Electronic Supplementary Material (ESI) for Materials Horizons.
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Supporting Information

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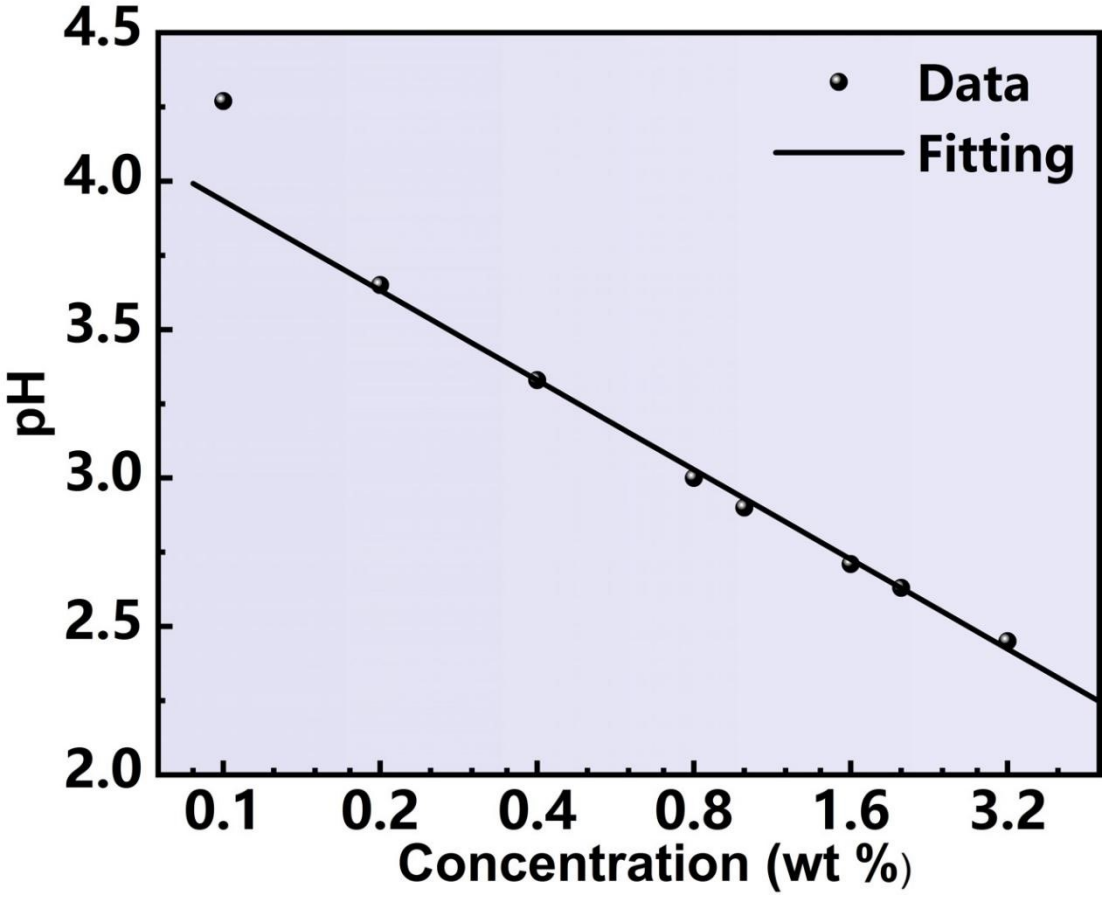

Figure S1 The pH value as a function of acid dopant ratio.

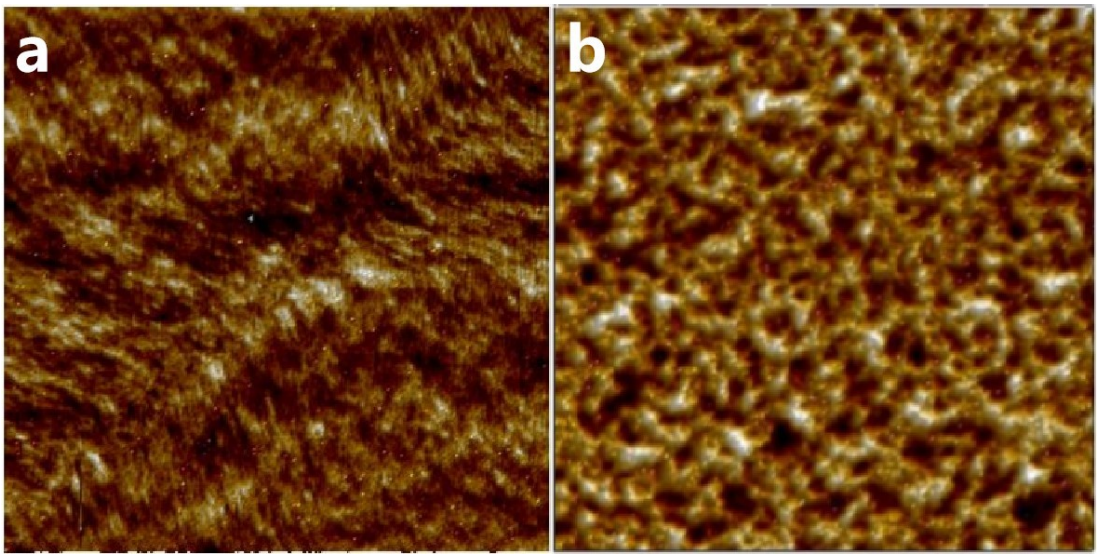

Figure S2 The AFM images of a) 2.0 wt% acid-doped PVA layer b) PDVT-10 layer.

