

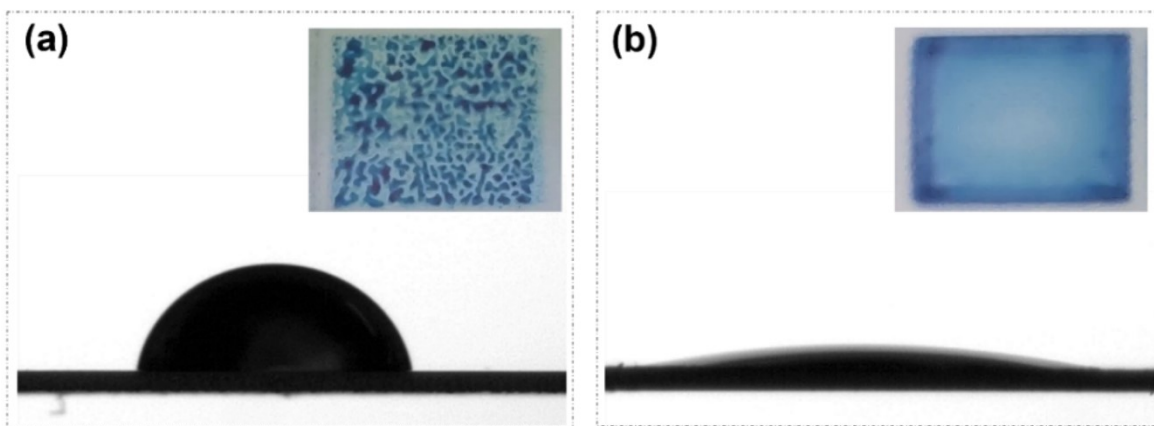
## Supporting Information

### **Metallosupramolecular polymer deposited via inkjet printing for fast-switching pixelated electrochromic devices**

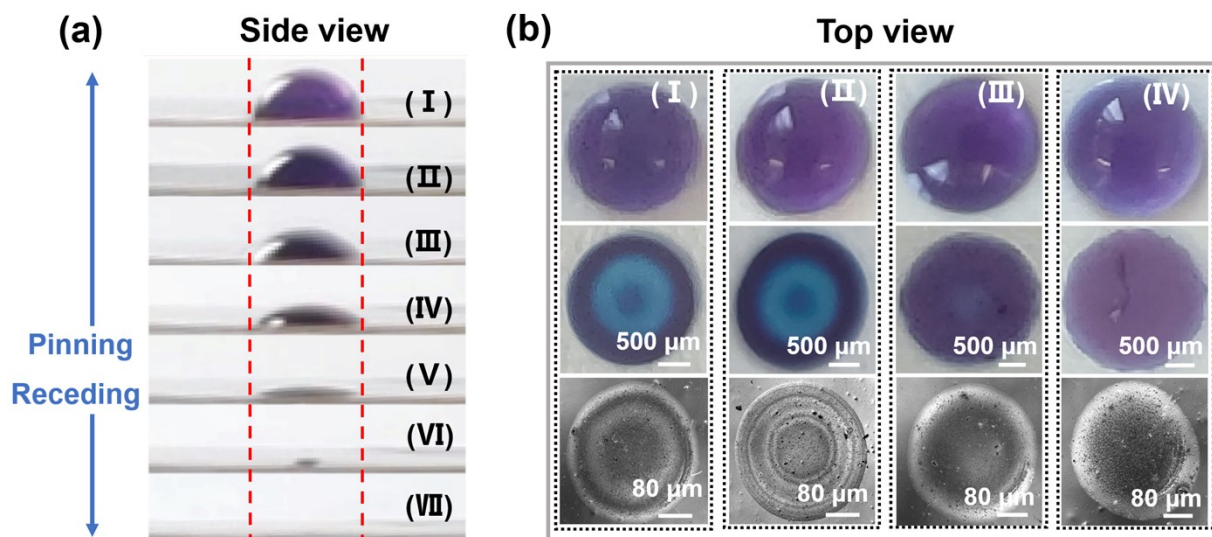
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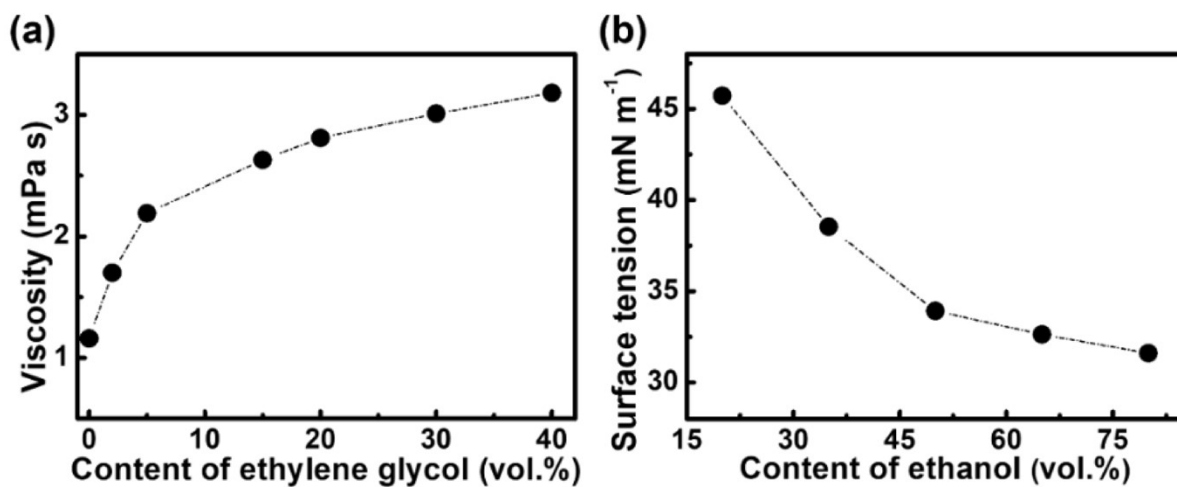
\*Corresponding author: gaoguo@hit.edu.cn; jiangzaixing@hit.edu.cn.



