Supporting Information

for

Interfacial Coordination Nanosheet Based on Non-Conjugated Three-Arm Terpyridine: A Highly Color-Efficient Electrochromic Material to Converge Fast Switching with Long Optical Memory

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Content

1. ^{1}H NMR spectra of 3tpy ligand

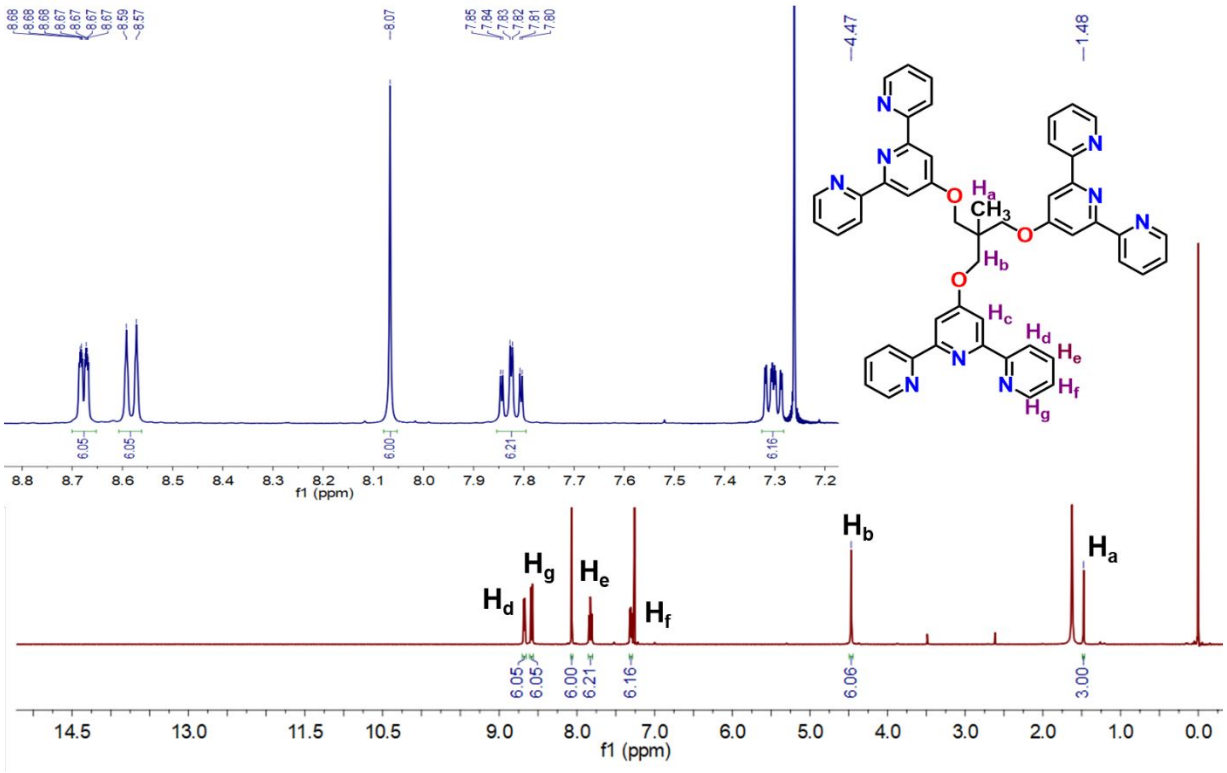

Figure S1. ^{1}H NMR spectra of 3tpy ligand in $CDCl_3$ solvent at room temperature. The chemical structure and the assignment of the different hydrogens are given. The aromatic zone is enlarged in the inset.