As shown in Figure S2a, the AFM images of 2.0 wt% acid-doped PVA layer indicated that the root mean square value was 0.546 nm and morphology of layers was relatively homogenous and smooth. The surface morphology of the PVA/PDVT-10 layer was further characterized by AFM as demonstrated in Figure S2b. The root mean square value was 1.21 nm and large crystal grain size was obtained, indicating the good quality of semiconductor layer.

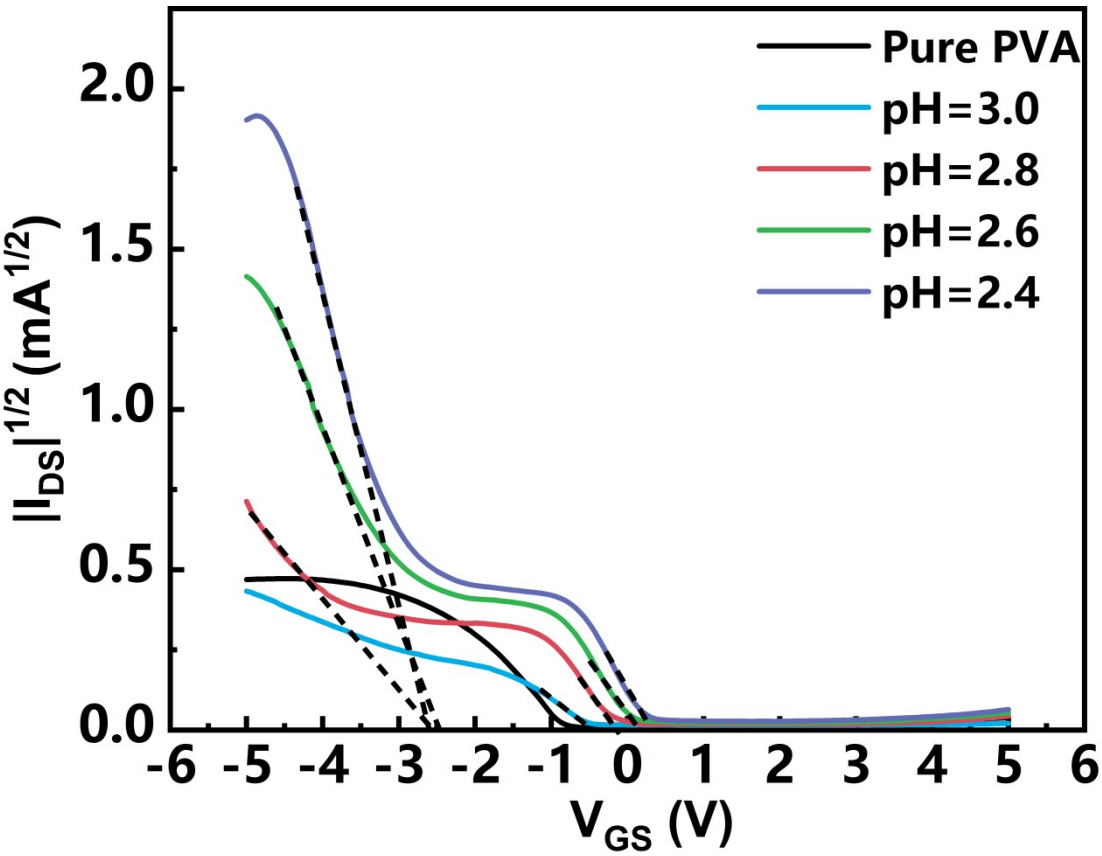

Figure S3 Transfer curves ($I_{DS}^{1/2}$ vs V_{GS}) of the devices with variant pH values

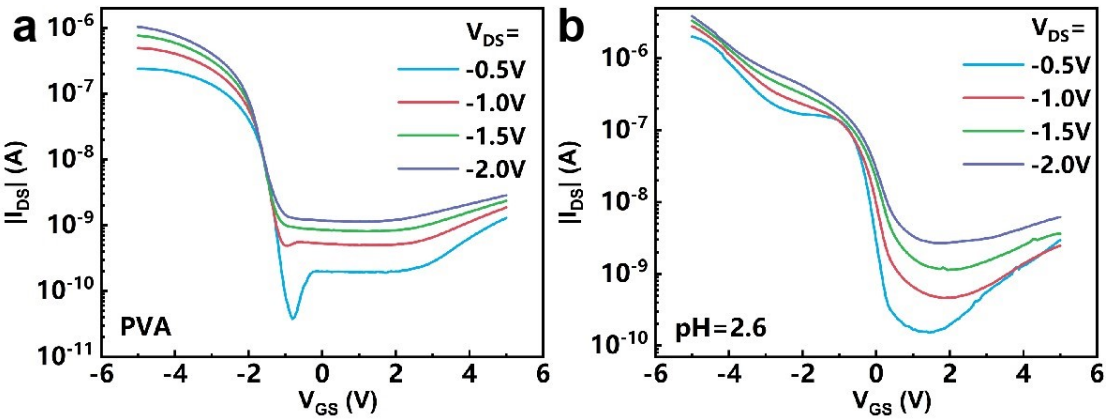

Figure S4 Transfer curves of a) a pure PVA device and b) pH=2.6 device at variant drain voltages.

