ARTICLE

Electronic supplementary information

Affective computing for human-machine interaction via bionic organic memristor exhibiting selective in-situ activation

Bingjie Guo,†[a] Xiaolong Zhong,†[b] Zhe Yu,*[c] Zhilong He,[a] Shuzhi Liu,[a] Zhixin Wu,[b] Sixian Liu,[b] Yanbo Guo,[b] Weilin Chen,[b] Hongxiao Duan,[b] Jianmin Zeng,[b]Pingqi Gao,[c] Bin Zhang,[d] Qian Chen,[de] Heidong He,[e] Yu Chen*[d] and Gang Liu*[ab]

a. School of Chemistry and Chemical Engineering, Shanghai Jiao Tong University, Shanghai 200240, China.
b. Department of Micro/Nano Electronics, School of Electronic Information and Electrical Engineering, Shanghai Jiao Tong University, Shanghai 200240, China.
c. School of Materials, Sun Yat-Sen University, Guangzhou, Guangdong, 510275, China.
d. School of Chemistru and Molecular Engineering, East China University of Science and Technology, Shanghai 200237, China
e. Minhang Hospital, Fudan University, Shanghai 201199, China

† These authors contribute equally to this work.

Keywords: affective computing, human-machine interaction, bionic organic memristor, ligand-gated ion channel, selective in-situ activation

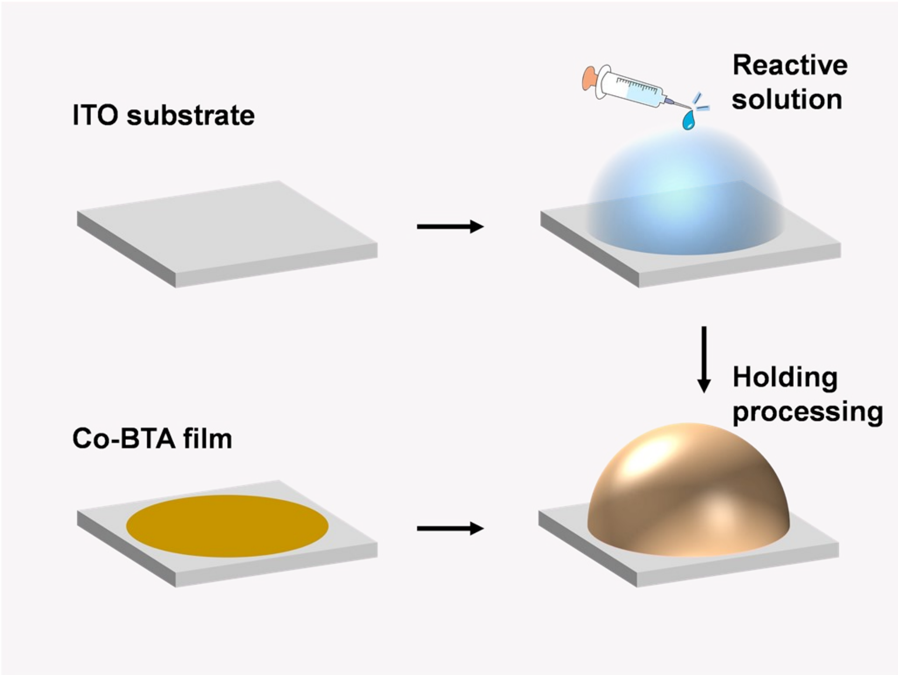

Figure S1. Schematic diagram of preparing Co-BTA film.