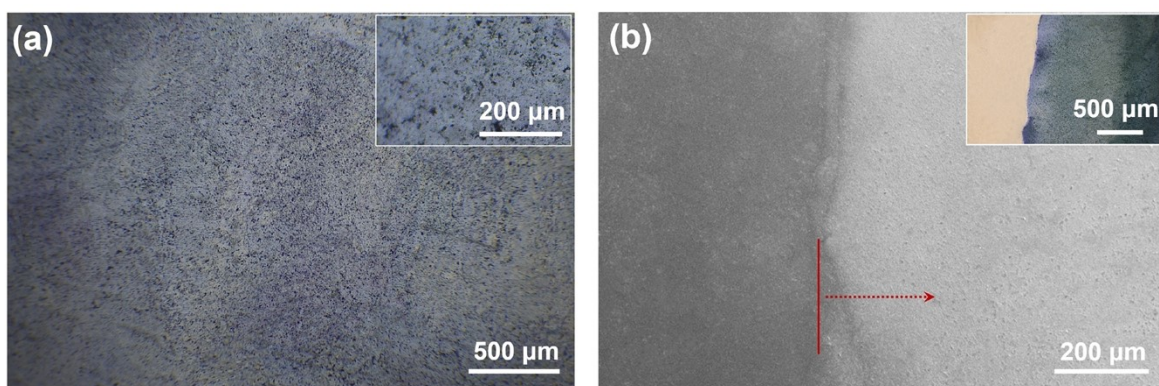
**Figure S1.** Contact angel of Fe(II)-MEPE polymer inks with (a) water (b) ethanol as solvent on ITO-coated PET substrate (the insets were the typical photograph of the printed film).



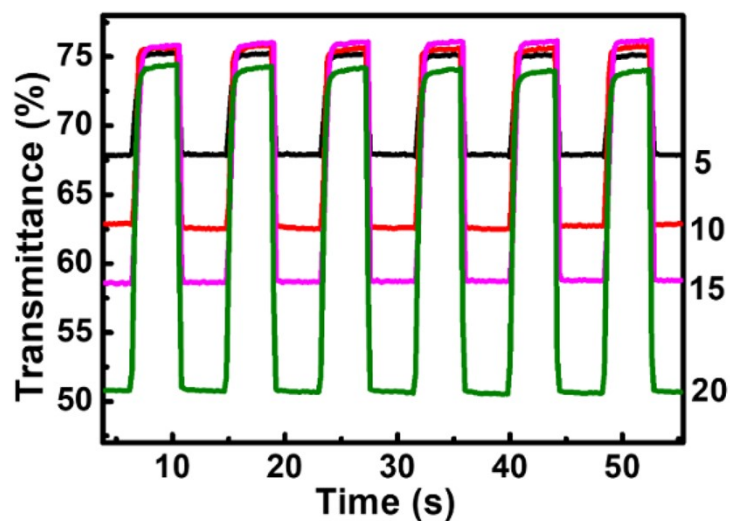
**Figure S2.** (a) The side view images of Fe(II)-MEPE polymer droplet with water as solvent during drying process on the ITO-coated PET substrate. (b) The top view images of 2  $\mu\text{L}$  Fe(II)-MEPE polymer droplets with water, ethanol, ethylene glycol, and water/ethanol/ethylene glycol (volume ratio of 100:50:10) as solvent from I to IV (top) and the top view images of dried Fe(II)-MEPE droplets (middle). The top view images of sub-microliter Fe(II)-MEPE droplets deposited (bottom).