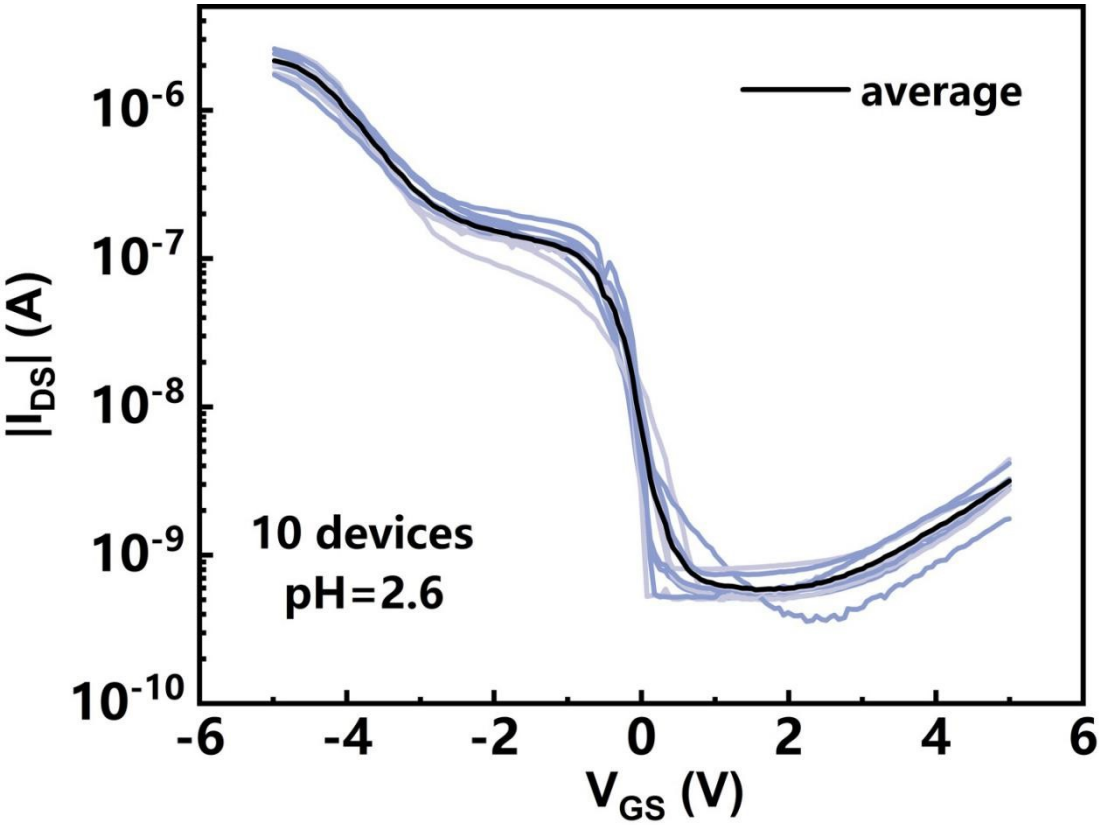

Figure S5 Transfer curves of 10 different devices with a pH value of 2.6.

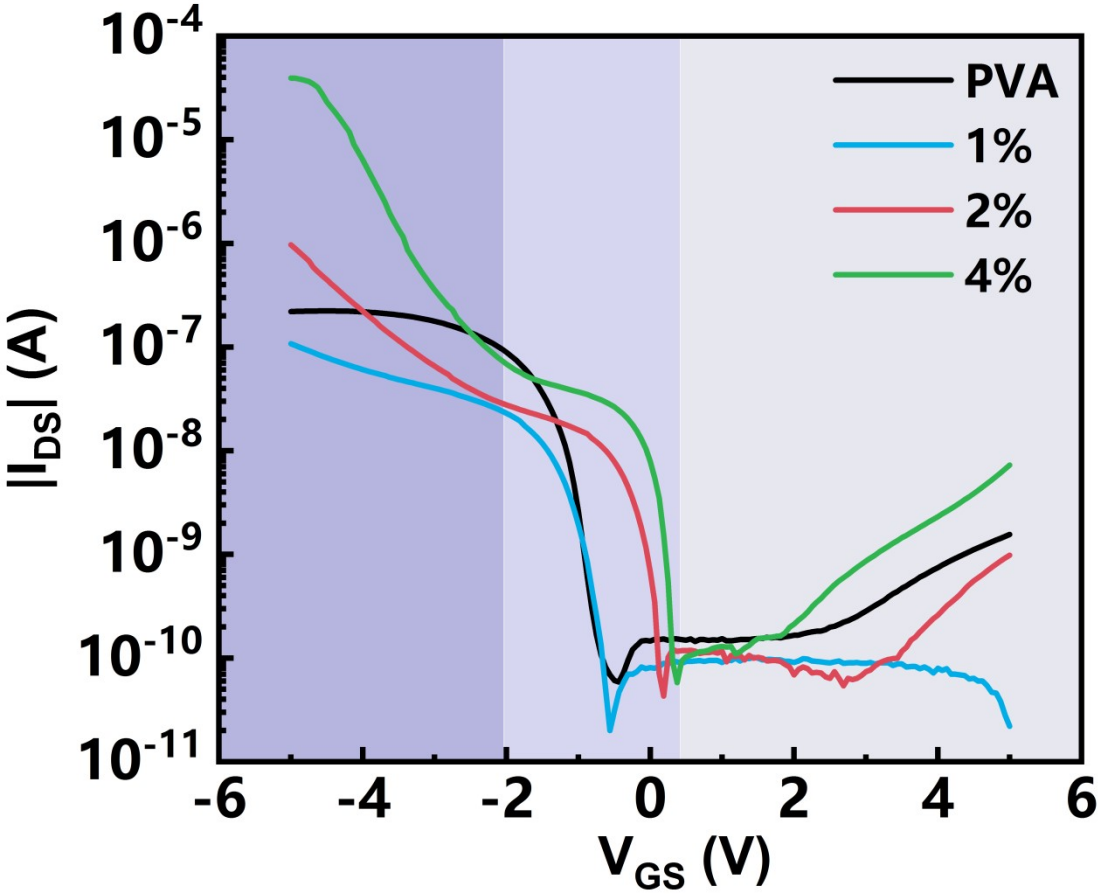

Figure S6 Transfer curves of Li salt doped PVA with variant dopant ratios.

