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Supporting Information

for

Organic Synaptic Devices based on Ionic Gel with reduced Leakage Current

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Experimental Section

Materials

High mobility p-type polymer semiconductor, poly[[2,3,5,6-tetrahydro-2,5-bis(2-octyldodecyl)-3,6-dioxopyrrolo[3,4-c]pyrrole-1,4-diyl]-2,5-thiophenediylthieno[3,2-b]thioph-ene-2,5-diyl-2,5-thiophenediyl] (DPP-DTT), was purchased from Ossila Ltd. P(VDF-TrFE) was purchased from piezotech Co., Ltd. P(VDF-HFP) with the molecular weight of 400000 and ionic liquid [EMI-TFSI] were purchased from TCI Co., Ltd. and Sigma-Aldrich Co., Ltd. respectively. All chemicals were used as received without further purification.

Device Fabrication

50 mg ml⁻¹ P(VDF-TrFE) was dissolved in 2-butanone and heated at 70°C until a homogeneous solution was formed. P(VDF-HFP) and ionic liquid [EMI-TFSI] were dissolved in acetone in a weight ratio of 1:4:7 and heated at 70°C. The P(VDF-TrFE) and the P(VDF-HFP)/[EMI⁺TFSI⁻] gel solutions were mixed with a volume ratio of 98:2 and a homogeneous solution was achieved through stirring for 1h. The synaptic transistors were made using top gate/Bottom contact architecture on silicon wafers. The wafers as substrates were cleaned in the ultrasonic bath with acetone and isopropanol sequentially, followed by rinsing with methanol and water, and then dried by N₂ flow. The bottom electrodes (Au/Cr) were thermally evaporated onto the substrates through a shadow mask. The channel length (L) and width (W) were 0.08 mm and 2 mm, respectively. 5 mg ml⁻¹ DPP-DTT chloroform solution was spin-coated on the wafer substrate at 1500 rpm for 60 s and baked at 140°C for 1h in the glove box to remove the residual solvent. Then the dielectric layer was prepared by spin-coating mixed electrolyte solution at 1500 rpm for 60 s, followed by annealing at 110°C for 1h in the glove box. Finally, the top gate electrode was thermally evaporated on dielectric film through a metal mask.

Device characterization

The surface morphologies of dielectric films involving pure PVDF and IL/PVDF were observed by AFM (Bruker Dimension Icon). Infrared spectroscopy of pure P(VDF-TrFE), pure P(VDF-HFP), hybrid films were characterized by using a Nicolet 380 FT-IR spectrometer. The SEM images of the dielectric films were obtained from scanning electron microscopy (ZEISS, Sigma 300 VP). The electrical performance and synaptic behaviors of

the devices were characterized by a Keithley 4200-SCS semiconductor parameter instrument.



Figure S1. The FT-IR spectrums of the films (PVDF-TrFE, HEDF, PVDF-HFP)



Figure S2. The AFM images (phase angle) of the HEDF film and PVDF film



Figure S3. (a) The cross-section optical microscope image of the hybrid bi-polymer/ionic dielectric film with the thickness of $\sim 5\mu m$ (scale bar, 20 μm). (b) The frequency-dependent effective capacitance (inset, the device structure for the test).



Figure S4. The fabricating procedure of the HEDF-based synaptic transistor



Figure S5. The hysteresis loop windows based on different scan rates.



Figure S6. (a) The Δ EPSC behaviors of the HEDF-based transistor triggered by presynaptic spikes with various duration time (0.87, 1.28, 2.21, 3.10 s). (b) The dependence of the Δ EPSC values on spike duration time. (c) The dependence of the energy consumptions on spike duration time. (d) The Δ EPSC behaviors of the HEDF-based transistor triggered by presynaptic spikes with various intensities (-0.5, -1.0, -1.5, -2.0V). (e) The dependence of the Δ EPSC values on spike intensity. (f) The dependence of the energy consumptions on spike intensity.



Figure S7. (a) The typical EPSC behaviors of nine devices triggered by -1V with the width of 0.5s. (b) The typical EPSC behaviors of nine devices triggered by -1V with the width of 1.4s.