

## Supporting Information

### **Polymer Bulk-heterojunction Synaptic Field-Effect Transistors with Tunable Decay Constant**

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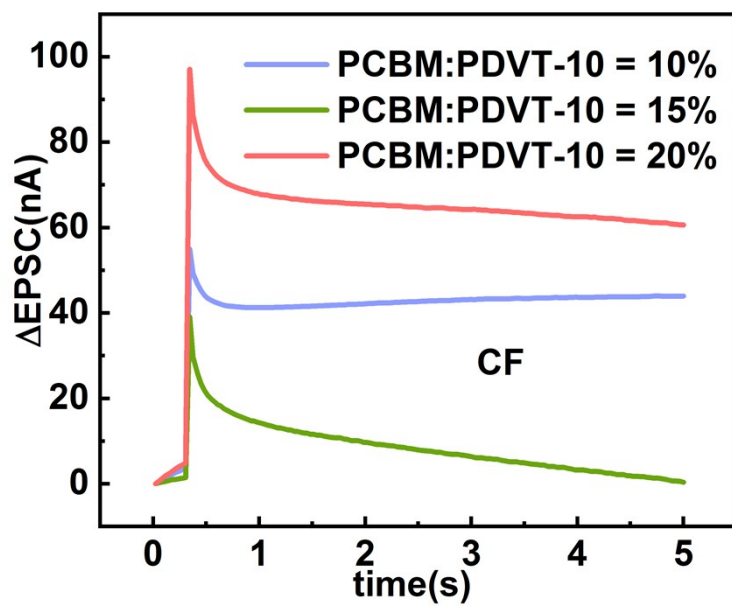
## 1. Experimental Section

*Preparation of Materials:* The p-type highly  $\pi$ -extended organic D-A copolymer poly[2,5-bis(alkyl)pyrrolo[3,4-c]pyrrolo-1,4(2H,5H)-dione-alt-5,5-di(thiophene-2-yl)-2,2-(E)-2-(2-(thiophen-2-yl)-vinyl)thiophene] (PDVT-10) ( $M_w = 183 \text{ kg mol}^{-1}$ ) was purchased from 1- Materials with a concentration 5 mg/mL and [6,6]-phenyl C<sub>61</sub> butyric acid methyl ester (PC<sub>61</sub>BM) was acquired from Solenne BV. The mixture of PDVT-10/PC<sub>61</sub>BM (at a weight ratio of 20:3) was dissolved in a mixture solvent of chloroform (CF)/chlorobenzene (CB). The weight ratio of PC<sub>61</sub>BM was optimized by comparison of different ratios of blend composition, and optimum synaptic performance was obtained for blends containing 15 wt % PCBM in all cases. In this work, the mixture solvent was mixed with CF and CB at various volume ratios 10:0, 9:1, 8:2, 7:3, 6:4, and 5:5, respectively. The solution of PDVT-10/PC<sub>61</sub>BM was heated at 60 °C for 1 day.

