Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Supplementary Information for

Ultrafast black color tunability of electrochromic dimming films using cobalt polyoxometalate-anchored nickel oxide nanoparticles

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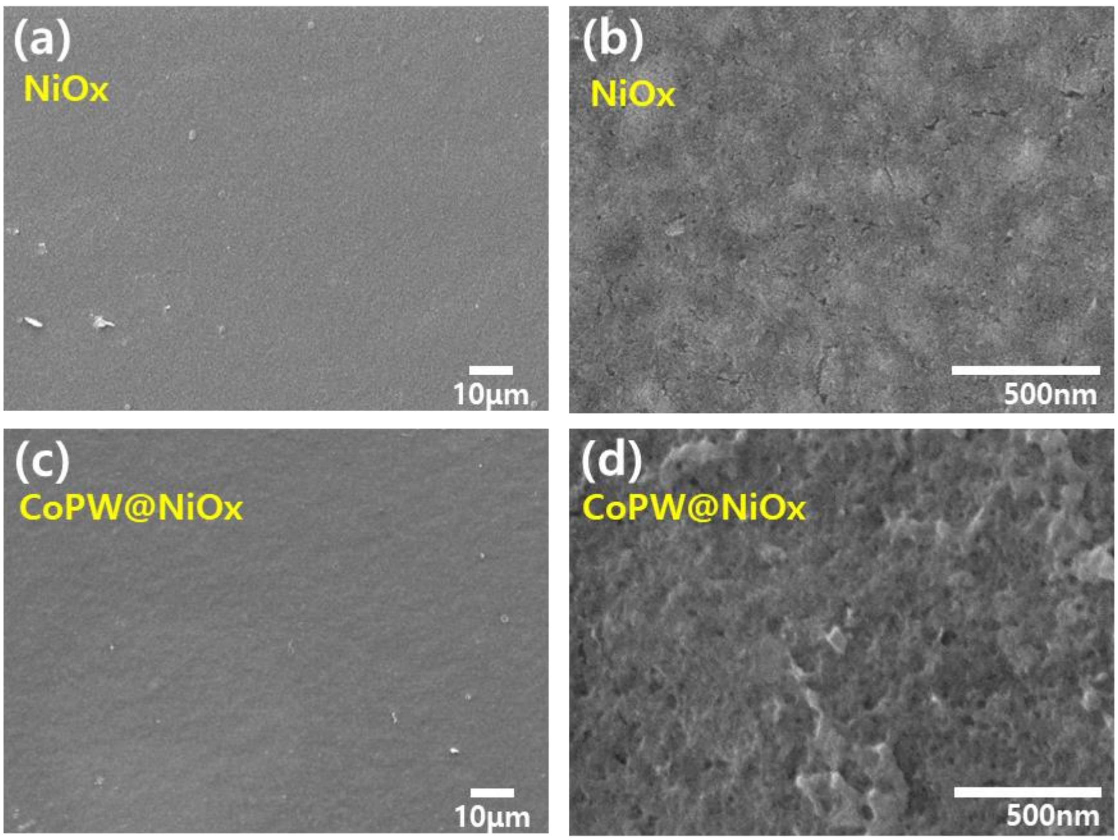


Fig. S1 Top view SEM image of NiOx film and CoPW anchored NiOx Film.

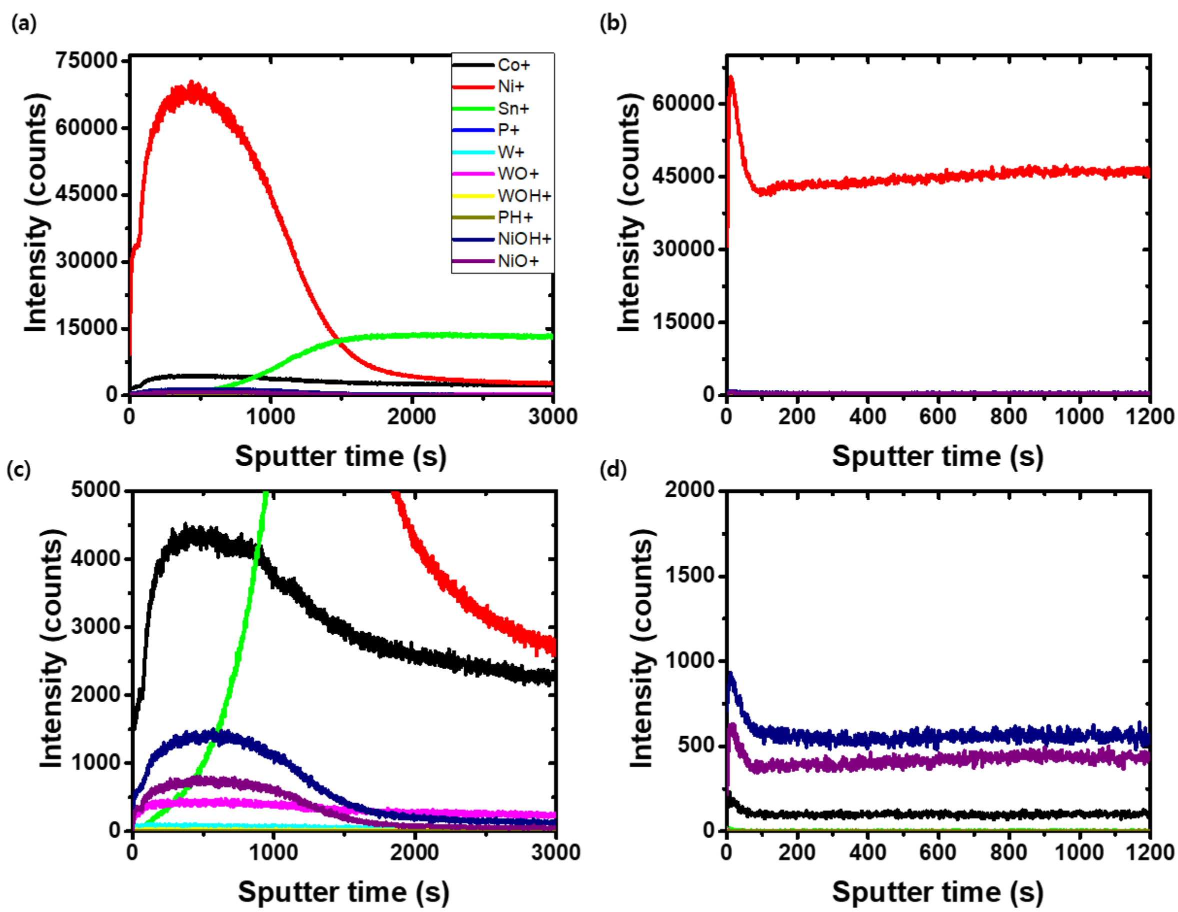
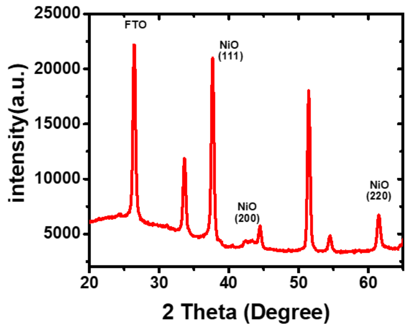