2. ^{13}C NMR spectra of 3tpy ligand

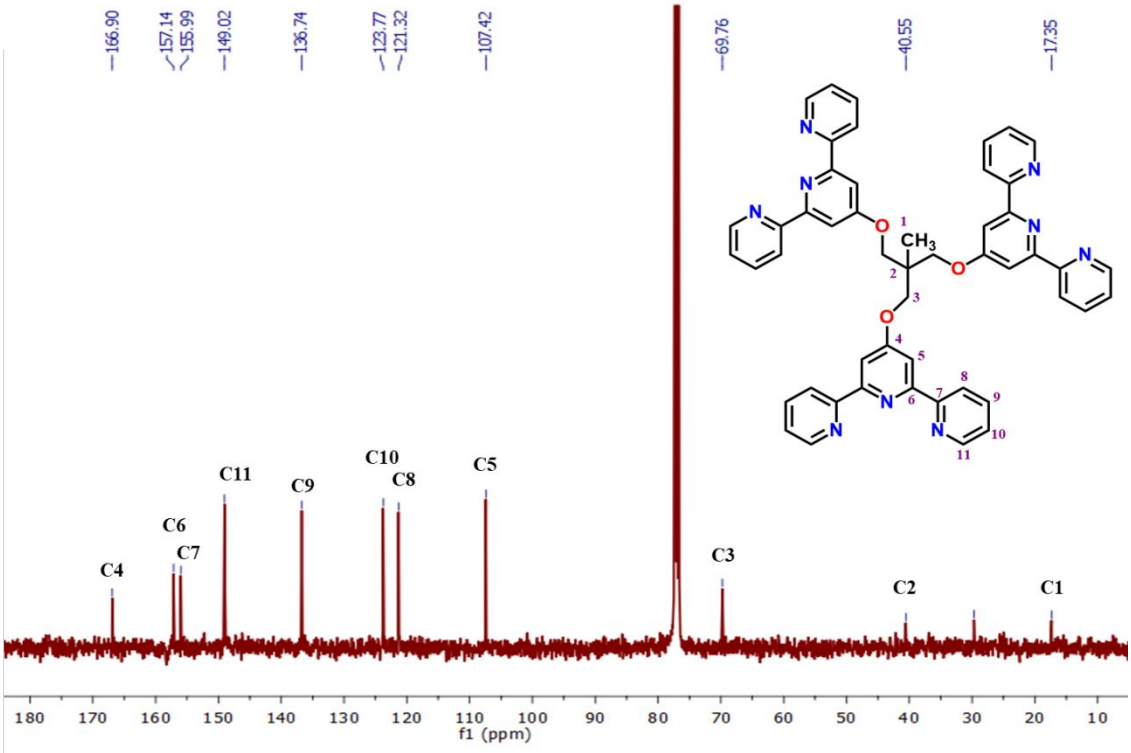

Figure S2. ^{13}C NMR spectra of 3tpy ligand in $CDCl_3$ solvent at room temperature. The chemical structure and the assignment of the different carbons are given.

3. ESI-Mass spectra of 3tpy ligand

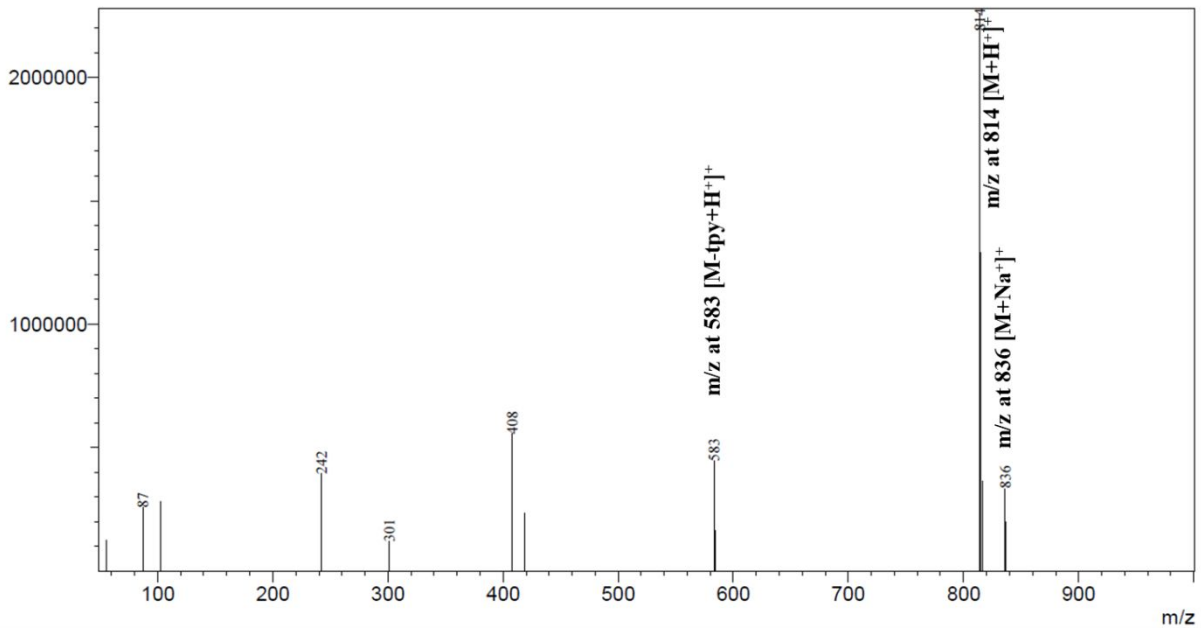

Figure S3. ESI-Mass spectra of 3tpy ligand. The important m/z values are assigned in the picture.