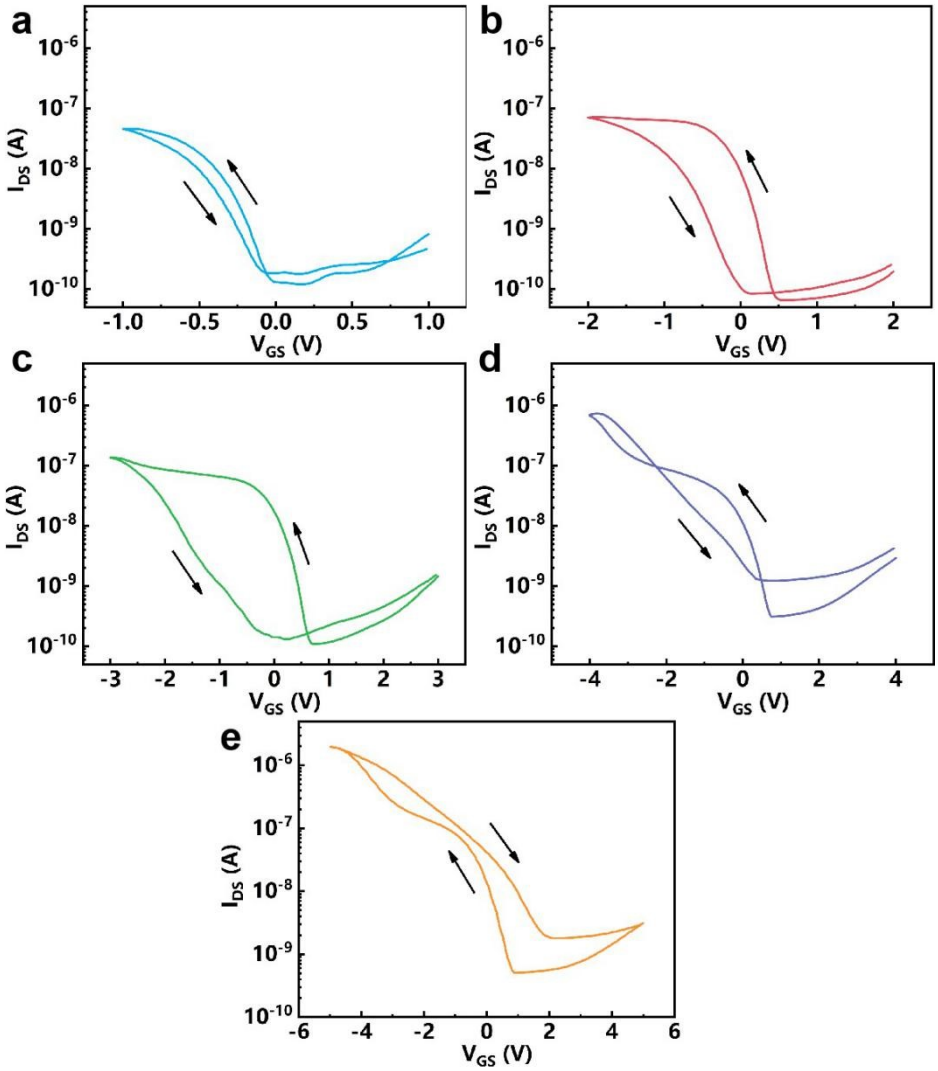

Figure S7 Dual-swept transfer characteristics curves of the device at the range of a) [-1V, 1V], b) [-2V, 2V], c) [-3V, 3V], d) [-4V, 4V] and e) [-5V, 5V].

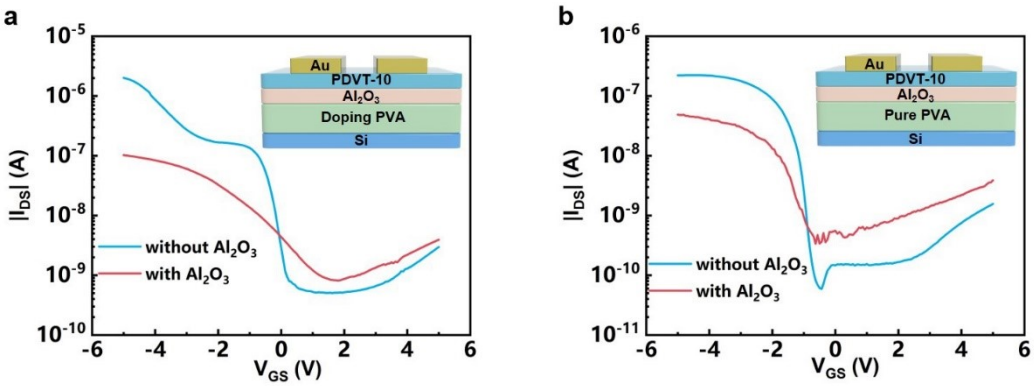

Figure S8 Transfer curves of a) pH=2.6 and b) PVA device with/without a Al_2O_3 isolated layer.