Fig. S2 TOF-SIMS depth profiling of (a, c) CoPW anchored NiOx Film and (b, d) NiOx film

(a)



(b)

No.	h	k	l	d (Å)	F(real)	F(imag)	|F|	2θ	I	M	λ	Phase
1	0	1	1	13.02861	-497.408	-47.0415	499.628	6.779	48.8605	4	1	1
2	0	1	1	13.02861	-497.129	-47.2182	499.366	6.79544	24.2863	4	2	1
3	1	0	-1	11.07676	1186.27	96.0423	1190.16	7.97532	100.0000	2	1	1
4	1	0	-1	11.07676	1186.23	96.3907	1190.14	7.99467	49.7555	2	2	1
5	0	0	2	10.43596	540.896	45.8918	542.839	8.4659	18.4491	2	1	1
6	0	0	2	10.43596	540.708	46.0612	542.666	8.48644	9.1738	2	2	1
7	1	1	0	9.56492	-75.1666	-9.08007	75.7131	9.23846	0.6020	4	1	1
8	1	1	0	9.56492	-74.8167	-9.12045	75.3706	9.26088	0.2969	4	2	1
9	1	0	1	9.48727	295.195	27.4281	296.466	9.31424	4.5400	2	1	1
64	1	2	-3	5.13413	317.184	29.1888	318.524	17.3	1.4913	4	2	1
65	2	1	1	5.11713	-159.021	-20.0993	160.286	17.3156	0.7539	4	1	1
66	2	1	1	5.11713	-158.92	-20.1751	160.195	17.3579	0.3746	4	2	1
166	0	2	5	3.73275	-58.8421	-6.45451	59.195	23.8185	0.0531	4	1	1

Fig. S3 (a) XRD pattern for NiOx film and lattice distance (inset), (b) Simulated lattice distance of CoPW.

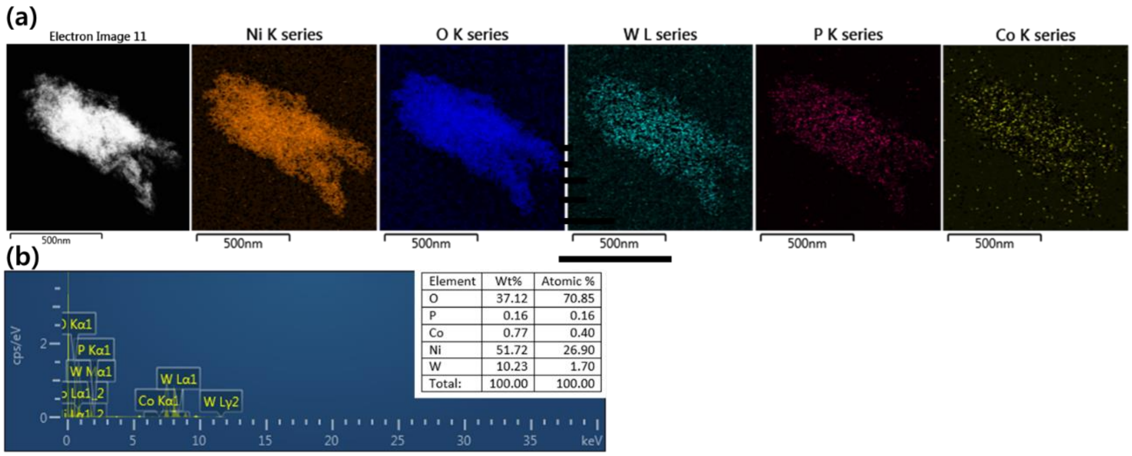


Fig. S4 (a) TEM-EDS mapping of the NiOx-CoPW nanoparticles; nanoparticles from CN400. The sample for TEM analysis was prepared by scratching off the film and dispersed in water. Finally, the water solution was drop on a copper mesh for TEM grid. (b) The corresponding energy-dispersive x-ray spectrum and table of elements

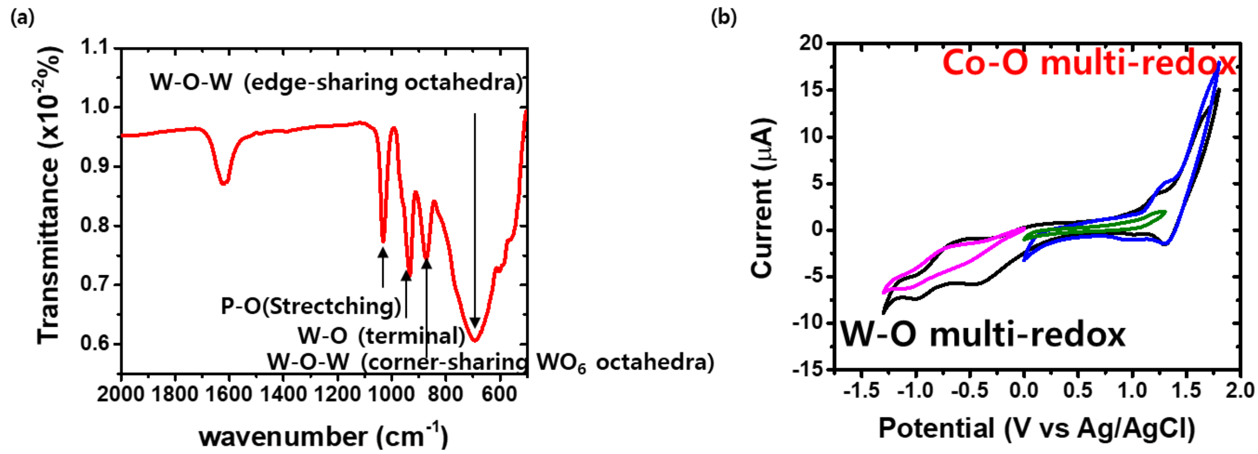


Fig. S5 (a) FT-IR spectra of KCoPW, (b) Cyclic voltammetry of TBACoPW 0.1M in acetonitrile in different potential range (black: 1.8 to -1.3 V, blue: 1.8 to 0 V, green: 1.3 to 0 V and magenta: 0 to -1.3 V, scan rate: 0.1 V/s). The two pairs of redox peaks at -0.54 V and -1.08 V are ascribed to the redox processes of tungsten atoms while the two oxidation peaks at 1.20, 1.67 V and reduction peaks of 1.02 V and 1.30 V correspond to the multi-electron transfer process of CoPW anions. These peaks were also observed in the CV of the CoPW anions in the salt with TBA cations.