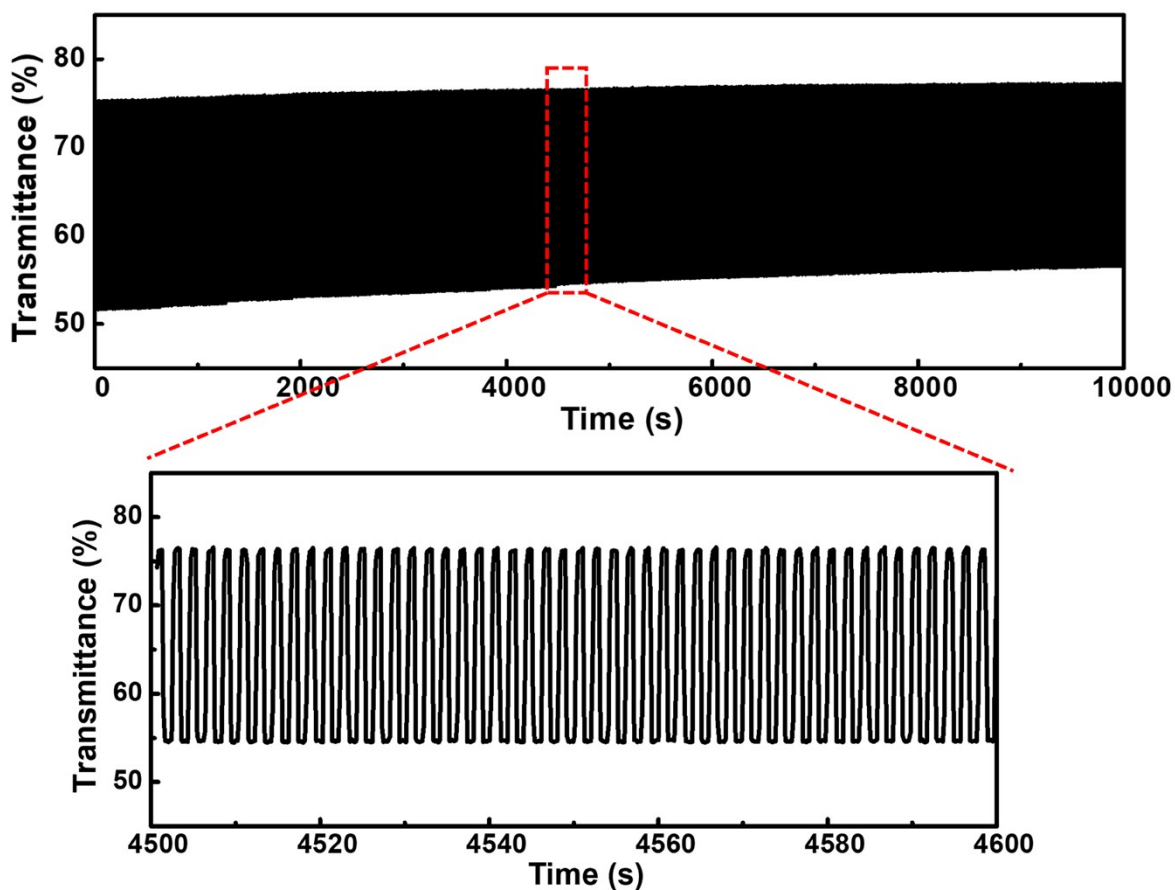
**Figure S3.** (a) The viscosity of Fe(II)-MEPE inks (with a fixed concentrate of 1 mg mL<sup>-1</sup>) with different content ethylene glycol relative to water. (b) The surface tension of Fe(II)-MEPE inks with different content ethanol relative to volent of water.



**Figure S4.** (a) A microscope image of the surface morphology of the inkjet-printed Fe(II)-MEPE film at a magnification of 4 × 16 (inset presents an image at a magnification of 10 × 16); (b) The SEM image of the boundary part of the inkjet-printed Fe(II)-MEPE polymer film (the inset was the optical microscope image).



**Figure S5.** Electrochromic switching behavior of electrochromic films with different printed layers of Fe(II)-MEPE polymer monitored at 584 nm at a voltage of 0 V for 4 s and +1.4 V for 4 s for cycles.



**Figure S6.** Transient transmittance profile at 584 nm during continuous coloration/bleaching switching 5000 cycles of the printed Fe(II)-MEPE polymer film.