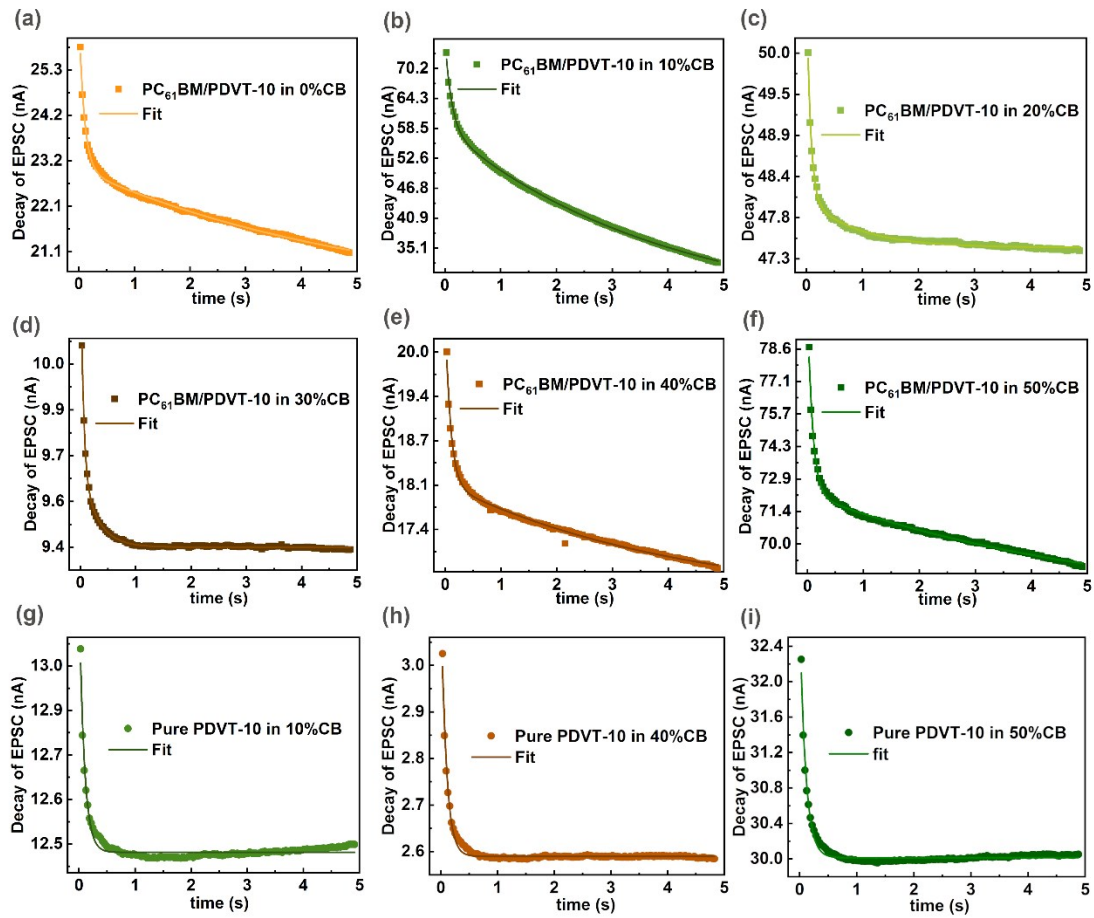
*Synaptic Transistors Fabrication:* OFET devices were fabricated in a bottom-gate top-contact (BGTC) configuration. Heavily n-type doped Si wafers with thermally grown 100-nm-thick SiO<sub>2</sub> as dielectric layers were used as substrates, which were fully cleaned by sequential ultrasonication in acetone, isopropanol, and deionized H<sub>2</sub>O, respectively, and dried with a nitrogen blower. The SiO<sub>2</sub> layer was modified by OTS at 60 °C for 20 min to form an OTS self-assembled monolayer. Subsequently, PDVT-10/PC<sub>61</sub>BM solutions with various CF and CB ratios were spin-coated on the dielectric layer at 1000 rpm for 1 min as semiconductor layers and were annealed at 30 °C for 30 min in the air as followed. Finally, 50-nm-thick source and drain electrodes made of

gold were deposited on the film by vacuum-evaporation.

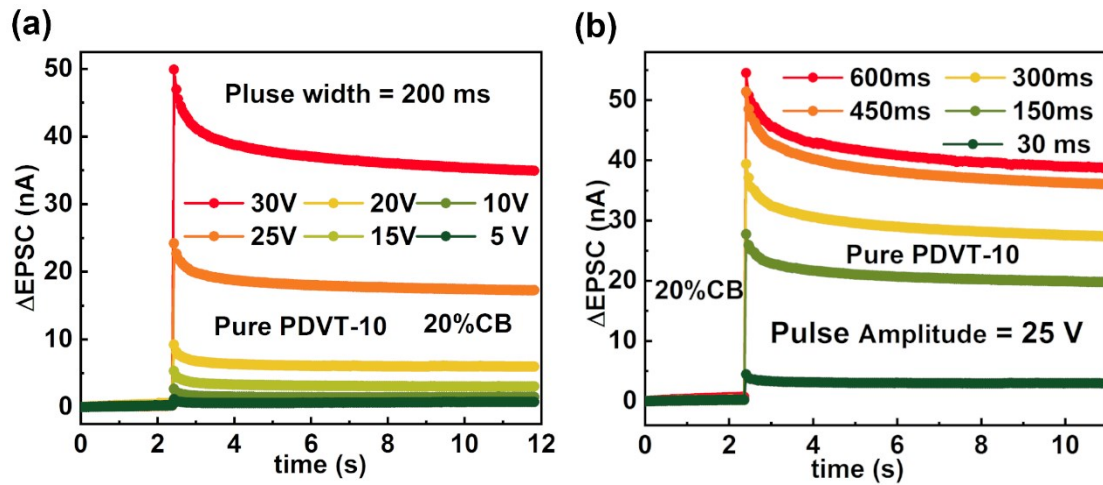
*Electric measurement:* The electric properties and basic synaptic behaviors were measured by the semiconductor parameter analyzer (Keithley B2902A) in an ambient environment. Atomic force microscopy (AFM) was performed using a Veeco Nanoscope IV in the tapping mode. Small-angle neutron scattering (SANS) was performed in China Spallation Neutron Source (CSNS).