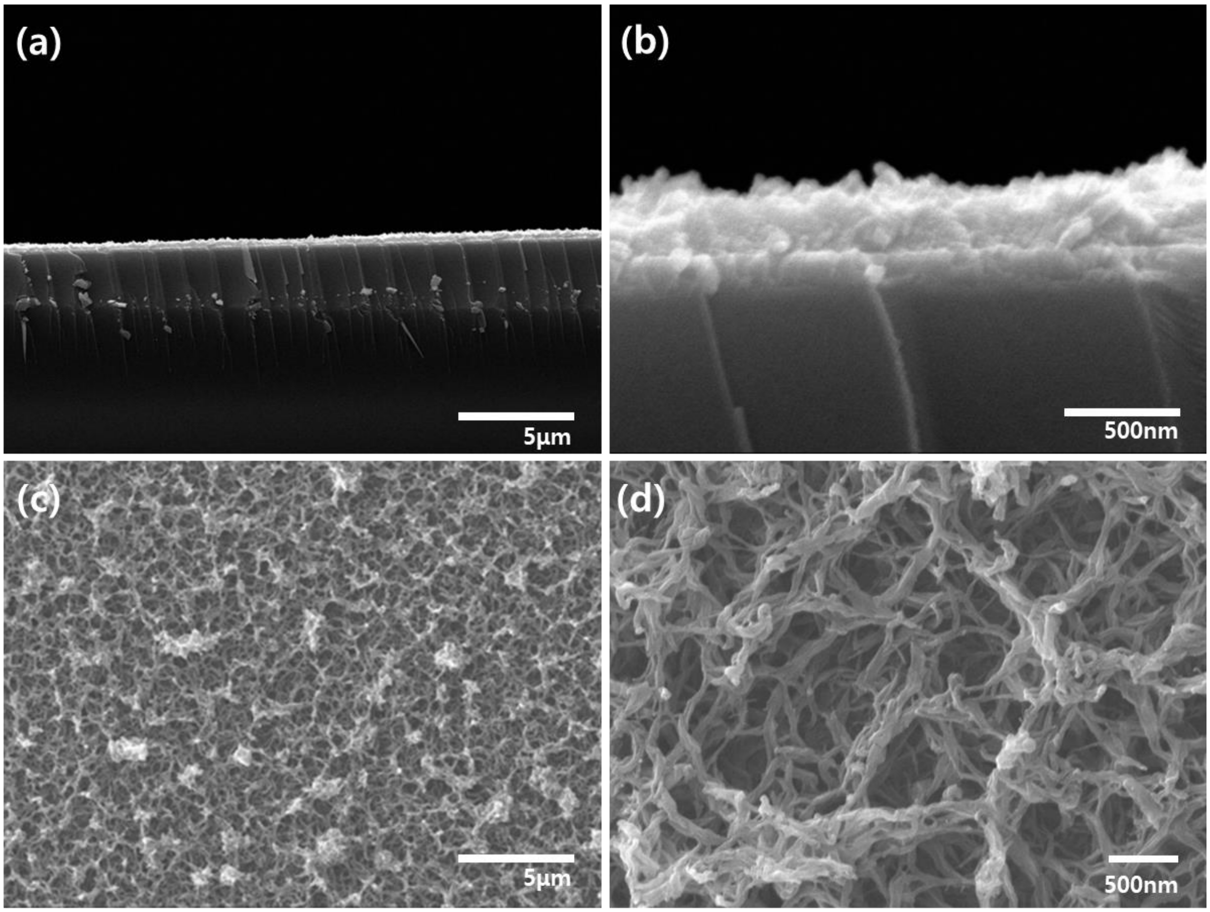


Fig. S6 SEM image of homogeneous electropolymerized PRBr electrode, (a) image of cross section (scale bar: 5μm), (b) image of cross section (scale bar: 500nm), (c) image of top section (scale bar: 5μm), (d) image of top section (scale bar: 500nm)

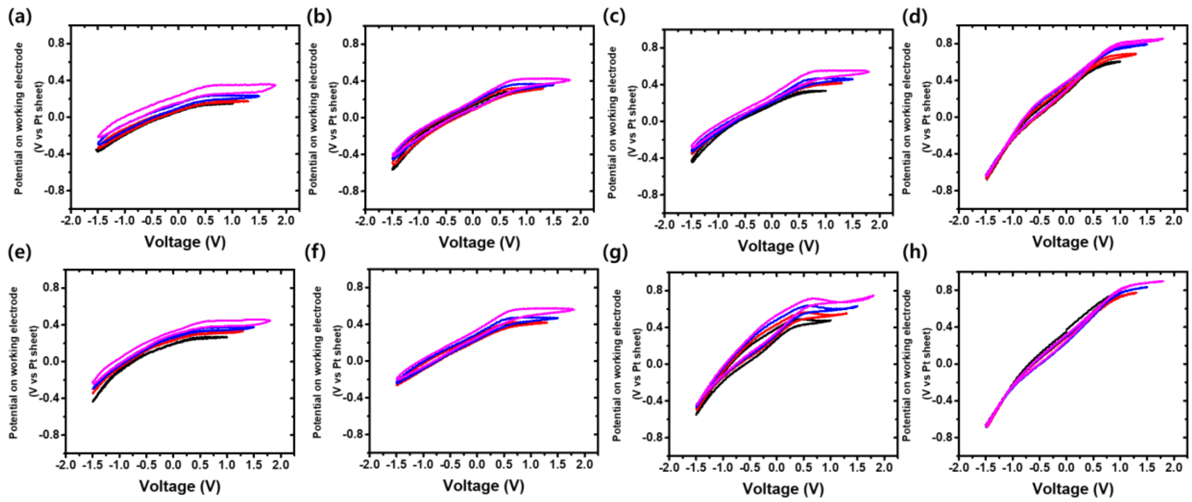


Fig. S7 The potential on the working electrode (solid line, V vs Pt sheet) under the different V_{ap} (black: 1.0/-1.5 V, red: 1.3/-1.5, blue: 1.5/-1.5 V, magenta: 1.8/-1.5 V). V_{ap} was applied by the potentiostat through the cyclic voltammetry with a scan rate of 100 mV/s for (a) PN200, (b) PN400, (c) PN550, (d) PN900, (e) PCN200, (f) PCN400, (g) PCN550 and (h) PCN900