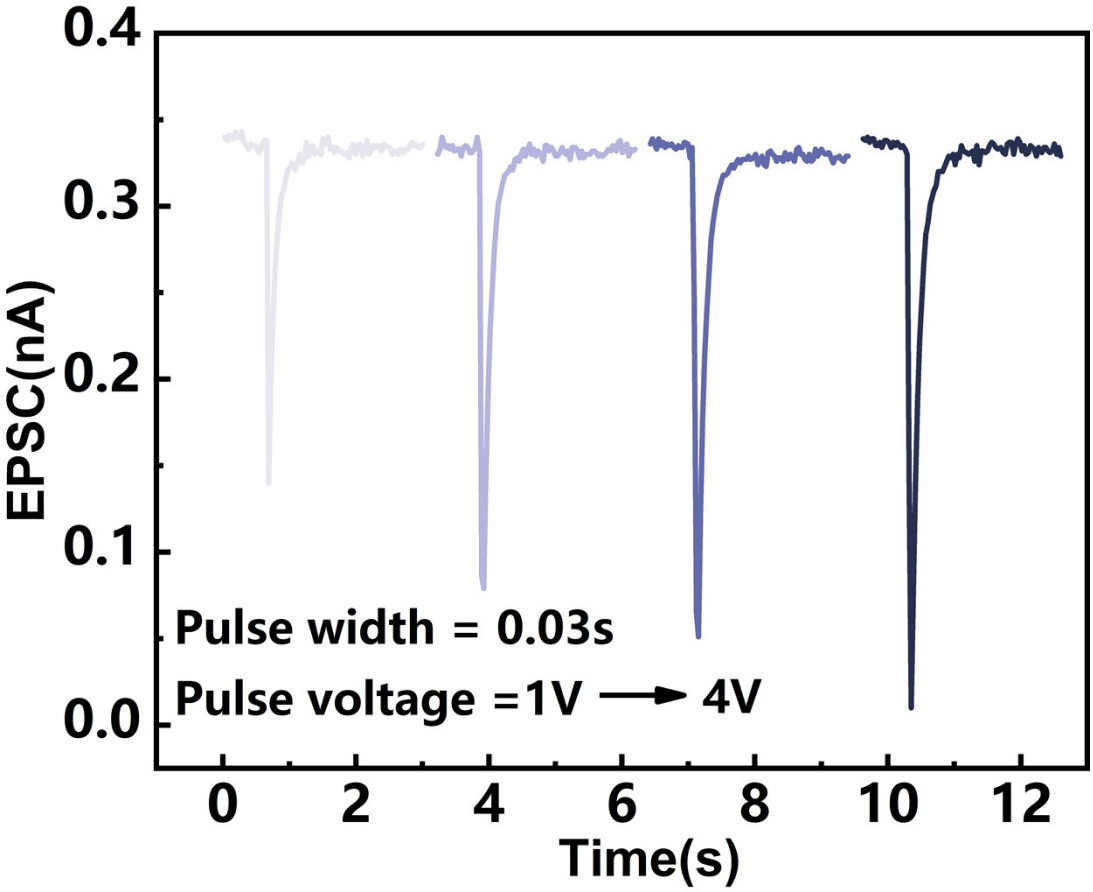

Figure S9 Response of EPSC triggered by pulses with variable positive voltages (from 1 V to 4V).