4. Optimization of the film thickness by varying reaction conditions

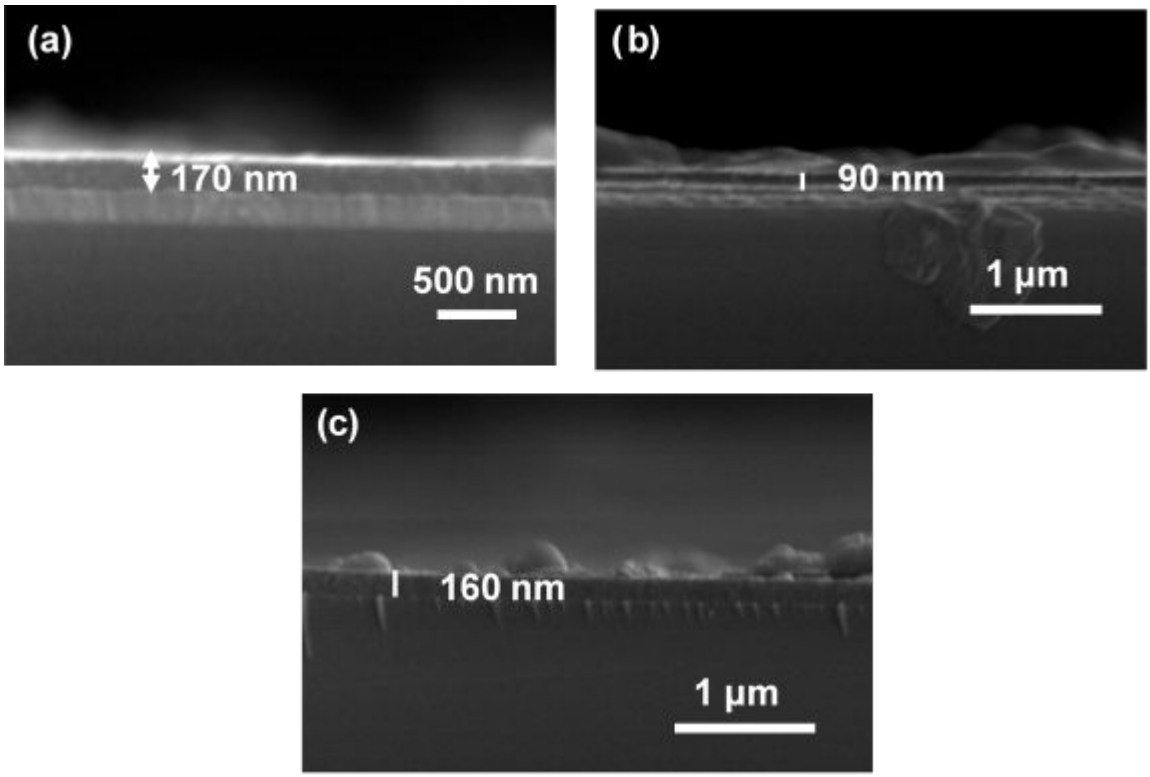

Figure S4. Thickness measurement through cross-sectional SEM study of 3tpy-Fe film using (a) 50 mM Fe^{2+} ion with 0.1 mM 3tpy for 36 h, (b) 50 mM Fe^{2+} ion with 0.1 mM 3tpy for 24 h, and (c) 25 mM Fe^{2+} ion with 0.1 mM 3tpy for 48 h.

Table S1. Summary of the thickness and optical contrast of the films formed by varying the conditions.

Entry	**Concentration of 3tpy (mM)**	**Concentration of Fe^{2+} ions (mM)**	**Reaction time (h)**	**Average film thickness (nm)**	**Optical contrast ΔT (%)#**
1	0.1 mM	50 mM	48 h	~350 nm	53 %
2	0.1 mM	50 mM	36 h	~170 nm	37 %
3	0.1 mM	50 mM	24 h	~90 nm	29 %
4	0.1 mM	25 mM	48 h	~160 nm	35 %

ΔT is optical contrast (transmittance difference between bleached and colored state during solid state EC measurement).