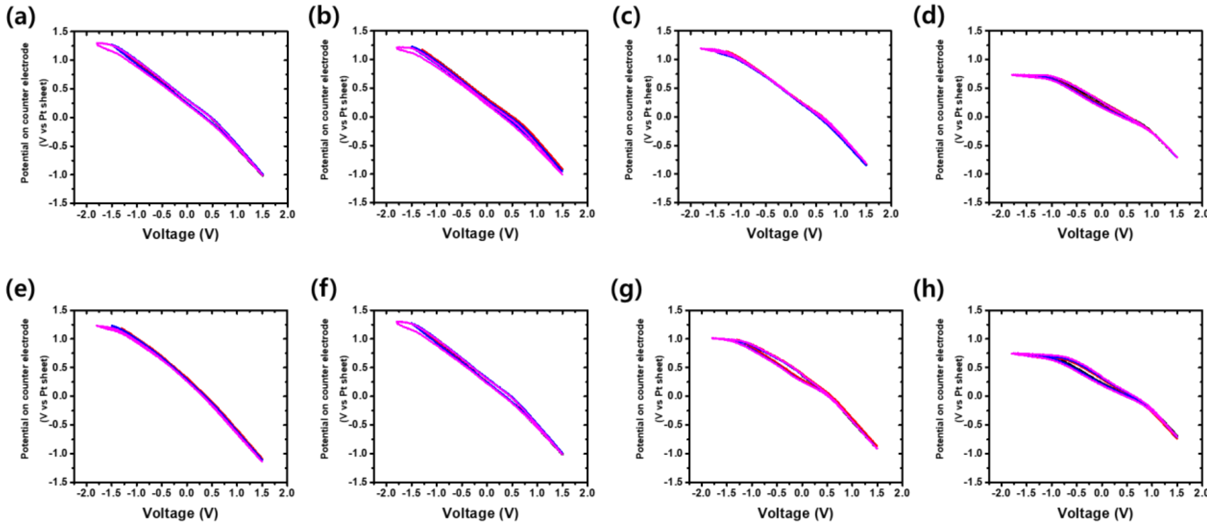


Fig. S8 The potential on the Counter electrode (solid line, V vs Pt sheet) under the different V_{ap} (black: 1.5/-1.0 V, red: 1.5/-1.3 V, blue: 1.5/-1.5 V, magenta: 1.5/-1.8 V). V_{ap} was applied by the potentiostat through the cyclic voltammetry with a scan rate of 100 mV/s for (a) PN200, (b) PN400, (c) PN550, (d) PN900, (e) PCN200, (f) PCN400, (g) PCN550 and (h) PCN900

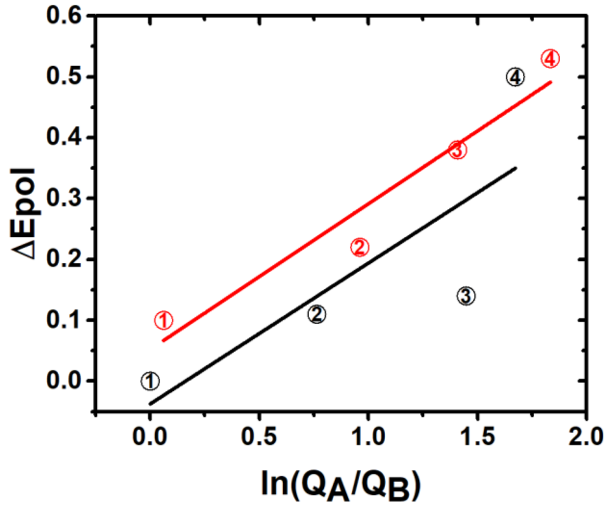


Fig. S9 Nernst relation between change of charge amount during EC reaction and change of E_{pol} by CoPW@NiOx(red) and NiOx(black) or thickness change. Charge amount during EC reaction was measured by cyclic voltammetry that represented as Q_x. Change of charge was estimated as charge ratio (Q_A/Q_B). (1: 200nm, 2: 400nm, 3: 550nm, 4: 900nm)

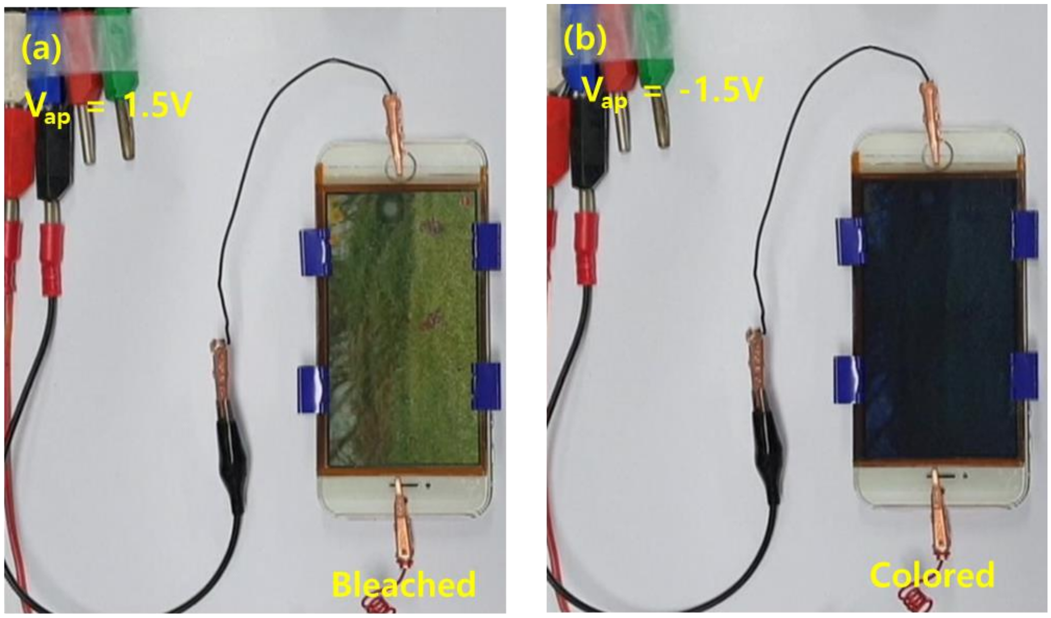


Fig. S10 Photographic image of a 4.1 inch smartphone (9 x 5.2 cm^2) size PCN400 in (a) the bleached state and (b) colored state.