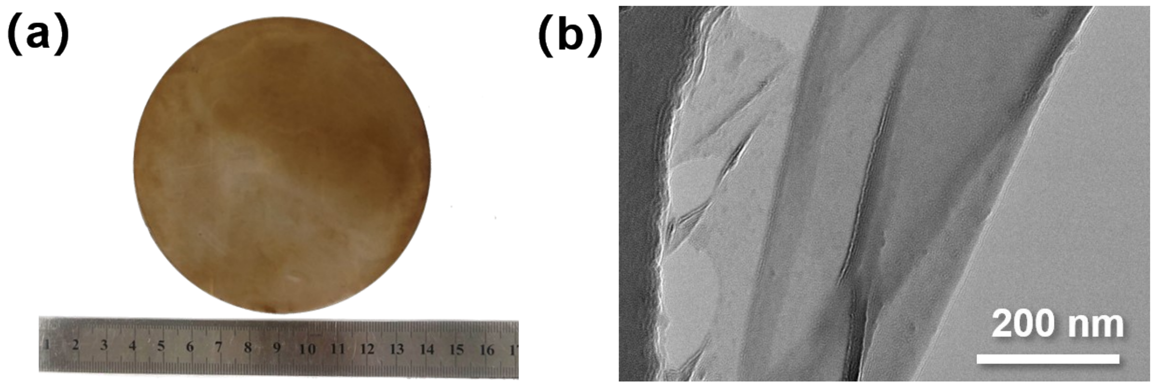

Figure S2. (**a**) The photo of large-are continuous Co-BTA film. It can be up to 4 inches in diameter. (**b**) Microtopography of the Co-BTA film. By directly depositing the film onto a copper mesh and observing its behavior, it becomes apparent that the film exhibits both continuity and curling in specific regions. This observation serves as evidence that the molecules self-assemble into continuous and flexible films under ammonium conditions. The presence of a continuous film indicates that the molecules have arranged themselves in an organized manner, forming a cohesive and interconnected structure.

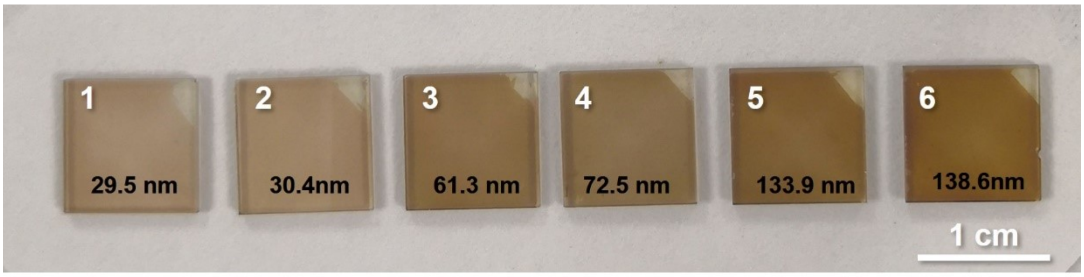

Figure S3. Photo of synthetic Co-BTA films on ITO glass. They were synthesized by different number of repeated growths (from 1 to 6 times), and the area of ITO glass is 1 cm^2. The upper right corner of each glass is the original transparent appearance. Obviously, the substrates were darkened with increasing cycles of film process, which show that the film thickness could be precise control with the process cycle numbers.

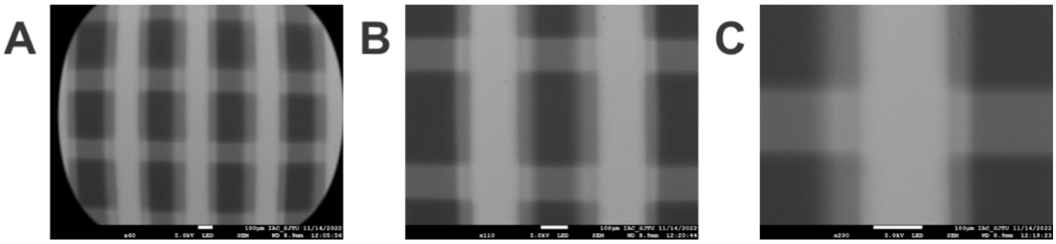

Figure S4. (a-c) Microstructure of fabricated bionic organic memristor network under the magnification of 60, 110, and 200.