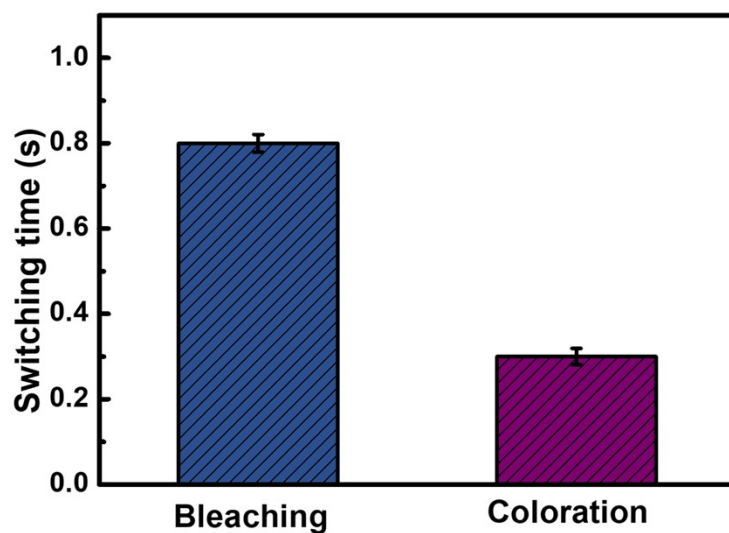


Figure S7. The Fe(II)-MEPE film switching speed histogram

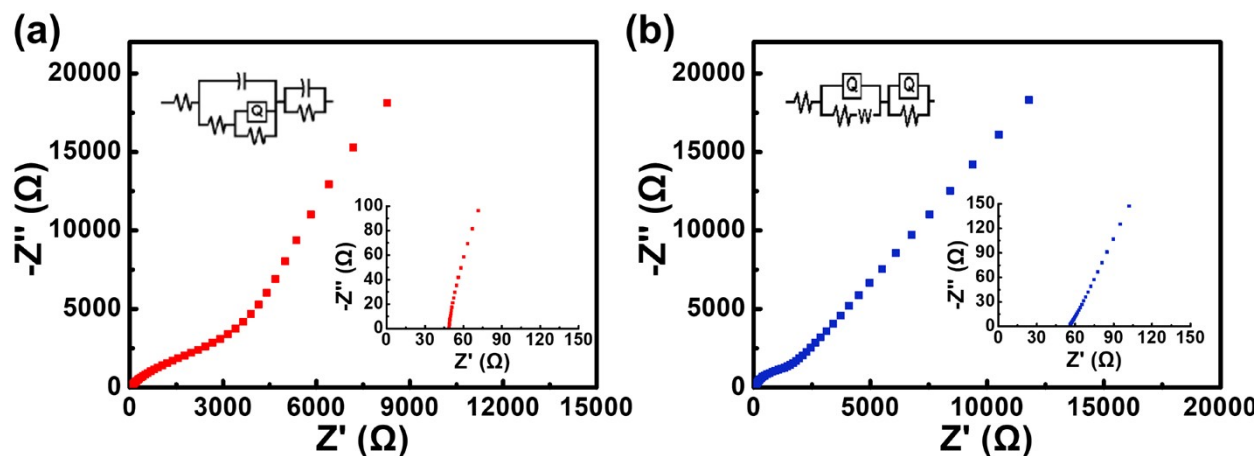
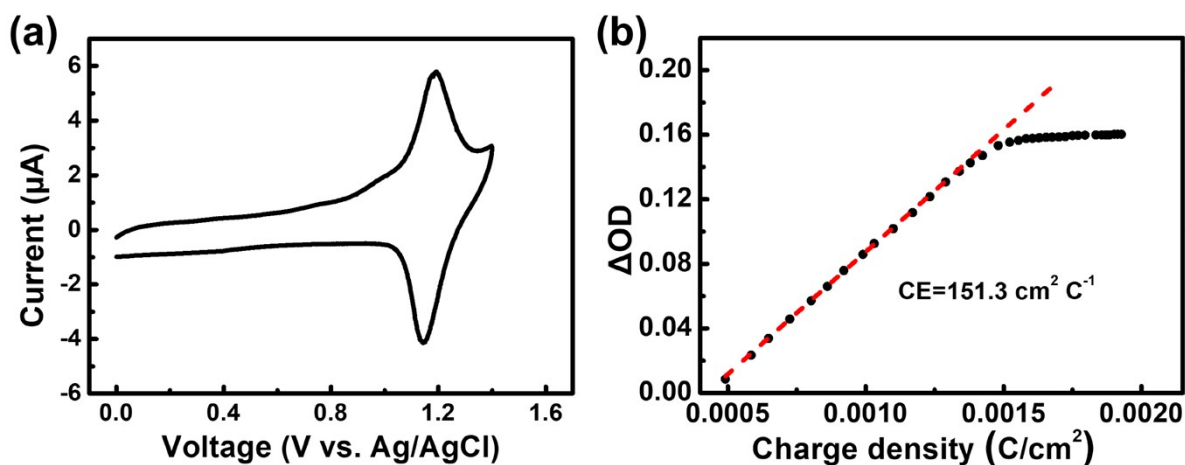
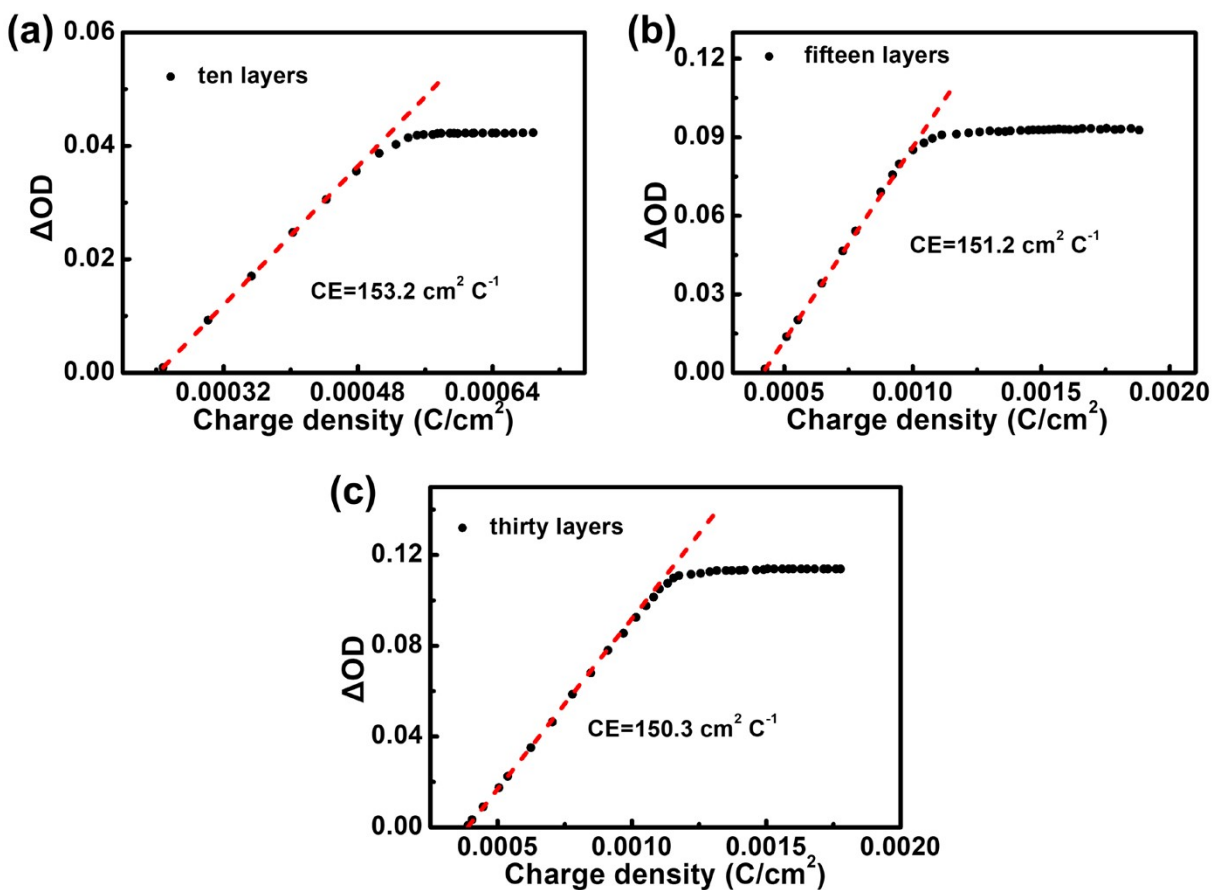


