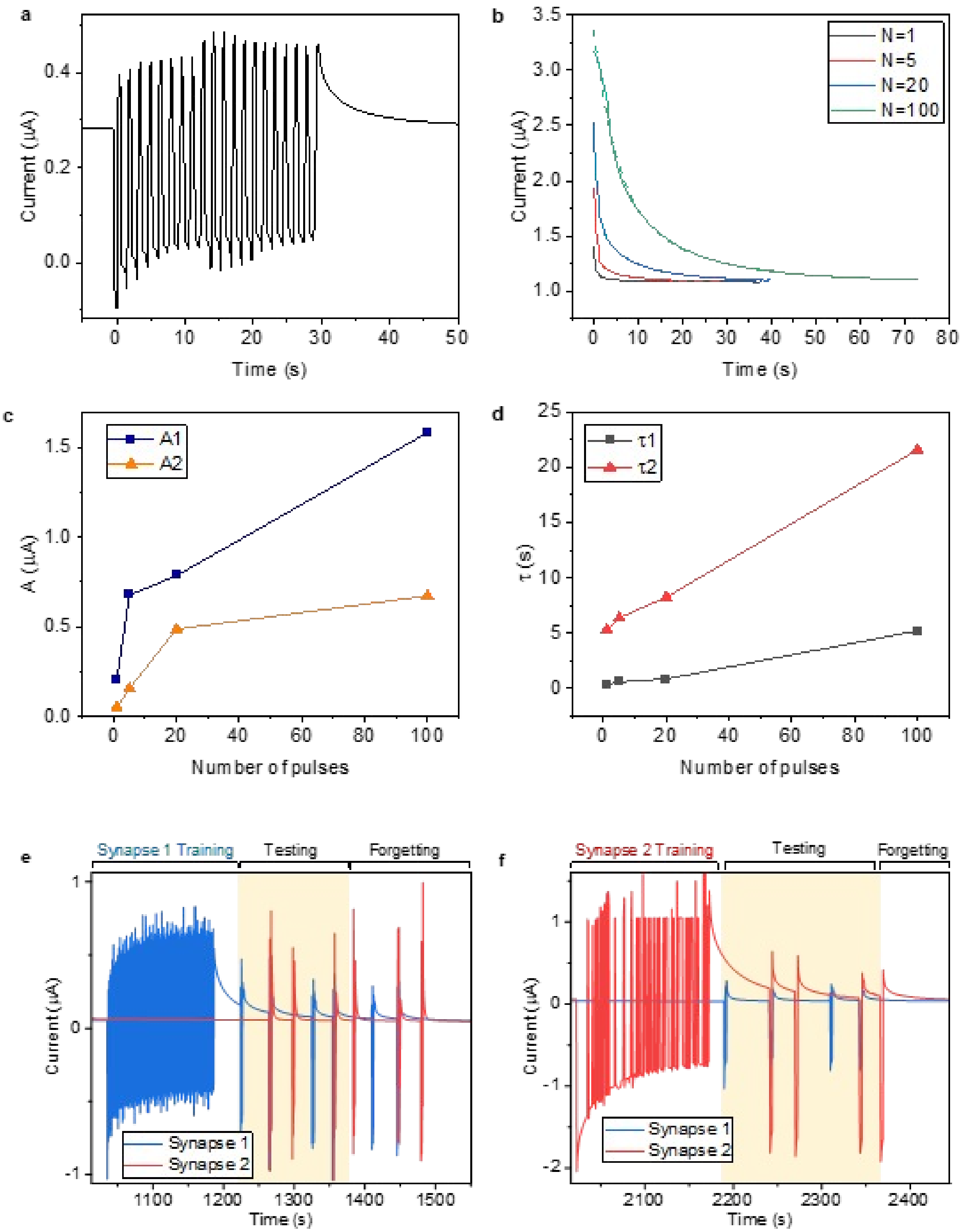

Figure S18 Electrical measurements of synapse using graphene dispersion with silver paste electrode, indicating the absence of redox reactions for synaptic behavior. **a,** Paired-pulse facilitation (PPF) of synapse. The plot of source-drain current exhibited the EPSC as current increased after each pulse and slowly decayed over time. Hundred +1 V pulses were applied with pulse duration of 0.2 s and rest duration of 0.1 s each. **b-d**, Results of excitatory post-synaptic decay curves from 1 to 100 pulses of +1 V with pulse duration of 0.2 s and rest duration of 0.1 s. (**b**) Results of EPSC (in solid lines) which are fitted with quite high accuracy with decay equation: $I = I_0 + A_1 exp(-t/\tau_1) + A_2 exp(-t/\tau_2)$, where $A_{1,2}$ are plotted in (**c**) and $\tau_{1,2}$ are plotted in (**d**). Fitted curves completely coincide with experimental curves. $A_{1,2}$ showed a continuously increasing amplitudes of the decay curve, while $\tau_{1,2}$ demonstrated a continuously increasing time constant of decay as number of pulses increased. **e-f,** Binary logic functions with trained and untrained synapses. EPSC of two synapses measured as one of the synapses was trained with 500 pulses of +1 V with pulse duration of 0.2 s and rest duration of 0.1 s. Either or both synapses were tested with 5 pulses of +1 V with pulse duration of 0.2 s and rest duration of 0.1 s, but trained synapse constantly remain of higher current for at least 100 s. (**a**) Synapse 1 (in blue) was trained. (**b**) Synapse 2 (in red) was trained.