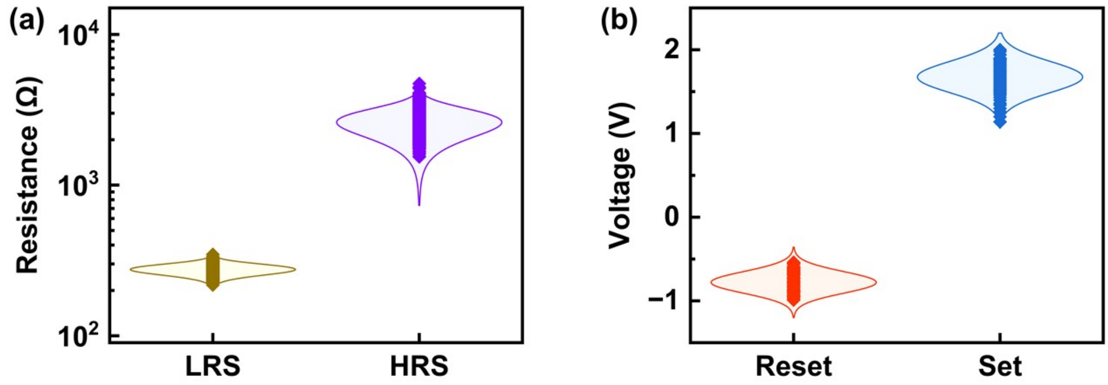

Figure S5. Performance statistics of bionic organic memristor based on Co-BTA in 600 I-V cycles, including LRS/HRS value distribution (a), and switching voltage distribution (b).

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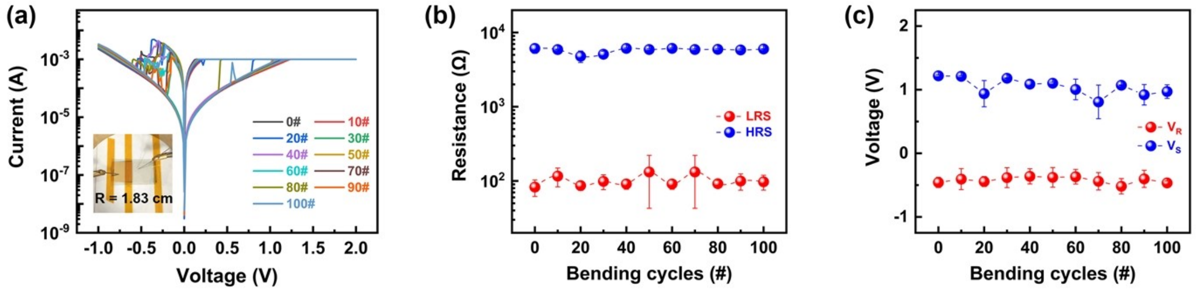

Figure S6. (a) The current-voltage characteristics cycles of memristor while bending for 100 times. The inset is the photograph of bending test. The changes in HRS/LRS values (b) and switching voltages (c) with the number of bending cycles.

Flexible Au/Co-BTA/ITO/PET Device Fabrication

The electrical performance of flexible Co-BTA devices were performed by the configuration of Au/Co-BTA/ITO/Polyethylene terephthalate (PET) in a ground electrode structure with a top Au electrode radius of 150 µm. Firstly, after ultrasonication in deionized water, ethanol, and acetone for 30 min, the ITO/PET substrate was dried using a Nitrogen gun. The Co-BTA film, serving as the active material, was synthesized via the liquid-solid interface method. Following this, a patterned mask with roundness shape (150 µm radius) was used to mask the Co-BTA/ITO/PET substrate. Finally, the top electrode Au was deposited onto the Co-BTA film through magnetron sputtering at 1.3 Pa for 30 min by using the mask. This process resulted in the formation of the Au/Co-BTA/ITO/PET in a ground electrode structure.

Characterization of Flexible Au/Co-BTA/ITO/PET Devices

All the mechanical deformations are introduced by convexly bending the sample device using a series of home-made bending radius of 1.83 cm as shown in inner picture of Figure S6a. All electrical measurements were conducted using a Keithley 4200 semiconductor parameter analyzer.