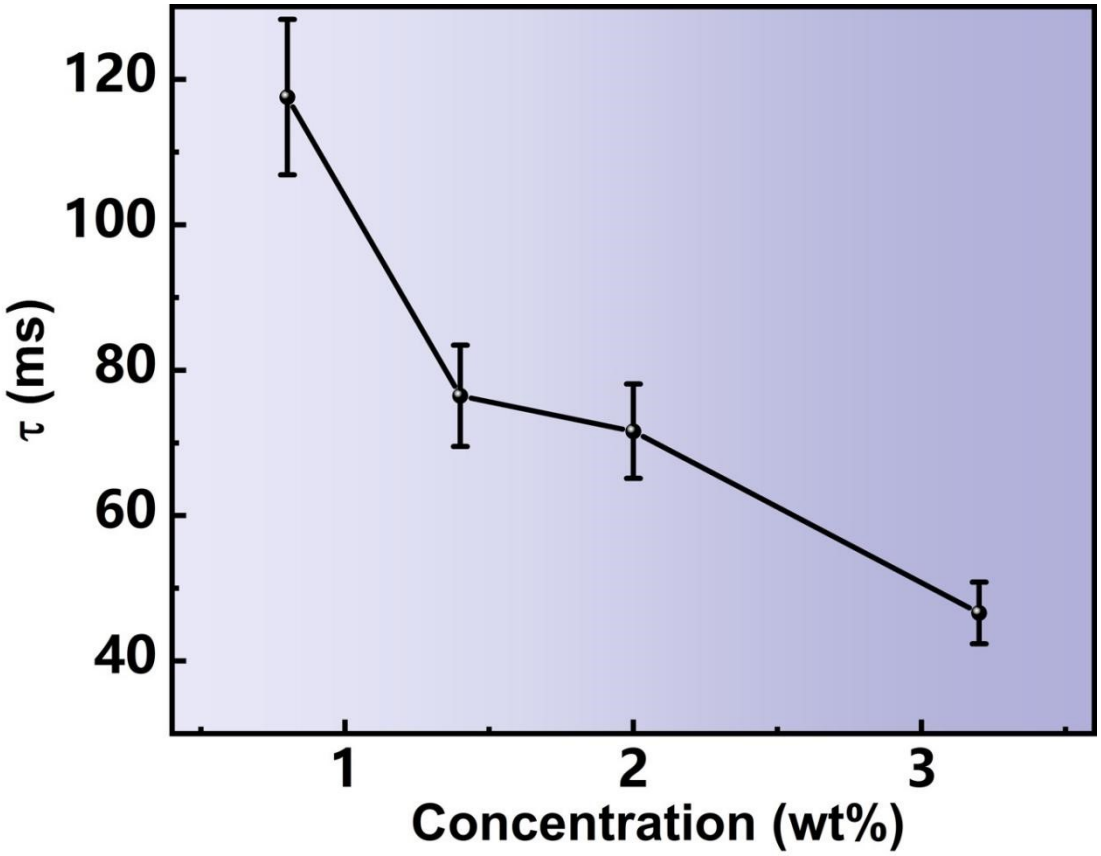

Figure S10 The decay constant (τ) as a function of acid concentration in the PPF fitting curves.

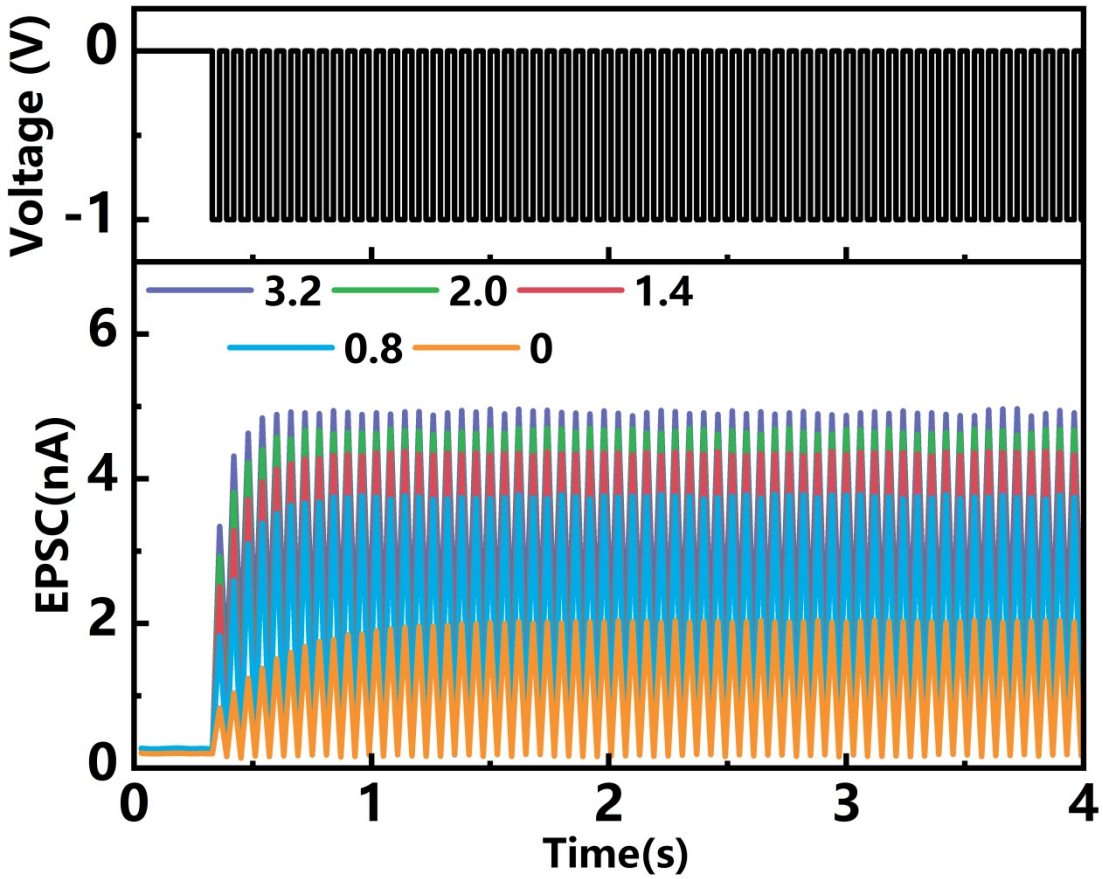

Figure S11 Response of the device triggered by a series of pulses with variant dopant ratios**.**