5. UV-Vis titration using 3tpy ligand as a function of Fe^{2+} ions

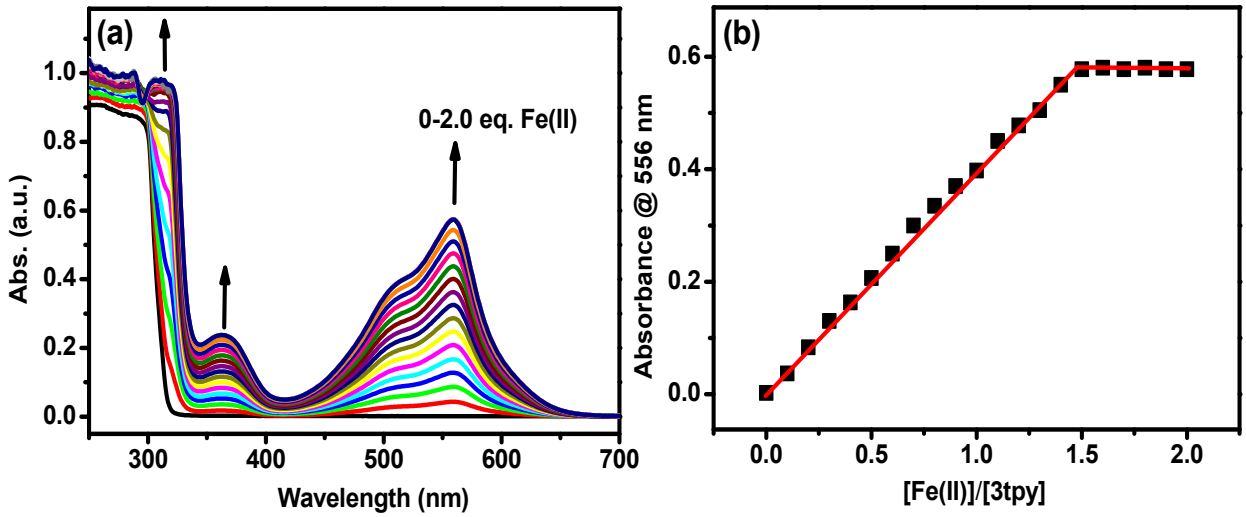

Figure S5. (a) UV-vis spectral change of a methanol solution of 3tpy during titration with $Fe(BF_4)_2$ at room temperature. (b) The absorption change at 556 nm as a function of the $[Fe(BF_4)_2]$/[3tpy] ratio.