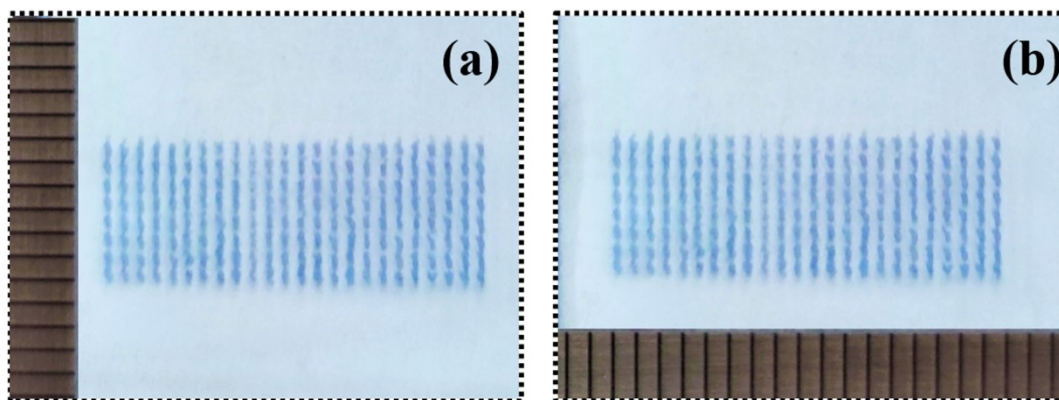
Figure S8. The AC impedance spectra of the ITO-coated PET electrode and the inkjet-printed Fe(II)-MEPE film in the presence of 0.1 M TBAClO<sub>4</sub>/ACN.