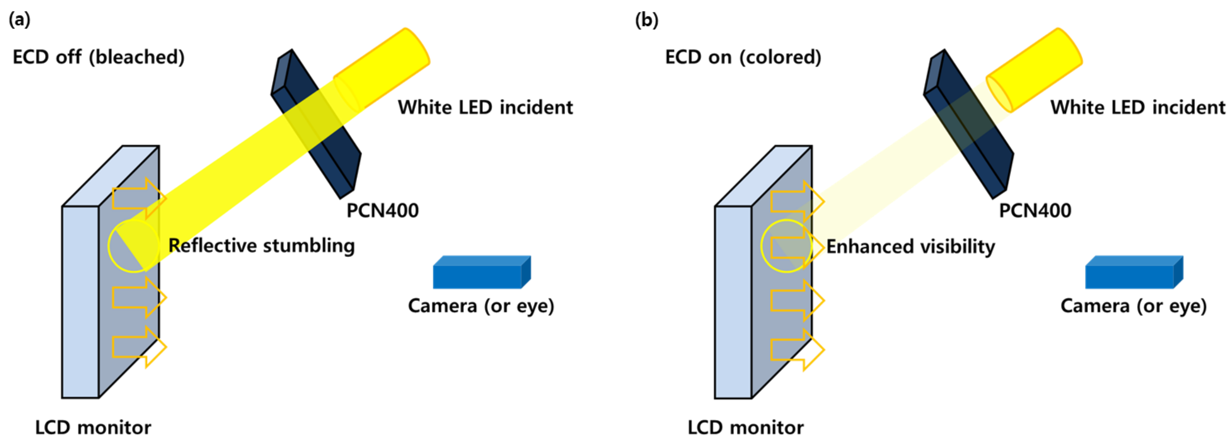


Fig. S11 A block diagram of the experimental setup used to block reflective light from a monitor when the ECD is (a) bleached state and (b) colored state.

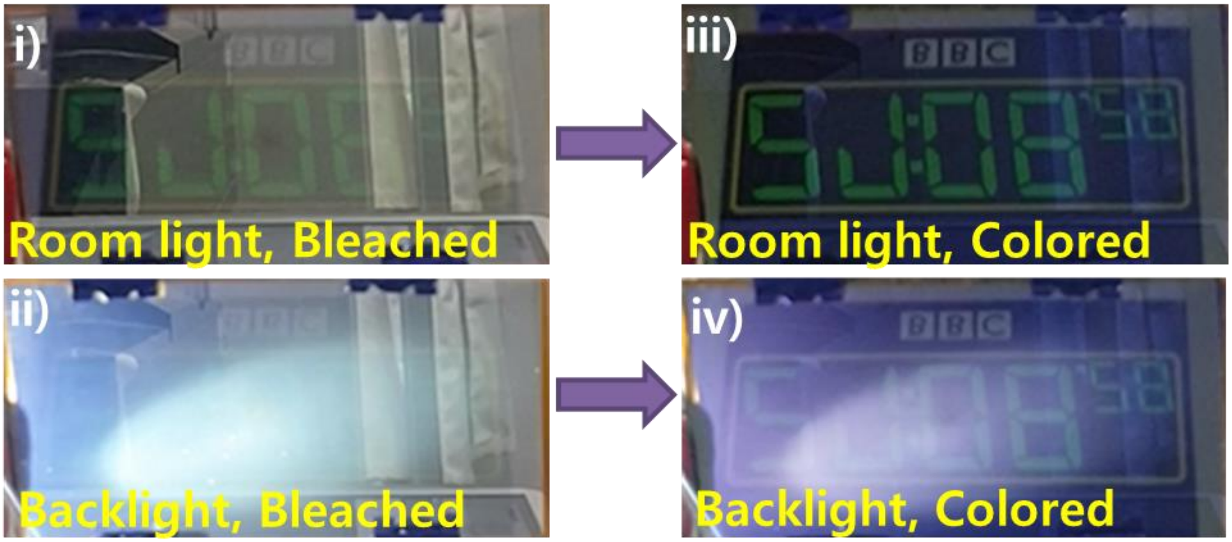


Fig. S12 Photographic images of enhancing visibility test in presence of projection display unit with a 4.1 inch PCN400.

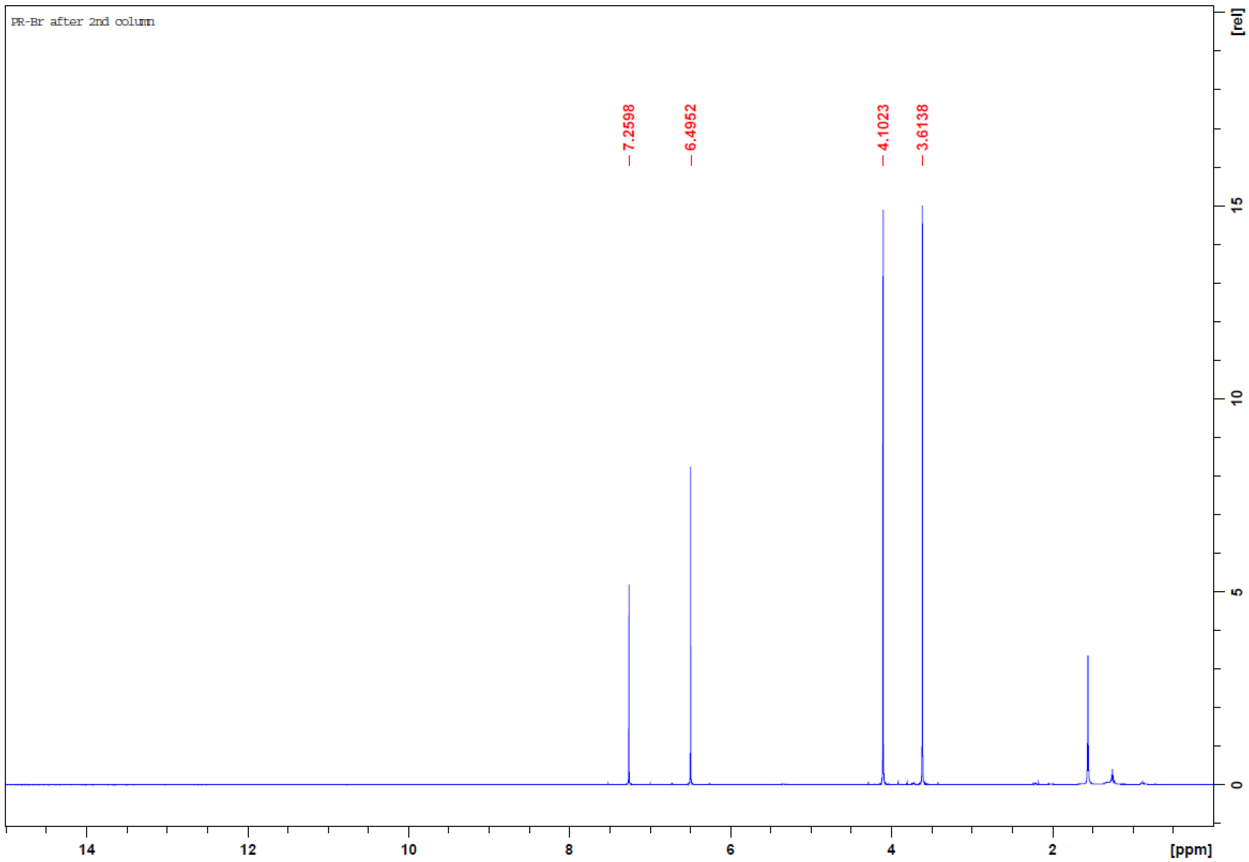


Fig. S13 ^{1}H NMR (300 MHz) spectrum of the 3,3-bis(bromomethyl)-3,4-dihydro-2H-thieno-[3,4-b][1,4]dioxepine. ^{1}H NMR (300 MHz, $CDCl_3$): 6.49 (s, 2H), 4.10 (s, 4H), 3.61 (s, 4H).