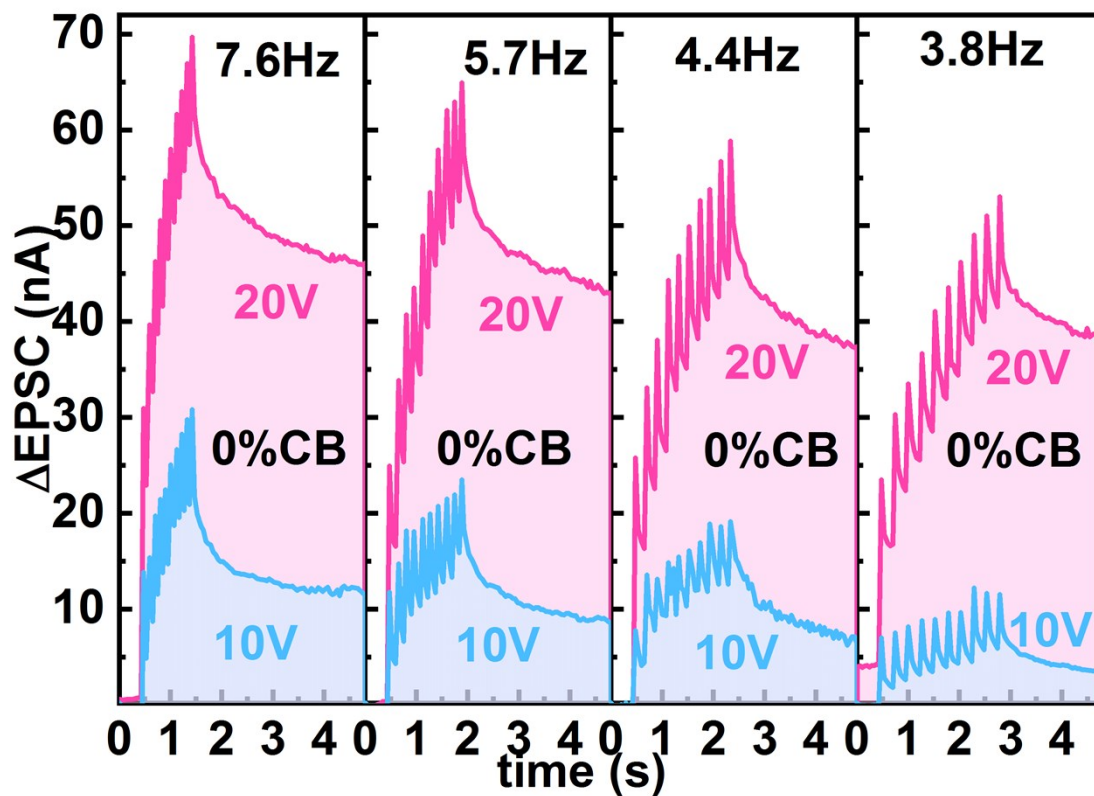
**Figure S1** Comparison of EPSC of synaptic device in 10%, 15%, and 20% CB respectively. EPSC of PDVT-10/ $\text{PC}_{61}\text{BM}$  synaptic transistors triggered by a presynaptic spike with  $V_{\text{pre}} = -35$  V,  $t_d = 60$  ms and  $V_{\text{ds}} = -10$  V.