6. XPS and TGA study of 3tpy-Fe film

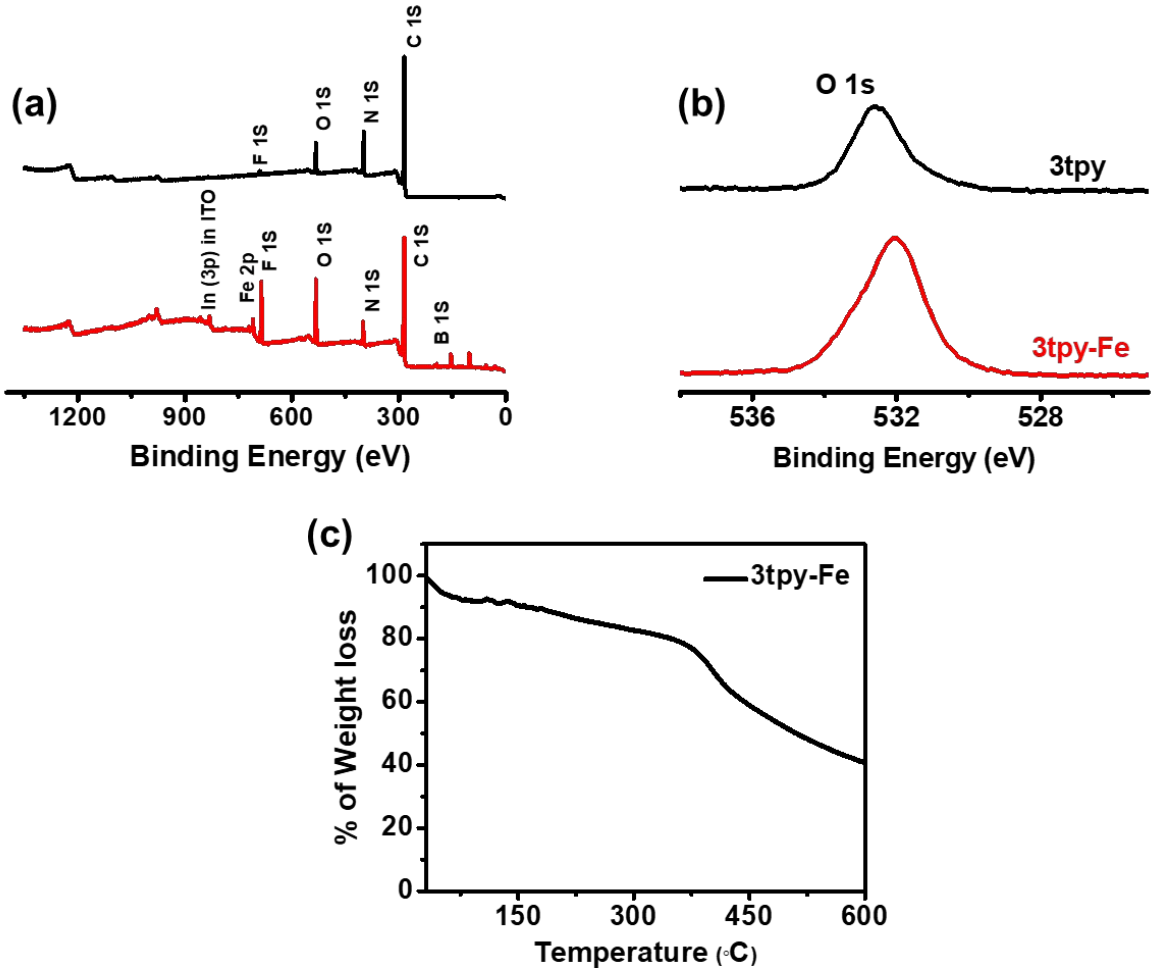

Figure S6. (a) XP survey spectra, (b) XP-spectra focusing on O 1s of 3tpy (black) and 3tpy-Fe (red). (c) Thermogravimetric analysis of 3tpy-Fe in N_2 environment.

7. **UV-Vis spectra comparison of 3tpy-Fe polymers prepared in bulk and interfacial polymerizations.**

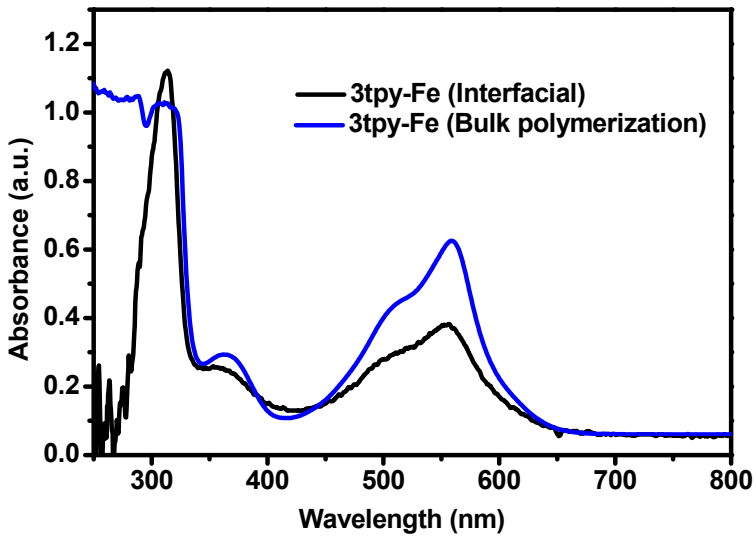

Figure S7. UV-Vis spectra comparison of 3tpy-Fe polymers prepared in bulk and interfacial polymerizations.

8. **Peak current vs. scan rate study of 3tpy-Fe film in three electrode system**

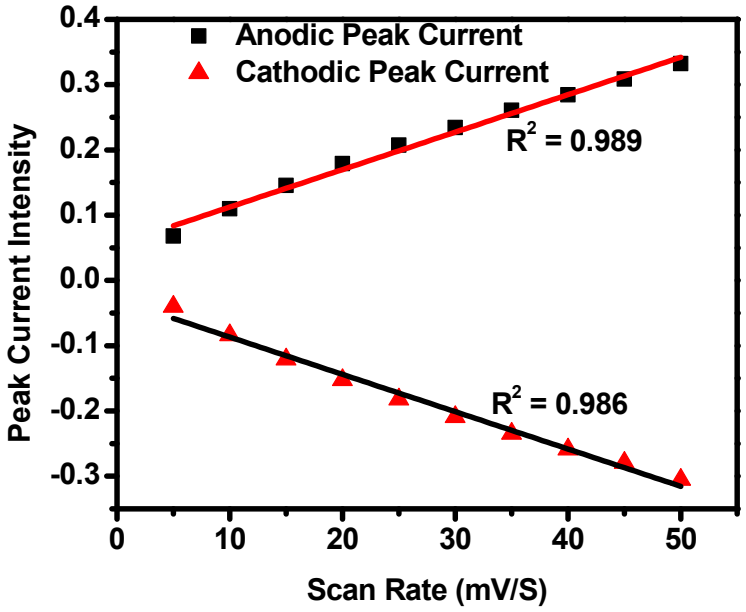

Figure S8. The linear correlations between the peak current and the scan rate during the redox cycle. The above one is for oxidation and below one for reduction and the corresponding R^2 values for fitting are given.