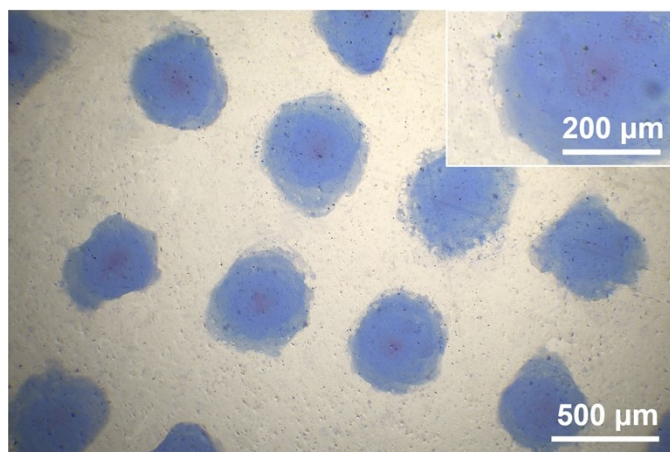
**Figure S9.** (a) CV (scan rate: 20 mV/s) and (b) plot of the optical density ( $\Delta OD$ ) versus the injected charge density for the printed Fe(II)-MEPE film. The coloration efficiency (CE) was extracted from the slope of the linear fitting in the linear scheme of the plot.



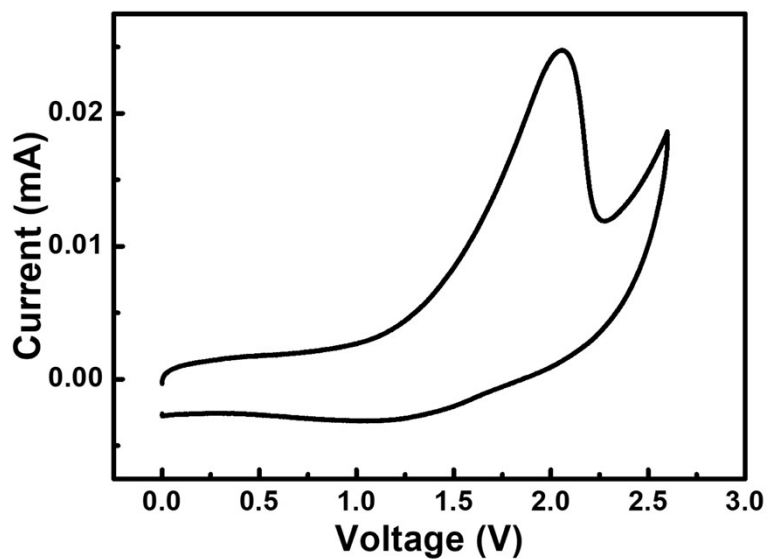
**Figure S10.** The coloration efficiency of printed Fe(II)-MEPE films in different layers



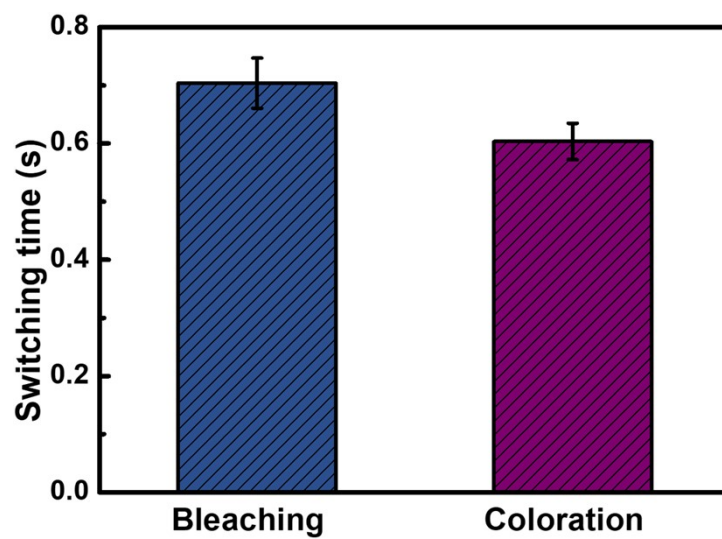
**Figure S11.** The pixelated Fe(II)-MEPE film with  $8 \times 24$  arrays composed of dot pixels ( $\sim 350 \mu\text{m} \times 700 \mu\text{m}$ ) on ITO-coated PET surface (the minimum scale of the ruler is 1.0 mm).