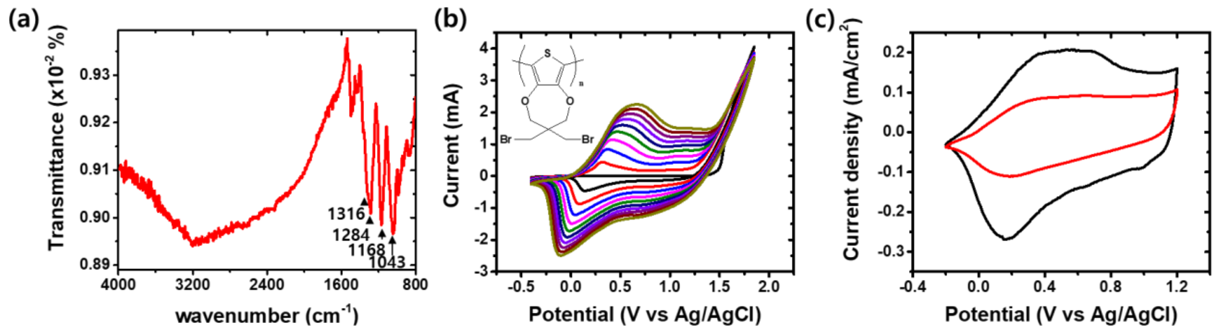


Fig. S14 (a) FT-IR spectra of PRBr collected from the electrodeposited PRBr on an ITO glass, showing a shoulder peak at 1316 cm^{-1} and a peak at 1284 cm^{-1}, characteristic for the stretching vibration of C-C and the C-S-C bond in the thiophene ring, respectively. The peaks at 1168 and 1043 cm^{-1} are attributed to the stretching vibration of C-O in the propylenedioxy bridge and in the ether groups. (b) Cyclic voltammetry during the electrodeposition of PRBr on the ITO glass using a solution of 0.01 M 3,3-bis(bromomethyl)-3,4-dihydro-2H-thieno-[3,4-b][1,4]dioxepine and 0.5 M $LiBF_4$ in propylene carbonate. (c) 3-electrode cyclic voltammetry of solution casting polymerized PRBr (red) and electropolymerized PRBr (black) under BMIMTFSI electrolyte (scan rate: 100 mV/s)

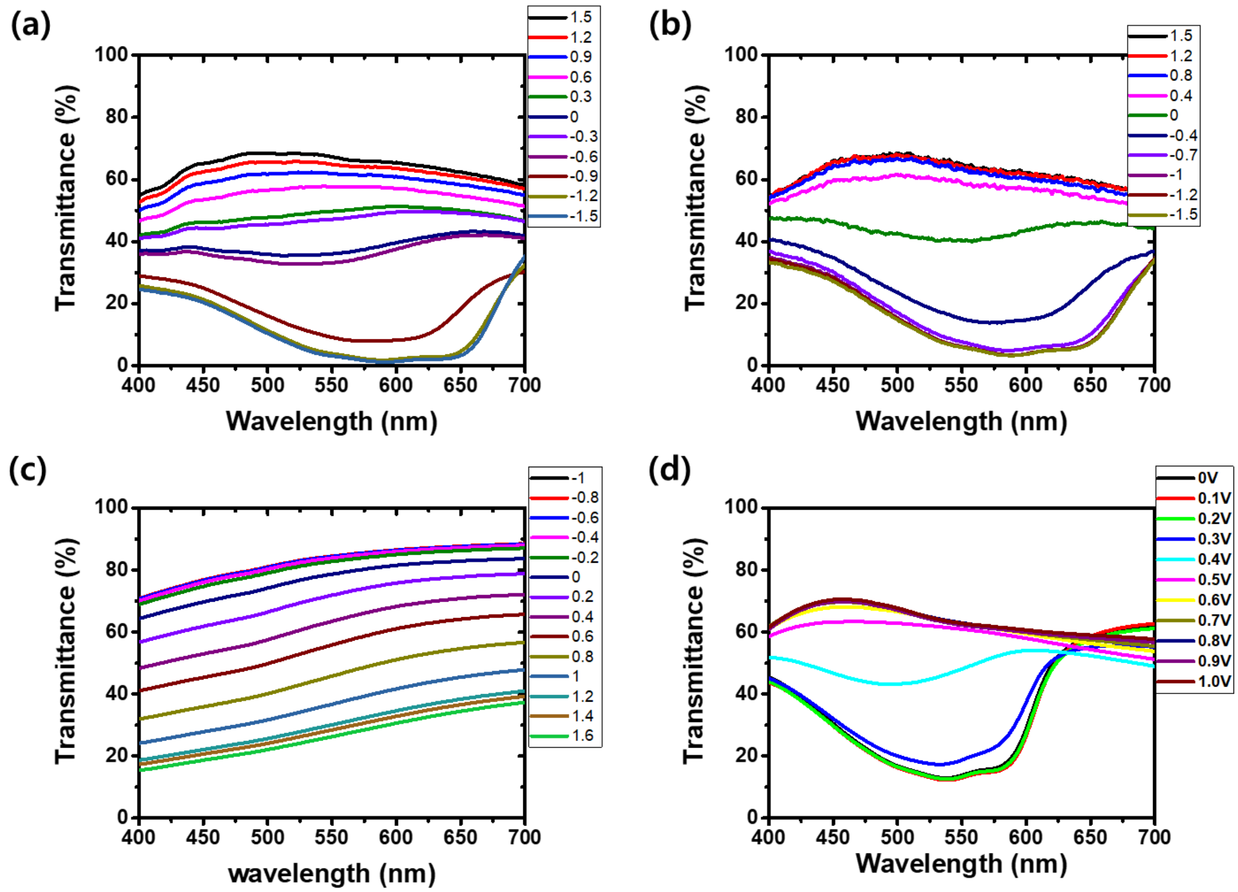


Fig. S15 Electrochromic behaviors of ECDs. Spectral change of PCN400 (a), PN400 (b), CN400 (c), and PN0 (d) over different V_{ap} (inset).