9. The EC properties of 3tpy-Fe film (350 nm) over ITO in three electrode system

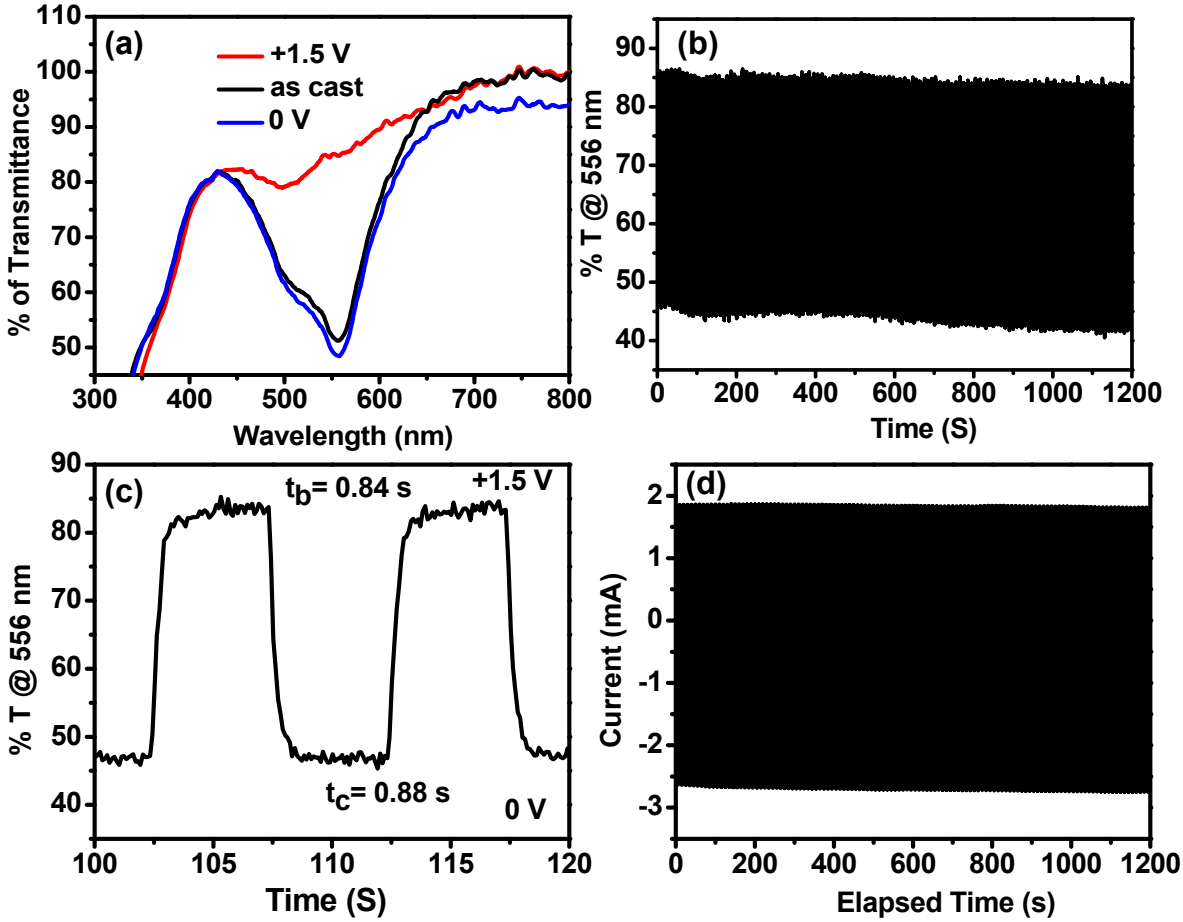

Figure S9. (a) Transmittance spectra of 3tpy-Fe film in three electrode system EC measurement by dipping the film in electrolyte solution. (b) Pulse switching stability (ΔT vs time) of 3tpy-Fe film at 556 nm in 5s interval time between +1.5 V and 0V. (c) Response time for bleaching and coloration of 3tpy-Fe film, and (d) Chronoamperogram (i vs time) during spectrochemical switching time measurement.