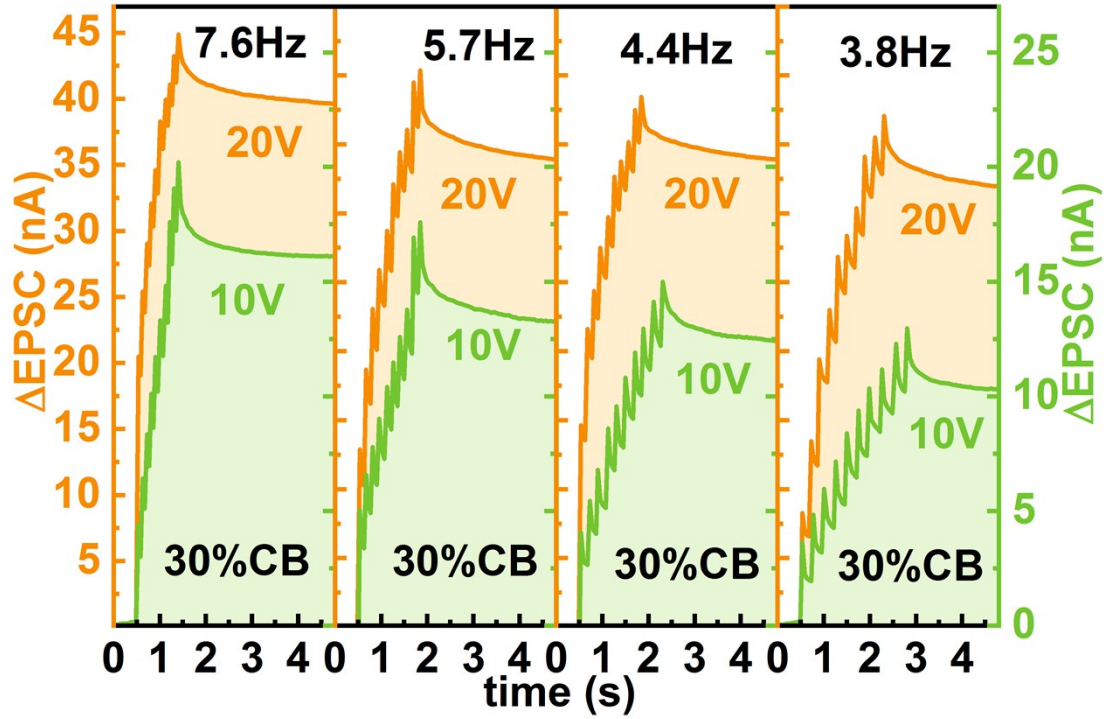
**Figure S2** For the synaptic device with PDVT-10/ $PC_{61}BM$  a) in CF, b) in 10%CB, c) in 20%CB, d) in 30%CB, e) in 40%CB, f) in 50%CB, the decay part of postsynaptic current can be fitted using a double-exponential function. For the synaptic device with pure PDVT-10 g) in 10%CB, h) in 40%CB, i) in 50%CB, the decay part of postsynaptic current can be fitted using a single-exponential function.



**Figure S3** a) The EPSC of synaptic device triggered by a single spike with the same time of duration 200 ms and different presynaptic spike voltages from 5 to 30 V. b) The EPSC of synaptic device triggered by a single spike with the same spike amplitude 25V and different presynaptic times of duration from 30 to 600 ms.