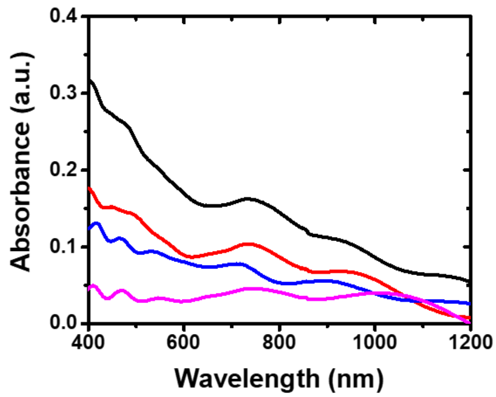


Fig. S16 UV-vis full spectrum of pristine NiOx. Absorption spectra of NiOx with 900 nm thickness (black), 550 nm thickness (red), 400 nm thickness (blue), 200 nm thickness (magenta).

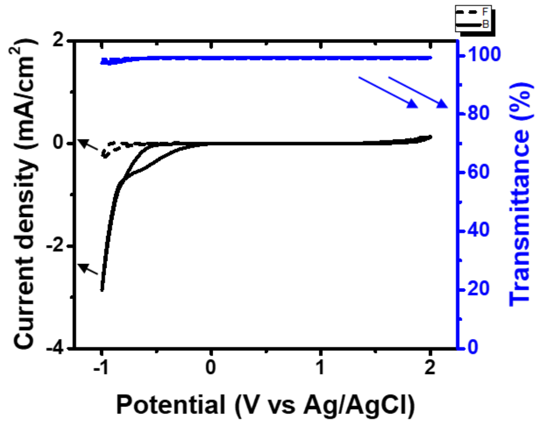


Fig. S17 Cyclic voltammetry (black) at a scan rate of 0.1 V/s and corresponding transmittance change at wavelength of 580 nm (blue) of CoPW (60 μg) coated TiO_2 nanoparticle film on FTO (solid line) as compared to those of a bared TiO_2 nanoparticle film on FTO (dashed line). The thickness of TiO_2 nanoparticle film in both devices was 550 nm.

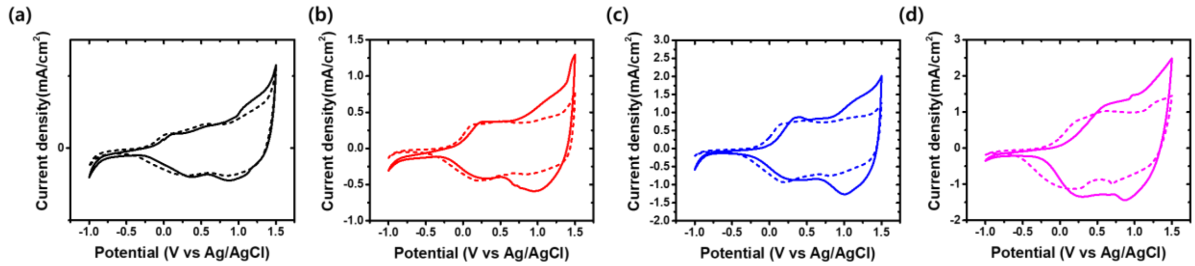


Fig. S18 3-Electrode cyclic voltammetry of (a) N200 (dotted line), CN200 (solid line), (b) N400 (dotted line), CN400 (dotted line), (c) N550 (dotted line), CN550 (solid line), (d) N900 (dotted line), CN900 (solid line).

Table S1. The structure and electrochromic properties of ECDs.

Code	*WE*[a]	CE	Structure[b]	t-NiO[c] (nm)	C_{CoPW}[d] wt (μg/cm^2)	SIMS (μg/cm^2)	V_{ap} (V)	Color	Transmittance[e] (%) T_b	T_c	ΔT[f] (%)	Response time[g] (s) $\tau_{c0.95}$	$\tau_{b0.95}$	Cap[h] (mF/cm^2)	Col_{EF} (cm^2/mC)
N200	NiOx	Pt	3-ED	200	0	0	1.5/-1.0		99	83	16	3.4	1	1.12	20.51
N400	NiOx	Pt	3-ED	400	0	0	1.5/-1.0		95	64	31	2.8	1.6	2.37	21.71
N550	NiOx	Pt	3-ED	550	0	0	1.5/-1.0		88	47	41	2.6	1.2	4.7	17.39
N900	NiOx	Pt	3-ED	900	0	0	1.5/-1.0	Brown-black	83	37	46	6	4.6	5.86	17.96
CN200	CoPW@NiOx	Pt	3-ED	200	15.9	15.9	1.5/-1.0		99	81	18	1.8	1.2	1.17	22.35
CN400	CoPW@NiOx	Pt	3-ED	400	27.4	30.8	1.5/-1.0		95	59	36	2.2	1.8	2.88	21.55
CN550	CoPW@NiOx	Pt	3-ED	550	47.8	40.8	1.5/-1.0		89	37	52	3.8	1.2	5.71	20.03
CN900	CoPW@NiOx	Pt	3-ED	900	56.3	48.2	1.5/-1.0		83	33	50	5.8	3.8	6.9	17.42
PN0	PRBr	FTO	2-ED	0	0	0	1.5/-1.5	Blue	40	36	4	6.2	7.4	0.06	190.7
PN200	PRBr	NiOx	2-ED	200	0	0	1.5/-1.5		37	3	34	0.8	2	0.62	262.3
PN400	PRBr	NiOx	2-ED	400	0	0	1.5/-1.5	Blueish black	69	3	66	0.8	0.8	1.14	330.9
PN550	PRBr	NiOx	2-ED	550	0	0	1.5/-1.5		59	1	58	0.8	0.8	1.4	306.9
PN900	PRBr	NiOx	2-ED	900	0	0	1.5/-1.5		58	2	56	0.8	1.8	1.1	243.7
PCN200	PRBr	CoPW@NiOx	2-ED	200	15.9	15.9	1.5/-1.5	Blue	44	3	41	0.8	2	0.66	302.2
PCN400	PRBr	CoPW@NiOx	2-ED	400	27.4	30.8	1.5/-1.5		74	4	70	0.4	0.8	1.24	367.4
PCN550	PRBr	CoPW@NiOx	2-ED	550	47.8	40.8	1.5/-1.5	Black	62	2	60	0.8	2.6	1.58	332.5
PCN900	PRBr	CoPW@NiOx	2-ED	900	56.3	48.2	1.5/-1.5		59	2	57	1	1.6	0.92	285.6

[a]Working electrode composed of PRBr on ITO glass (the thickness of PRBr =350 nm). [b]3-ED: 3-electrode system with Ag/AgCl as reference electrode, 2-ED: 2-electrode system in the presence of a Pt sheet. [c]Thikness of pristine and CoPW anchored NiOx film. [d]Mass of CoPW contents anchored on NiOx layer determined by measuring the sample weight (wt) and by SIMS. [e]Transmittance at bleached (T_b), colored (T_c) state , and [f]color contrast ($\Delta T = T_b - T_c$), monitored at wavelength of 580 nm, [g]Time for transmittance change for 95 % in coloration (τ_c) and bleaching (τ_b). [h]Capacitance determined at Galvanostatic charging discharging curve in 2-ED and Cyclic voltammetry in 3-ED.