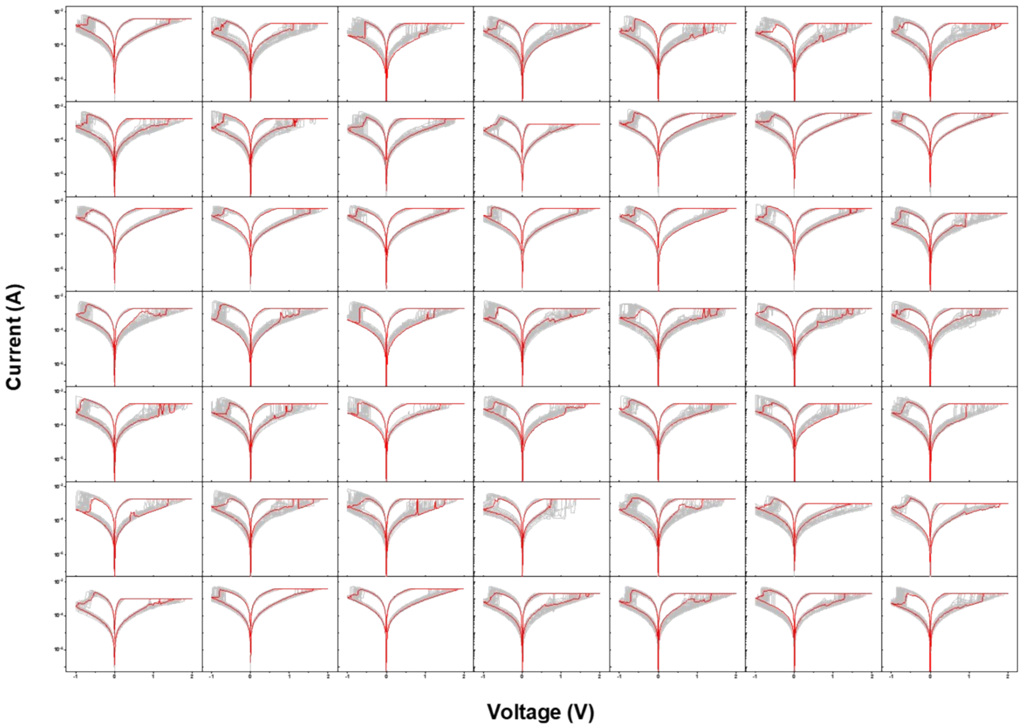

Figure S7. Characteristic I-V curves of 49 devices in the bionic organic memristor network. All the devices were subjected to voltage sweeping from 0V to 2V and from 0V to -1V, with 50 I-V cycles.