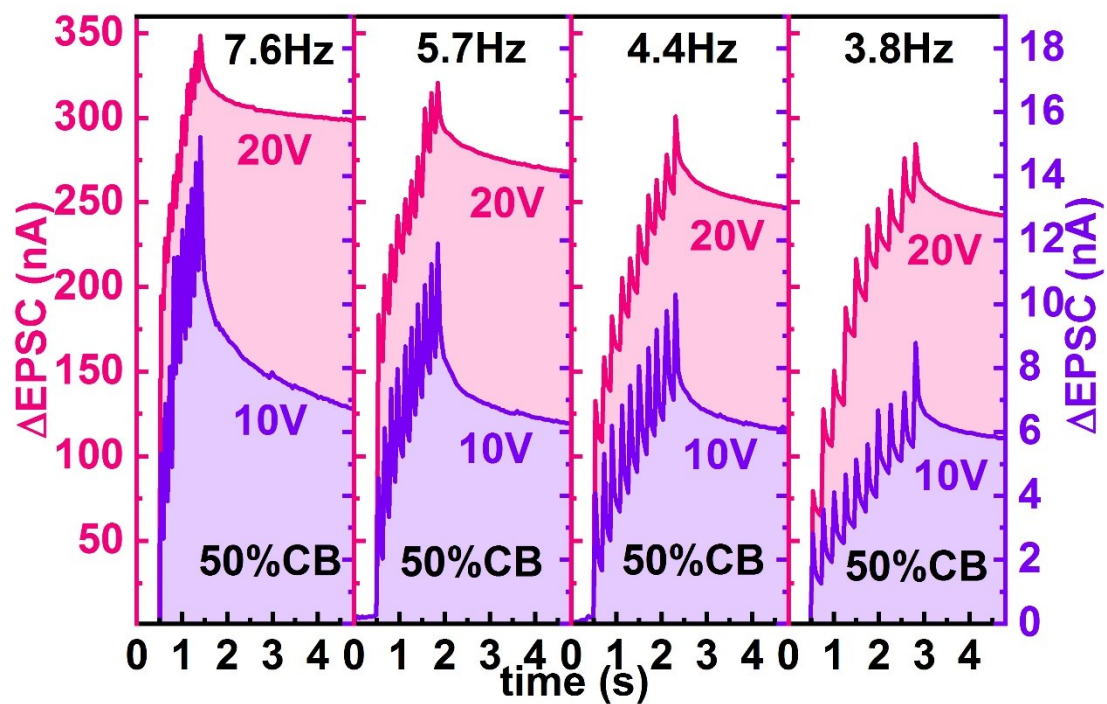


**Figure S4** The EPSC of PDVT-10/PC<sub>61</sub>BM synaptic device in CF in the frequency of the stimulus spikes rises from 3.8 to 7.6 Hz and the presynaptic voltage rises from 10 V to 20V.