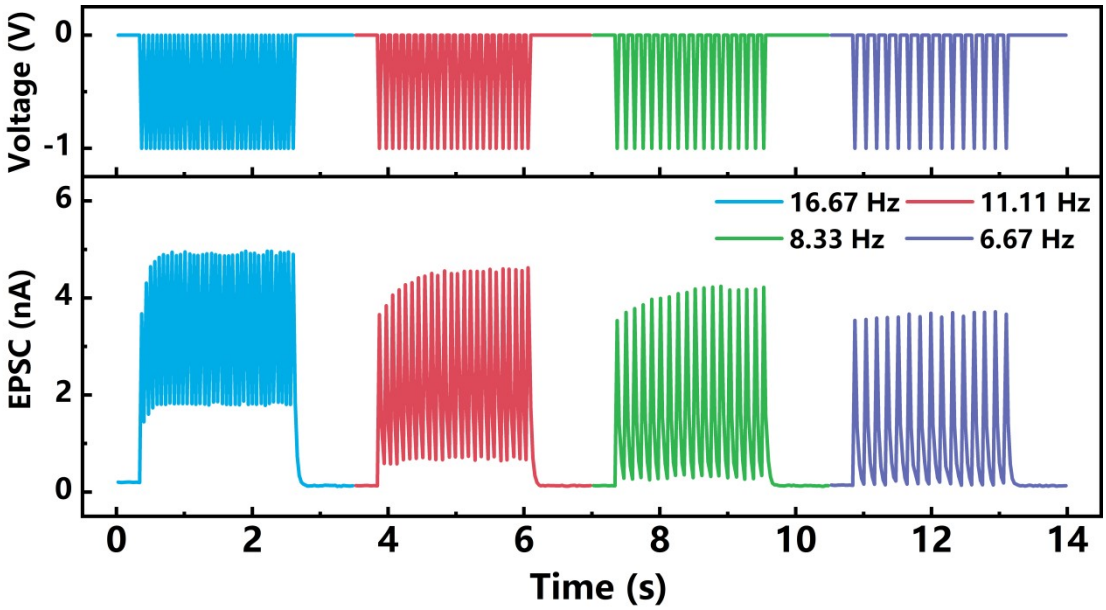

Figure S12 Response of the device triggered by pulses with different frequencies.