10. Electrochromic and chronoamperometry study of 3tpy-Fe film based ECD

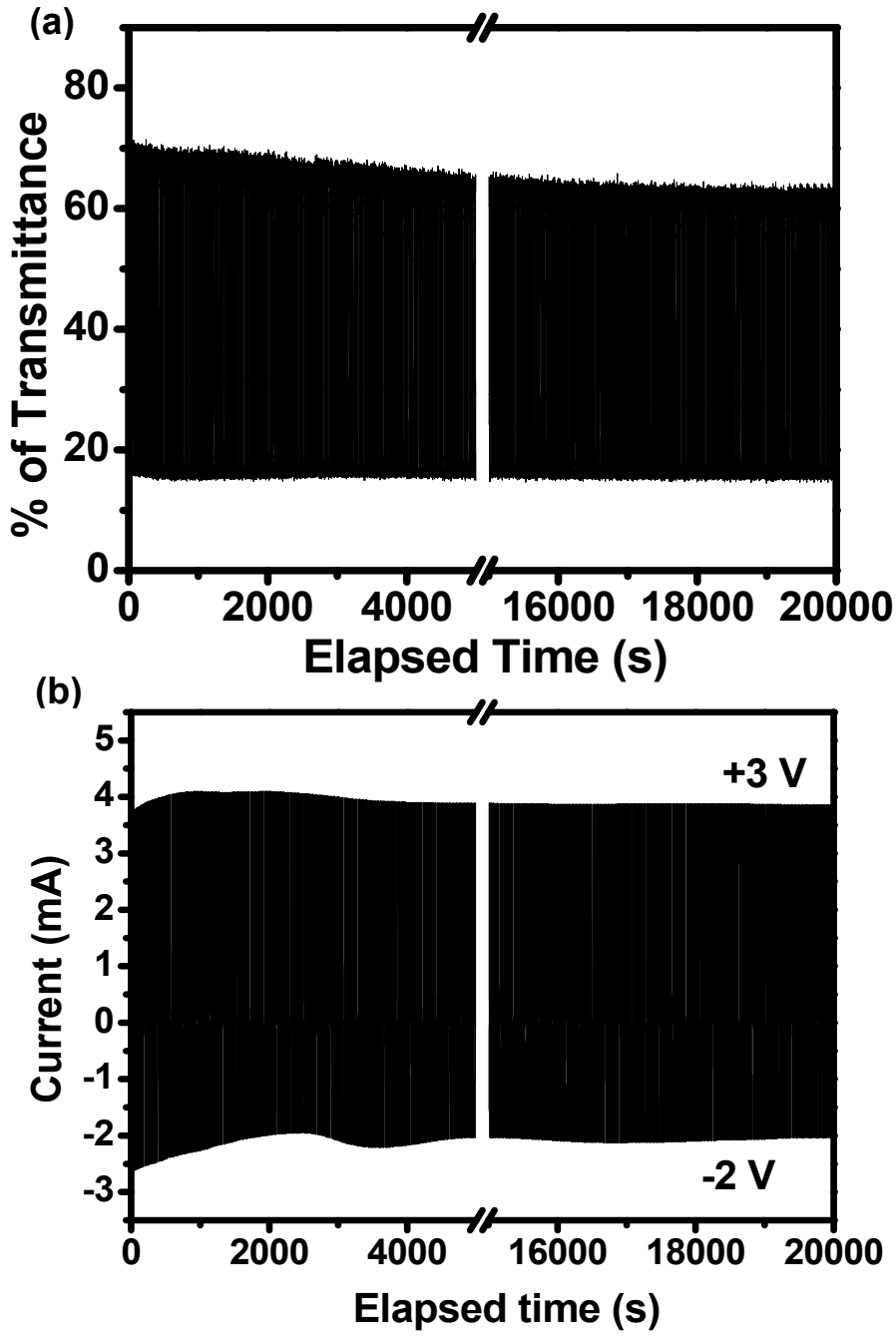

Figure S10. (a) Pulse switching cycle stability (*ΔT* vs. time) of 3tpy-Fe based ECD at 556 nm up to 1000 cycles in 10s interval time between +3V and -2V. (b) Chronoamperogram (*i* vs. time) during spectro-electrochemical switching.

Table S2: Comparison of electrochromic properties with other reported ECMs

Types of materials	**Molecular unit/Framework/ binding ligand**	**Optical contrast/ transmittance change (ΔT %)**	**Coloration efficiency (η cm^2 C^{-1})**	**Solid state ECD fabrication**	**Durability in solid state ECD**	**Memory study**	**Reference**
Metallo-supramolecular polymer (MSP)	Terpyridine based ligand metal complex	Not reported as done with respect to Abs. change	Not reported	Not reported	Not reported	Close to 15 min	*J. Am. Chem. Soc.* **2008**, *130*, 2073-2081
Metallo-supramolecular polymer (MSP)	Terpyridine based ligand	41.6 (for 1D) and 50.7 for 3D polymers	263.8 for 1D and 383.4 for 3D	Not reported	Up to 50 cycles	Not done	*ACS Appl. Mater. Interfaces* **2014,** *6,* 9118-9125
Metal organic molecular assembly	Polypyridyl complex	65	474	Reported	Up to 75 cycles	Not done	*J. Am. Chem. Soc.* **2017**, *139*, 11471-11481
MSP-nanocomposite	Terpyridine based polyFe-nanoclay composite	62.8	572.77	Reported	Up to 1000 cycles	Not done	*Sol. Energy Mater. Sol. Cells* **2020**, *208,* 110392.
Inorganic oxide	WO_3	97.7	118.3	Reported	Up to 10 cycles	Not done	*Chem. Sci.* **2016**, *7*, 1373-1382
Pasmonic	Au and Al metallic nanoslit arrays on polyaniline	90	Not reported	Not reported	Not reported	Not done	*Nat. Commun.* **2016**, *7*, 10479
π-conjugated polymer	dioxythiophene	72	375	Not reported	Not reported	Not done	*Chem. Mater.* **2002**, *14*, 3964-3970
Electropolymerized film of metal-complex	biscyclometalated ruthenium complex	40	250	Not reported	Not reported	Memory close to 1 h	*J. Am. Chem. Soc.* **2011**, *133*, 20720-20723
CONASHs	Terpyridine-metal complex	Not reported as done with respect to Abs. change	Not reported	Reported	Up to 5 cycles	Not done	*J. Am. Chem. Soc.* **2015**, *137*, 4681-4689
CONASHs	Bis(2,2′-bipyridine) based ligand	62	431	shown	Up to 300 cycles	Not done	*ACS Appl. Mater. Interfaces* **2019**, *11, 11893−11903*
CONASH	**Terpyridine-metal complex with non-conjugated ligand**	**53.19**	**470.16**	**Shown**	**Up to 1000 cycles**	**50% retention in 25 min**	**This work**