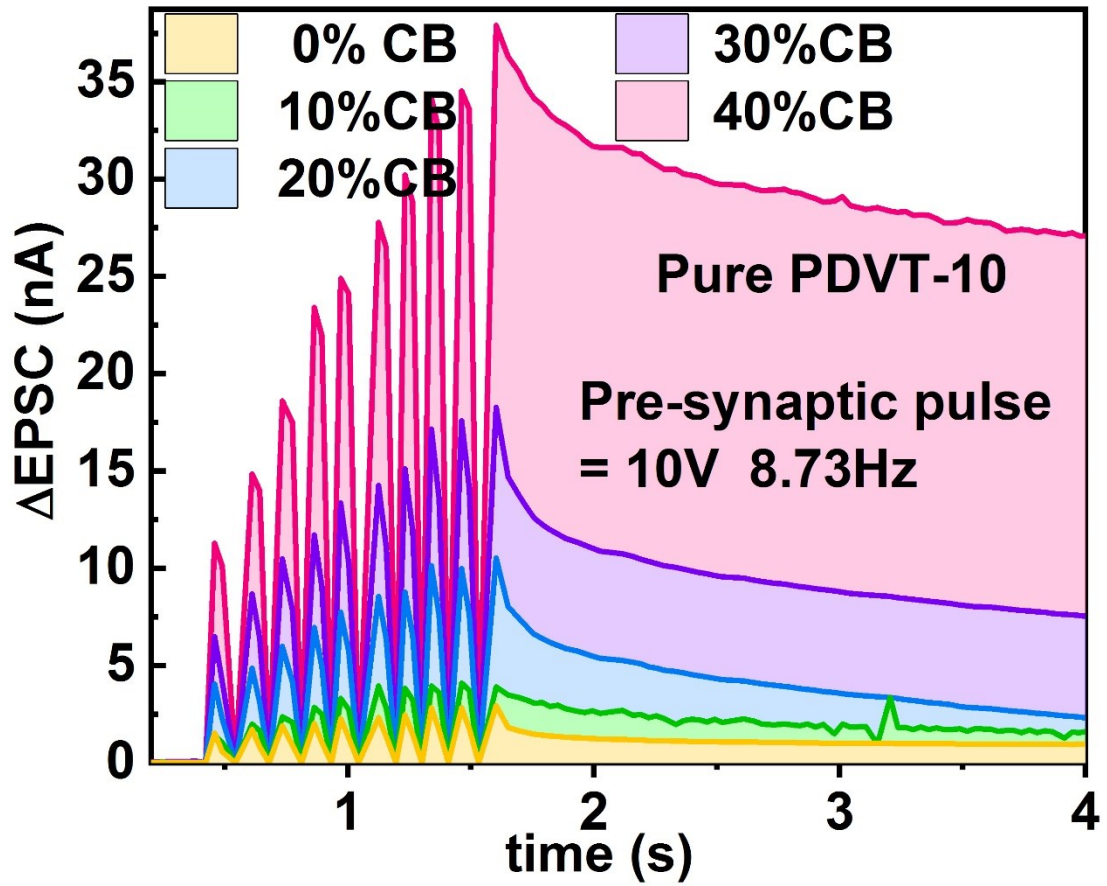


**Figure S5** The EPSC of PDVT-10/PC<sub>61</sub>BM synaptic device in 30%CB in the frequency of the stimulus spikes rises from 3.8 to 7.6 Hz and the presynaptic voltage rises from 10 V to 20V.