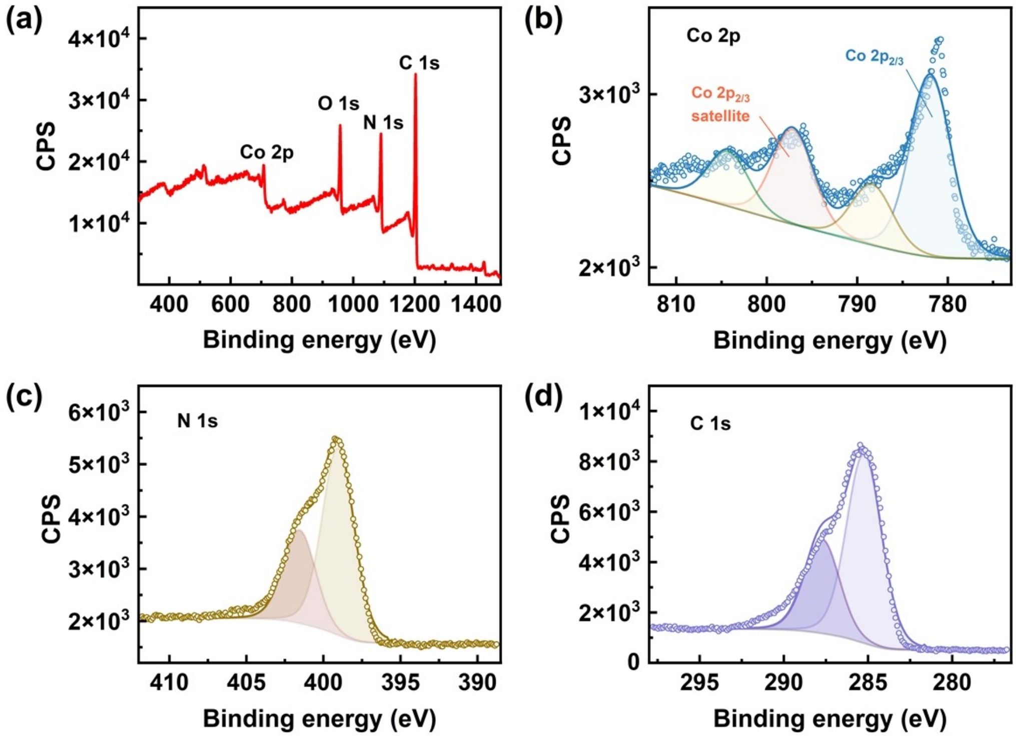

Figure S8. (a) XPS survey spectra of Co-BTA film. The Co-BTA film mainly consists of Co, N, and C elements. XPS high resolution spectra of (b) Co 2p region, (c) N 1s region, and (d) C 1s region of Co-BTA film. Circles represent the experimental data, the solid lines are the envelope of fitting, and the dash lines are backgrounds and fitting components. Co-BTA film were prepared on glass substrate with full coverage.

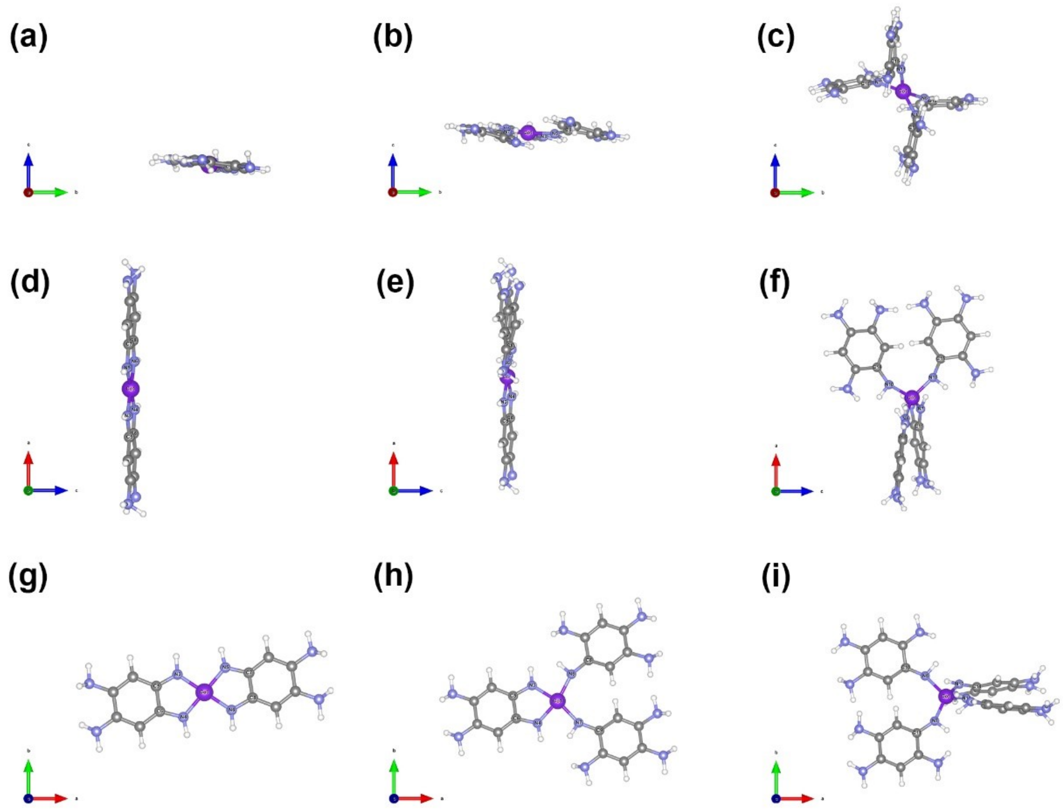

Figure S9. Simulated chemical structures of Z=2 (a, d, g), Z=3 (b, e, h), and Z=4 (c, f, i). Perspectives are labeled as axes a (a, b, c), b (d, e, f), and c (g, h, i), respectively. The increase in the number of coordinated BTA molecules introduces a strong steric hindrance effect, which affects the degree of spatial twisting in the Co-BTA molecular structure.