11. EC memory as a function of thickness of 3tpy-Fe film

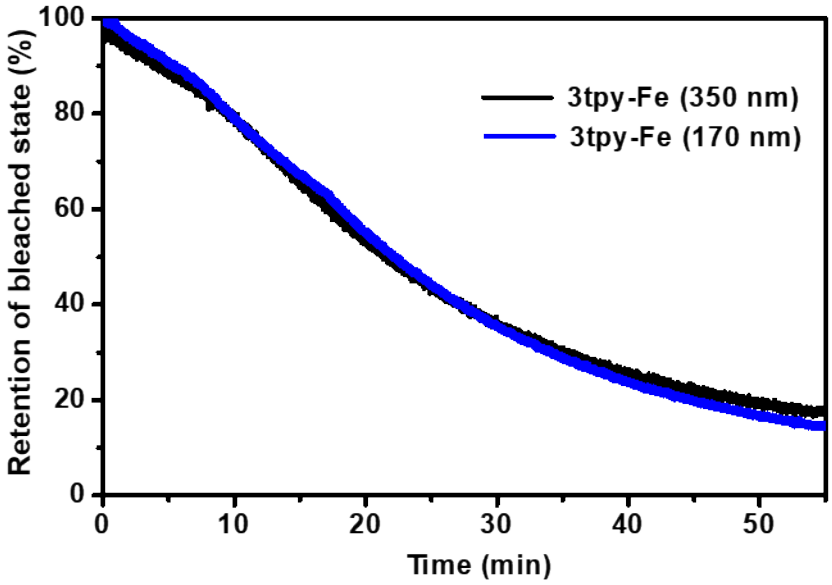

Figure S11. Transmittance decay (normalized electrochromic memory) curve of 3tpy-Fe films with different thickness from bleached state to colored state at open circuit condition.

12. Thickness measurement of polyFe and 3tpyC-Fe film

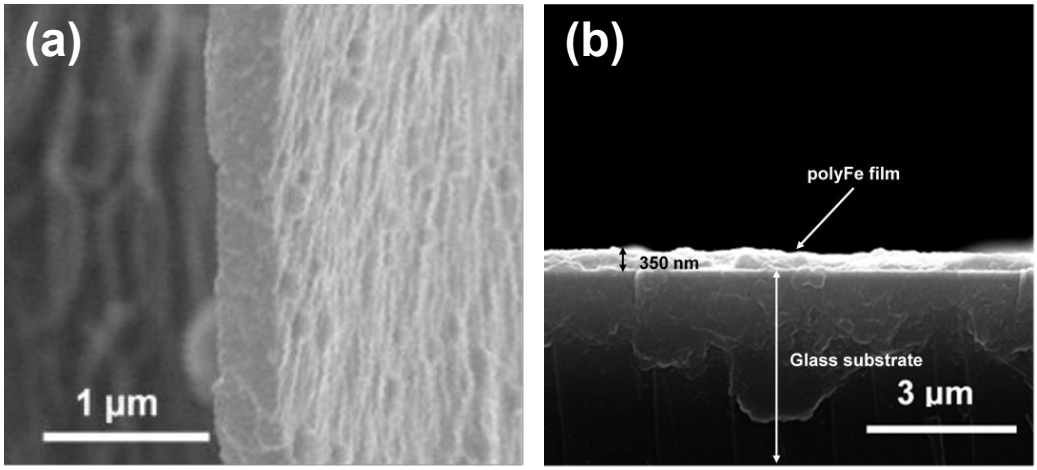

Figure S12. Thickness measurement through cross-sectional FESEM study of (a) 3tpyC-Fe and (b) polyFe film over a glass substrate. The average thickness of the film was measured to be around 350 nm.