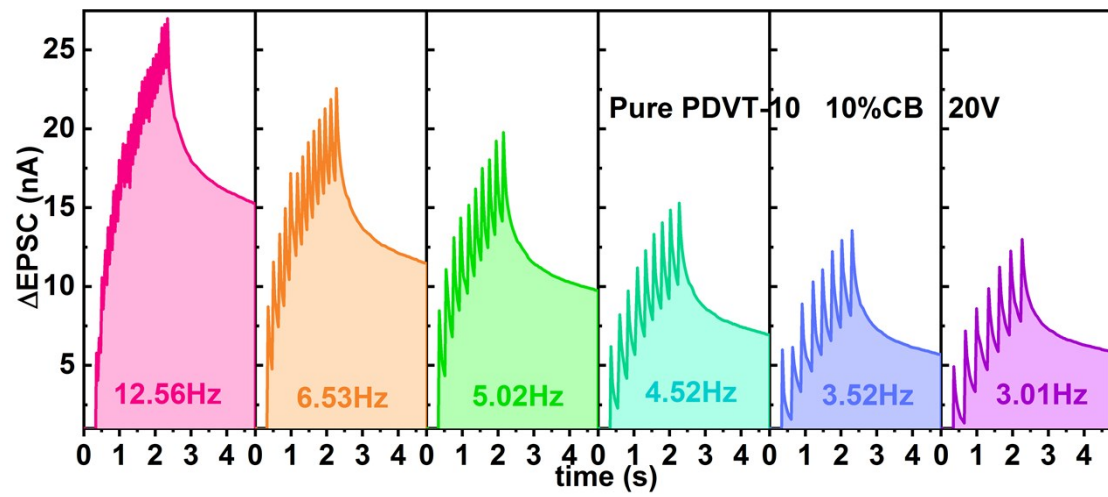




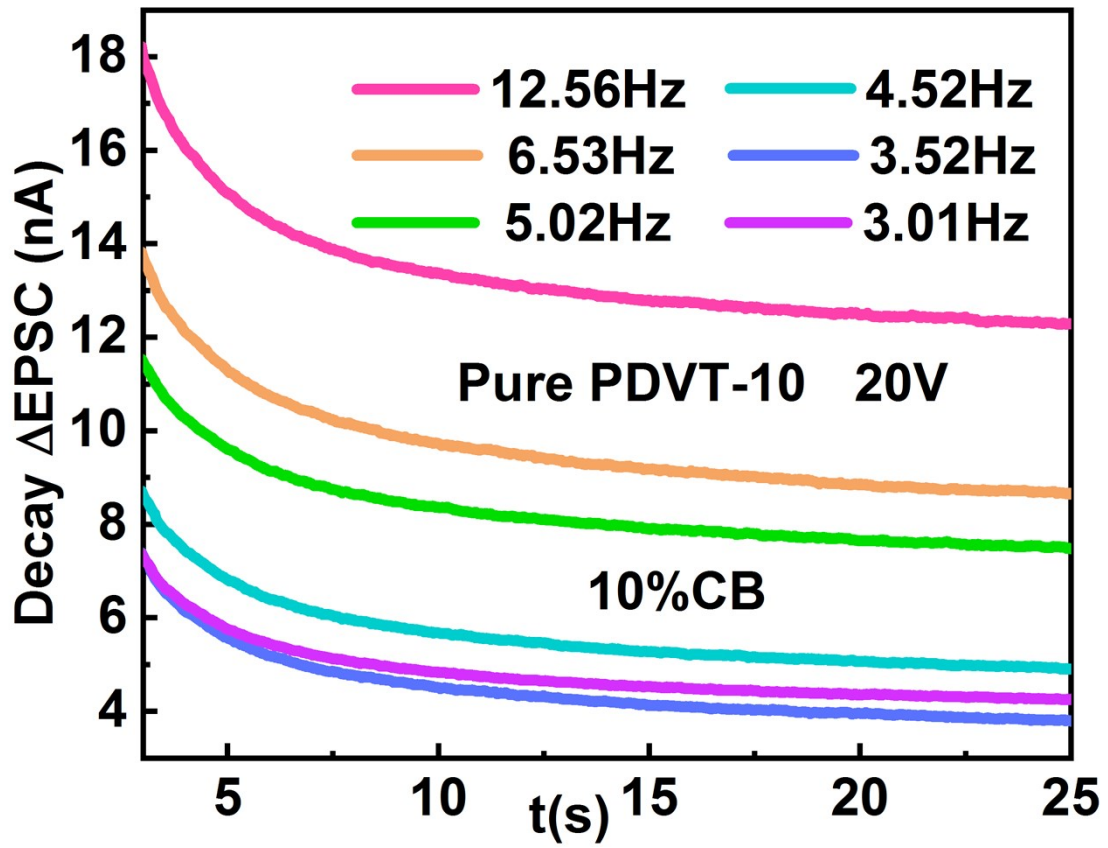
**Figure S6** The EPSC of PDVT-10/PC<sub>61</sub>BM synaptic device in 50%CB in the frequency of the stimulus spikes rises from 3.8 to 7.6 Hz and the presynaptic voltage rises from 10 V to 20V.



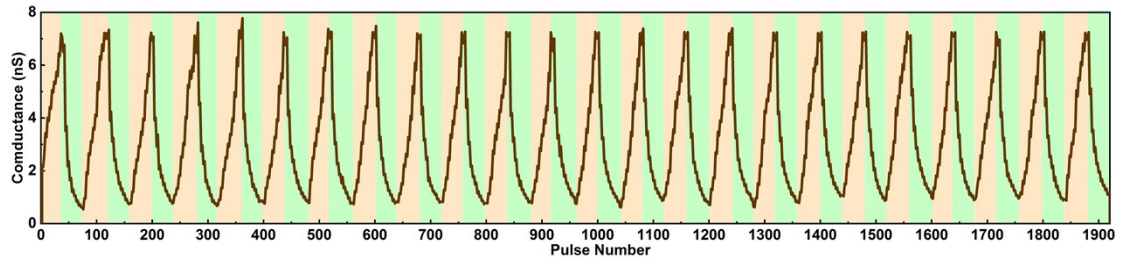
**Figure S7** The EPSC of pure PDVT-10 synaptic device in vary ratios of CB from 0% to 50% . The frequency of repeated synaptic spikes was 8.73 Hz and 10V.