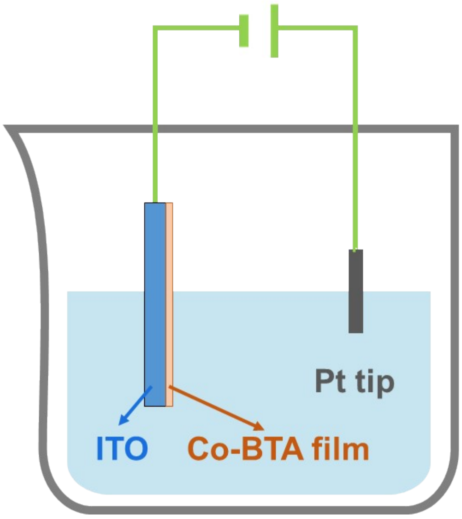

Figure S11. Illustration of electrochemical Pt-tip/Co-BTA/ITO device. An electrochemical Pt-tip/Co-BTA/ITO device was designed and applied in mechanism experiments to study the redox state of the Co-BTA film. The device consisted of a power source, Pt tip, and ITO-coated Co-BTA film. A continuous voltage sweep was applied to the Co-BTA film via the power source. Then, the Co-BTA/ITO glass was taken out and dried by blowing, which was used for the next test. The simulated oxidation and reduced states of the Co-BTA film during resistive switching were acquired by applying write voltages (1, 2, and 3 V) and erase voltages (-1, -2 V) for one minute. To relate the transport phenomena to the electronic structure of the active materials, in-situ Raman spectrum and ultraviolet-visible (UV-vis) spectra were performed with the Pt-tip/Co-BTA/ITO devices.