**Figure S12.** A microscope image of the surface appearance of the inkjet-printed pixels on an ITO-coated PET film using the Fe(II)-MEPE polymer in ternary solvent at a magnification of  $4 \times 16$  (inset presents an image at a magnification of  $10 \times 16$ ).

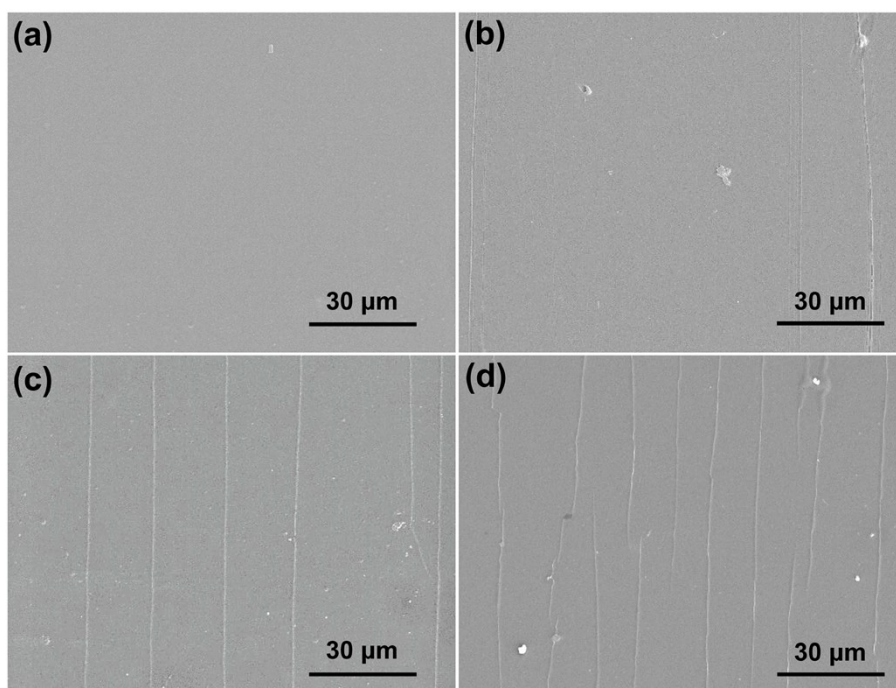


**Figure S13.** The cyclic voltammetry curve (scan rate: 20 mV/s) of the PECD.

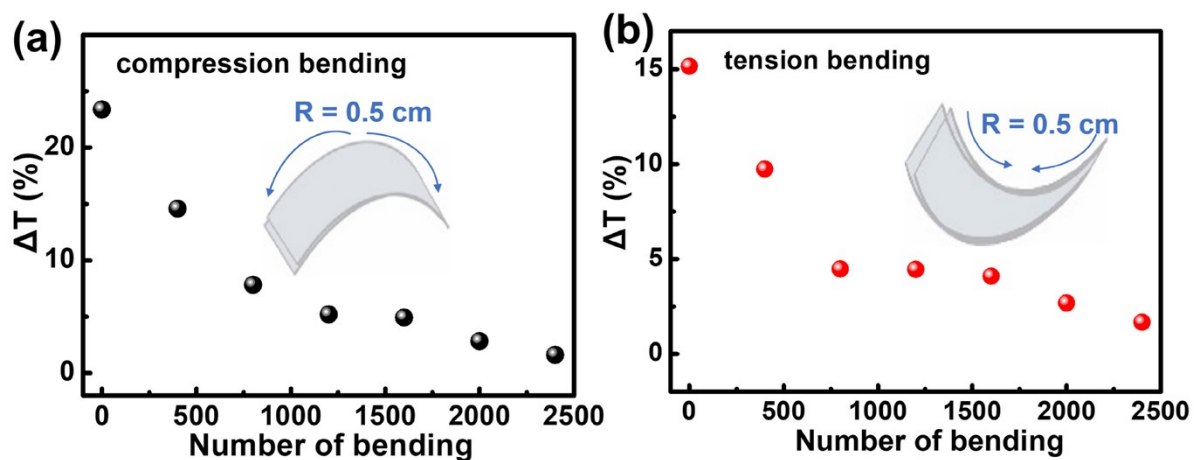


**Figure S14.** The PECD switching speed histogram.

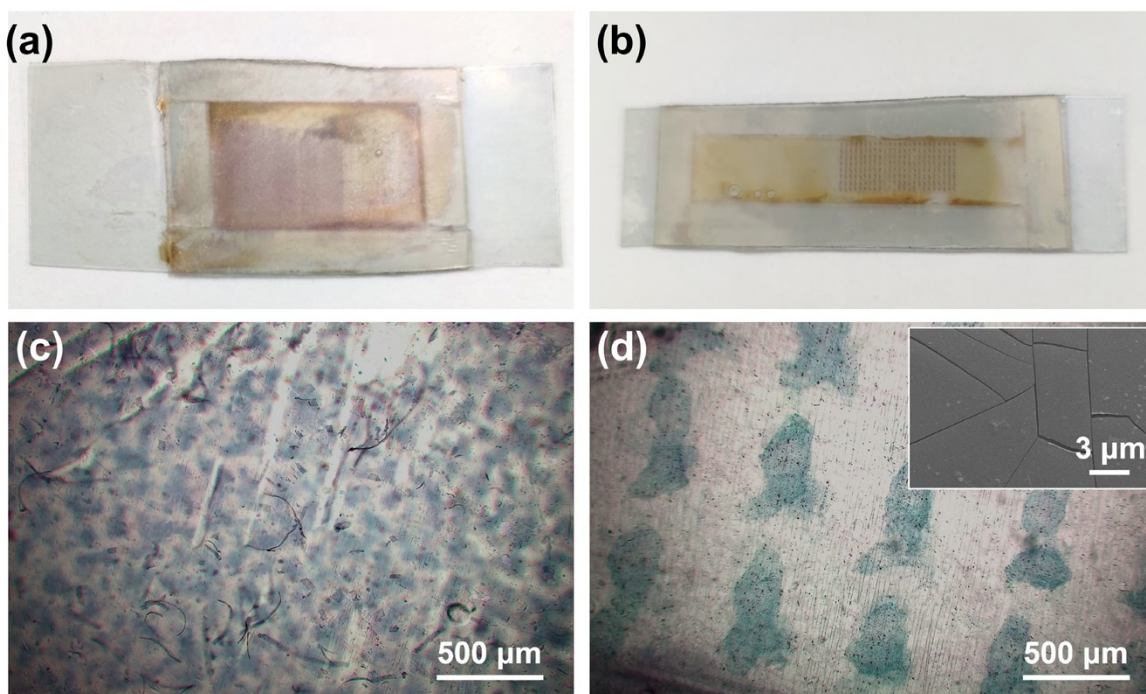




**Figure S15.** SEM images of ITO-coated PET substrate according to the bending cycles ((a) 0 s, (b) 800 s, (c) 1600 s, and (d) 2400 s).



**Figure S16.** (a, b) Changes of  $\Delta T$  in film ECD as a function of number of compression and tension bending cycles (bending radius 5 mm).



**Figure S17.** Photographs and optical microscope images at a magnification of  $4 \times 16$ : (a and c) printed film ECD and (b and d) PECD after 2400 bending cycles (inset showed the SEM image of pixelated Fe(II)-MEPE after 2400 bending cycles).

**Table S1.** The physical parameters of the alternative main solvents and inks.

ink	viscosity (mPa s)	surface tension (mN m <sup>-1</sup> )