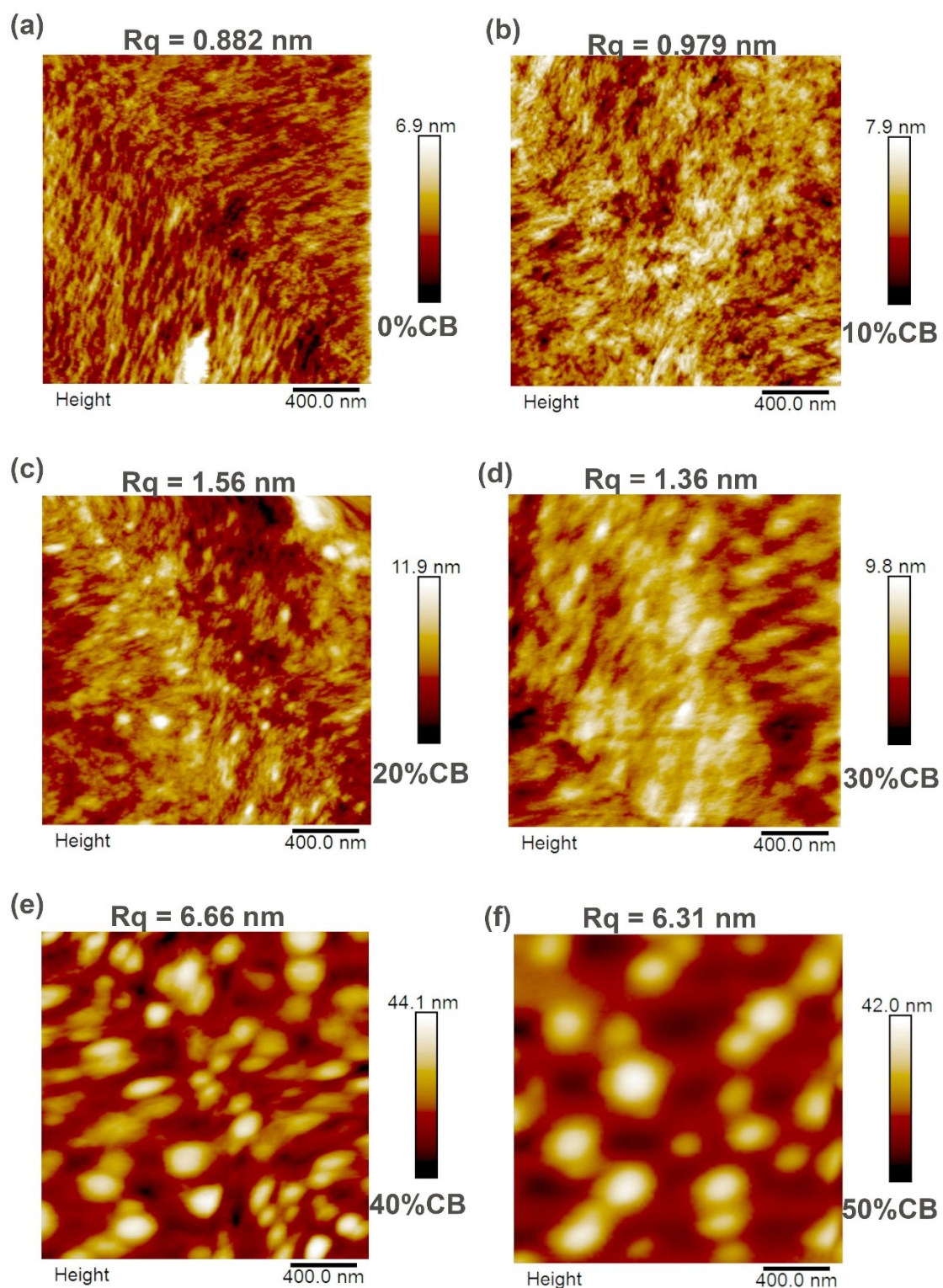
**Figure S8** The EPSC of pure PDVT-10 synaptic device in 10%CB in the frequency of the stimulus spikes rises from 3.01 to 12.56 Hz.



**Figure S9** The decay part of EPSC of pure PDVT-10 synaptic device in 10% CB in the frequency of the stimulus spikes rises from 3.01 to 12.56 Hz.



**Figure S10** The reproducibility of PDVT-10/PC<sub>61</sub>BM SFET for 24 cycles (10 pulses positive pulse and 10 negative pulses with duration of 200 ms and interval of 200 ms).



**Figure S11** AFM tapping mode topographies of the surfaces of PDVT-10/PC<sub>61</sub>BM blends in (a) 0 % CB, (b) 10 % CB, (c) 20 % CB, (d) 30 % CB, (e) 40 % CB, and (f) 50 % CB.