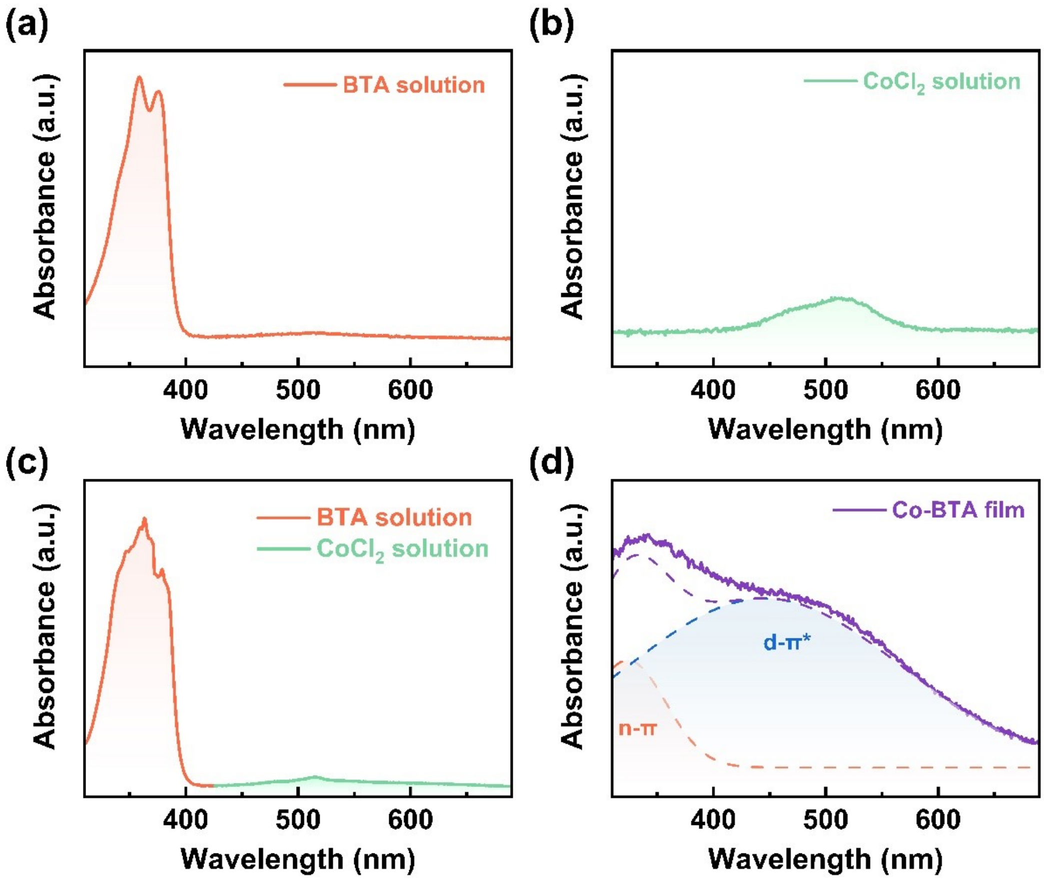

Figure S12. UV-Vis spectrum of (a) BTA solution, (b) $CoCl_2$ solution, (c) BTA and $CoCl_2$ blending solution, and (d) Co-BTA film. A blank glass baseline and zero-background correction were collected prior to the sample measurements. The UV-Vis spectrum was obtained for the sample film on a high-conductance ITO glass substrate with a scan rate of 2 nm/s under ambient conditions. Peaks at 360 nm and 510 nm correspond to BTA·4HCl and $CoCl_2·6H_2O$, respectively. When Co cations coordinated with BTA, the peaks at 510 nm redshift to 465 nm, indicating the influence of d-π* metal-to-ligand charge transfer.

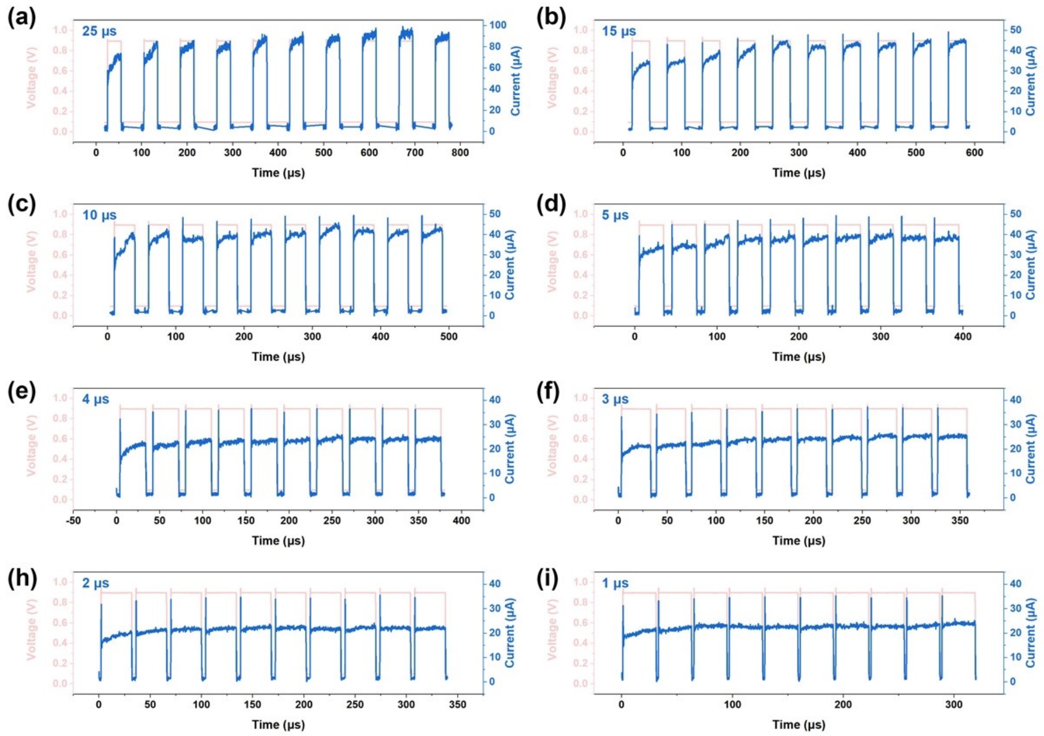

Figure S13. The current change induced by a continuous 10 electrical voltage pulse stimulation with the fixed amplitude value (0.9 V) and reading bias (0.1 V). The pulses width is 10 µs with different intervals of (a) 25 µs, (b) 15 µs, (c) 10 µs, (d) 5 µs, (e) 4 µs, (f) 3 µs, (g) 2 µs, and (h) 1 µs.