water	0.8	71.9
ethylene glycol	17.3	48.4
ethanol	1.1	21.9
original ink	2.1	36.8
water/ethanol/ethylene glycol (100:50:5)	2.2	33.9

**Table S2.** The ratio of Fe(II)-for viscosity

number	water (mL)	ethanol (mL)	ethylene glycol (mL)
1	100	50	0
2	100	50	2
3	100	50	5
4	100	50	15
5	100	50	20
6	100	50	30
7	100	50	40

specific solvent MEPE inks used testing.

**Table S3.** The specific solvent ratio of Fe(II)-MEPE inks used for surface tension testing.

number	water (mL)	ethanol (mL)	ethylene glycol (mL)
1	100	20	5
2	100	35	5
3	100	50	5
4	100	65	5
5	100	80	5

**Table S4.** The EIS fitting results of the electrode.

electrode	$R_s$ ( $\Omega \cdot \text{cm}^2$ )	$R_{ct1}$ ( $\Omega \cdot \text{cm}^2$ )	$R_{ct2}$ ( $\Omega \cdot \text{cm}^2$ )	$W$ ( $\Omega \cdot \text{cm}^2$ )	$R_{ct3}$ ( $\Omega \cdot \text{cm}^2$ )
ITO-coated PET film	48.9	12.5	$5.9 \times 10^3$	—	$9.8 \times 10^4$
printed Fe( II )-MEPE film	55.1	$1.0 \times 10^3$	$7.8 \times 10^{13}$	$7 \times 10^4$	—