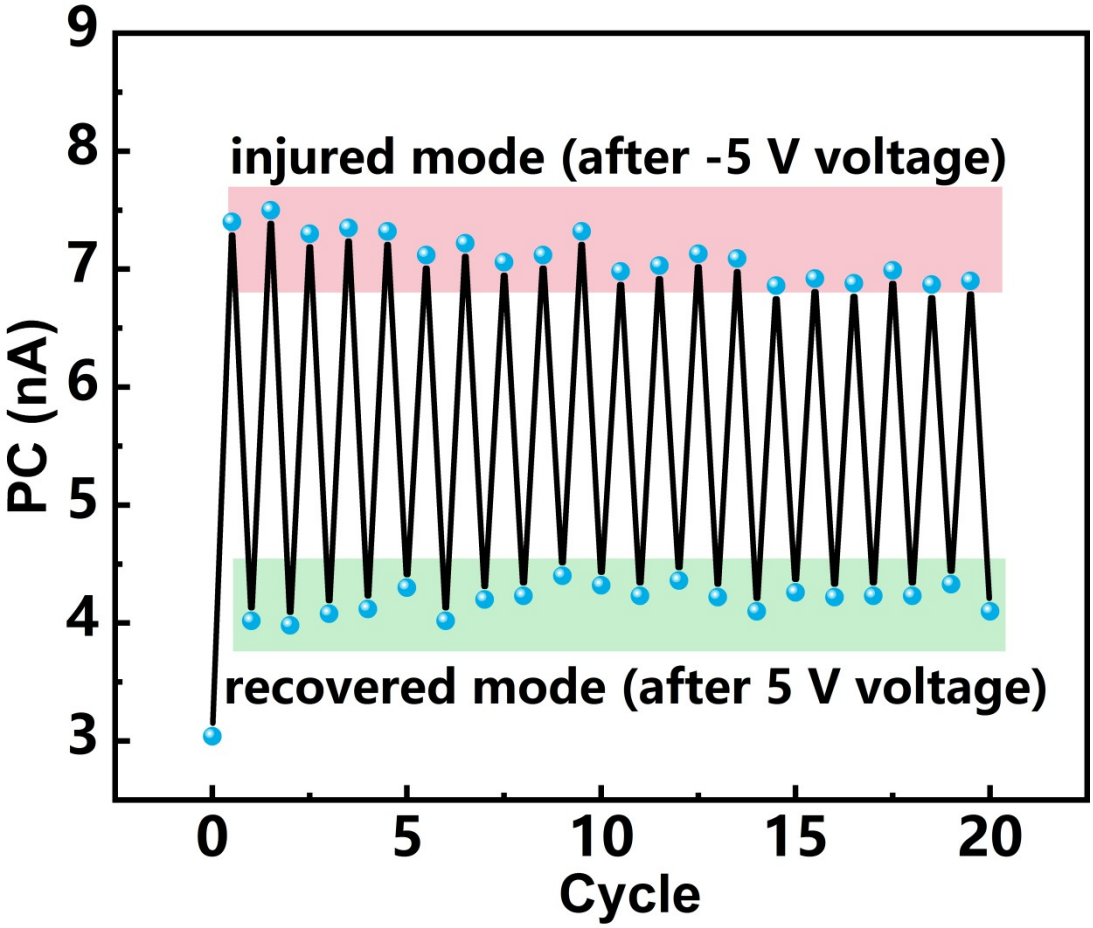

Figure S13 The switching behavior of the device

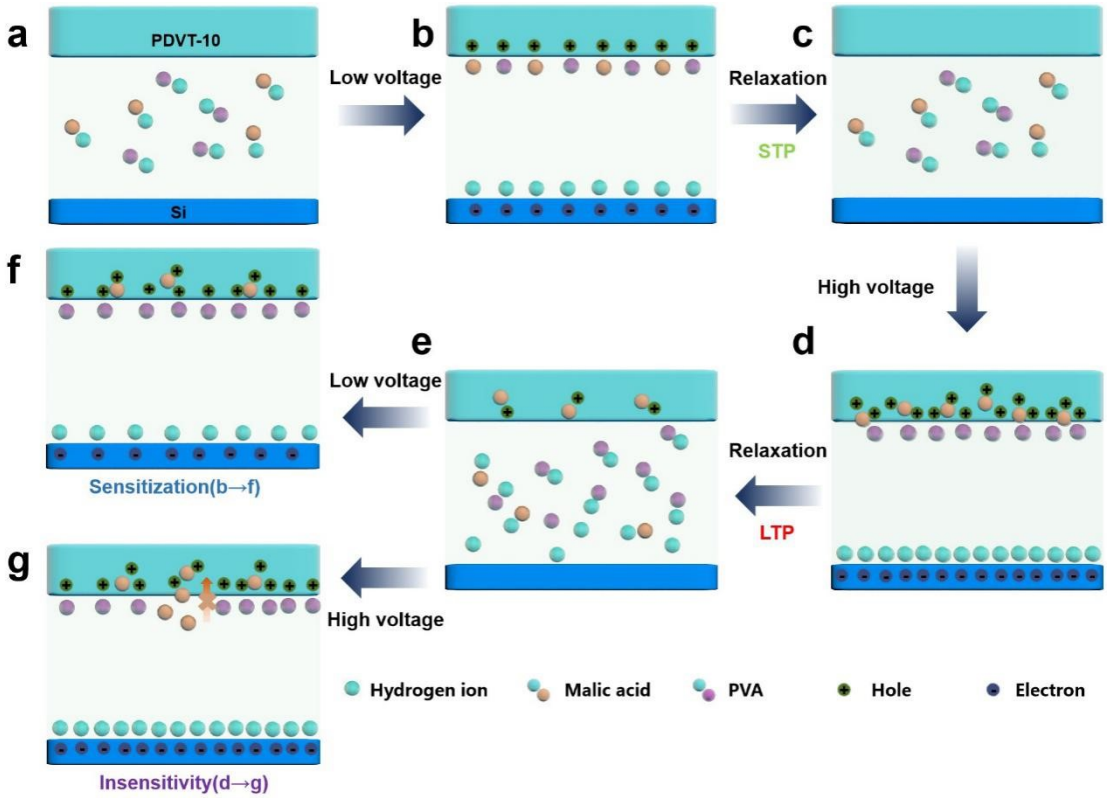

Figure S14 The mechanism of the moving ions during the process of applying different voltages.

In general, the switching speed of electrical double layer regime is milliseconds while electrochemical transistors are limited by the migration rate of ions between the electrolyte and semiconductor layer, exhibiting slow switching speed (more than seconds).[1] Hence, the free ions were polarized in the EDL when a pulse with low voltage was applied to the nociceptor while the polarized ions would decay for dozens of milliseconds, and the current return to initial state rapidly, which was termed as “fast pain” in Process b and c. When a high voltage was applied to the nociceptor, acid radicals migrate toward the semiconductor layer and tit is hard for them to return to the EDL. As a result, the current would decay to a relative high current, which was termed as “chronic pain” in Process d and e. After that, a low and high voltage was applied to the nociceptor again as the Process f and g showed,

respectively. Under the synergy of electrical double layer and electrochemical doping effect, more holes would be induced in the semiconductor, thereby causing a higher response in the nociceptor which was termed as “sensitization”. As a contrast, the ions in semiconductor would prevent ions from migrating toward the semiconductor layer again, and so there would be less ions existing in the semiconductor. Therefore, the current would be lower than that in the Process d so that “insensitivity” was termed.

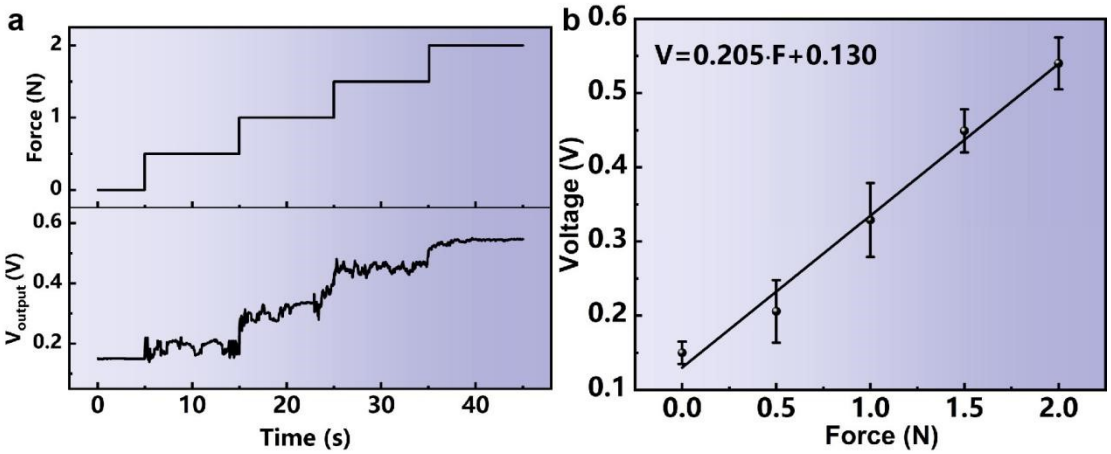

Figure S15 a) Variety of output voltage as the force increased, and b) the corresponding fitting curve of voltage as a function of force.

	Stage Ⅰ	Stage Ⅱ	Stage Ⅲ
Force (N)	5	~15	5
Stimuli from piezoresistor (V)	1.15	3.3	1.15
Above threshold	No	Yes	No
Nociceptor mode	No-injured	Injured	Injured
Action	No	Yes	Yes
EPSC			

Table S1 Operating parameters in the process of pain reflex movement

Reference

[1] S. H. Kim, K. Hong, W. Xie, K. H. Lee, S. Zhang, T. P. Lodge, C. D. Frisbie, *Adv. Mater.* **2013**, 25, 1822.