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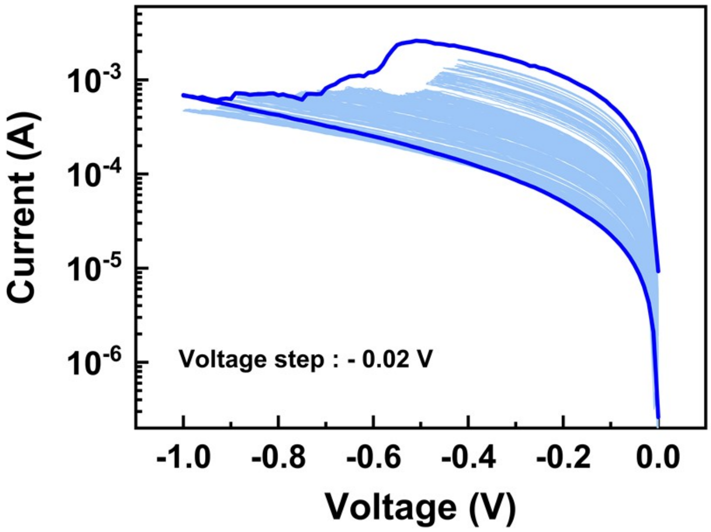

Figure S14. The continuous conductance states modulation by applying reset voltage from -0.4 V to -1 V with step of -0.02 V.

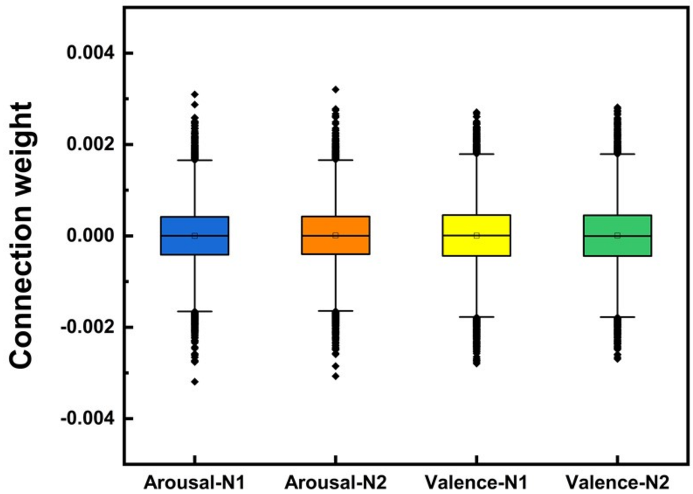

Figure S15. Box plots of the statistical analysis of the connection weights of the output layer neurons.

Table S1. Summary of dihedral angle of three molecule

Molecule	Atom				Dihedral angle (degree)
Z = 2	C5	N3	Co1	N5	179.31
	C7	N5	Co1	N6	0.72
	C8	N6	Co1	N4	179.09
	C6	N4	Co1	N3	0.39
Z = 3	C5	N3	Co1	N6	171.54
	C8	N6	Co1	N7	21.55
	C9	N7	Co1	N4	169.04
	C6	N4	Co1	N3	0.27
Z = 4	C10	N6	Co1	N10	143.95
	C14	N10	Co1	N11	52.07
	C15	N11	Co1	N7	91.12
	C11	N7	Co1	N6	24.83

Table S2. Summary of Co-N bond length of three molecule

Molecule	Bond	Bond length (Å)
Z = 2	N3-Co1	1.82
	N5-Co1	1.82
	N6-Co1	1.83
	N4-Co1	1.83
Z = 3	N3- Co1	1.86
	N6- Co1	1.87
	N7- Co1	1.87
	N4- Co1	1.87
Z = 4	N6- Co1	1.93
	N10- Co1	1.89
	N11- Co1	1.89
	N7- Co1	1.82