Table S2. Electrochemical properties of ECD under different applied voltages (positive voltage extension).

	PR-Br Thickness (nm)	NiOx Thickness (nm)	C_{CoPW} (μg/cm^2)	T_b [a] (%)	T_c [a] (%)	ΔT_{580nm} [a]	$E_{pol, Oxi}$ [b] (V)	$E_{pol, Red}$ [b] (V)	$E_{ce, Oxi}$ [b] (V)	$E_{ce, Red}$ [b] (V)
1.0V ~ -1.5V										
PN0	350	0	0	33	31	2	0.16	0.12	1.62	-0.84
PCN200	350	200	15.9	34	2	32	0.27	-0.44	1.06	-0.73
PN200	350	200	0	33	3	30	0.15	-0.38	1.12	-0.85
PCN400	350	400	27.4	69	3	64	0.39	-0.26	1.24	-0.61
PN400	350	400	0	62	3	59	0.30	-0.57	0.93.	-0.70
PCN550	350	550	47.8	52	2	50	0.57	-0.49	1.01	-0.43
PN550	350	550	0	50	1	49	0.33	-0.44	1.06	-0.67
PCN900	350	900	56.3	55	2	53	0.64	-0.67	0.83	-0.36
PN900	350	900	0	56	2	54	0.72	-0.66	0.84	-0.28
1.3V ~ -1.5V										
PN0	350	0	0	35	33	2	0.23	0.15	1.65	-1.07
PCN200	350	200	15.9	39	2	37	0.34	-0.35	1.15	-0.96
PN200	350	200	0	33	3	30	0.18	-0.34	1.16	-1.12
PCN400	350	400	27.4	70	3	67	0.43	-0.26	1.24	-0.87
PN400	350	400	0	66	3	63	0.32	-0.51	0.99	-0.98
PCN550	350	550	47.8	60	2	58	0.59	-0.49	1.01	-0.71
PN550	350	550	0	57	1	56	0.38	-0.39	1.11	-0.92
PCN900	350	900	56.3	58	2	56	0.74	-0.64	0.86	-0.56
PN900	350	900	0	57	2	55	0.74	-0.72	0.78	-0.56
1.5V ~ -1.5V										
PN0	350	0	0	40	36	4	0.29	0.17	1.67	-1.21
PCN200	350	200	15.9	44	3	41	0.38	-0.30	1.2	-1.12
PN200	350	200	0	37	3	34	0.24	-0.28	1.22	-1.26
PCN400	350	400	27.4	73	4	69	0.48	-0.25	1.25	-1.02
PN400	350	400	0	69	3	66	0.36	-0.45	1.05	-1.14
PCN550	350	550	47.8	62	2	60	0.64	-0.46	1.04	-0.86
PN550	350	550	0	59	1	58	0.42	-0.35	1.15	-1.08
PCN900	350	900	56.3	59	2	57	0.82	-0.62	0.88	-0.68

PN900	350	900	0	58	2	56	0.79	-0.71	0.79	-0.71
1.8V ~ -1.5V										
PN0	350	0	0	45	40	5	0.38	0.22	1.72	-1.42
PCN200	350	200	15.9	55	3	52	0.46	-0.24	1.26	-1.34
PN200	350	200	0	54	3	51	0.36	-0.20	1.3	-1.44
PCN400	350	400	27.4	74	4	70	0.58	-0.20	1.3	-1.22
PN400	350	400	0	70	3	67	0.42	-0.41	1.09	-1.38
PCN550	350	550	47.8	65	2	63	0.74	-0.45	1.05	-1.06
PN550	350	550	0	60	1	59	0.47	-0.32	1.18	-1.33
PCN900	350	900	56.3	60	2	58	0.89	-0.63	0.87	-0.91
PN900	350	900	0	59	2	57	0.86	-0.76	0.74	-0.94

[a]T_b = transmittance at bleached state, T_c = transmittance at colored state, $\Delta T = T_b - T_c$, monitored at 580 nm under given E_{ap}, [b] E_{pol} and E_{CE} are the measured potential at WE and CE under the given V_{ap}, respectively. $E_{pol.oxi}$ and $E_{pol.red}$ are recorded potential on working electrode when the working electrode in ECD undergoes oxidation state and reduction state, respectively. $E_{CE.oxi}$ and $E_{CE.red}$ are recorded potential on counter electrode under oxidation state and reduction state.

Table S3. Electrochemical properties of ECD under different applied voltages (negative voltage extension).

	PR-Br Thickness (nm)	NiOx Thickness (nm)	C_{CoPW} (μg/cm^2)	T_b [a] (%)	T_c [a] (%)	ΔT_{670nm} [a]	$E_{pol, Oxi}$ [b] (V)	$E_{pol, Red}$ [b] (V)	$E_{ce, Oxi}$ [b] (V)	$E_{ce, Red}$ [b] (V)
1.5V ~ -1.0V										
PN0	350	0	0	43	42	1				
PCN200	350	200	15.9	49	25	24	0.4	0	1.00	-1.1
PN200	350	200	0	46	25	21	0.4	0.05	1.05	-1.1
PCN400	350	400	27.4	62	21	41	0.56	-0.12	0.88	-0.94
PN400	350	400	0	61	32	29	0.52	0.04	1.04	-0.98
PCN550	350	550	47.8	50	21	29	0.68	-0.12	0.88	-0.82
PN550	350	550	0	55	15	40	0.62	0.04	1.04	-0.88
PCN900	350	900	56.3	57	13	44	0.78	-0.33	0.67	-0.72
PN900	350	900	0	56	14	42	0.81	-0.31	0.69	-0.69
1.5V ~ -1.3V										
PN0	350	0	0	45	44	1				
PCN200	350	200	15.9	49	17	32	0.4	-0.13	1.17	-1.1
PN200	350	200	0	46	17	28	0.4	-0.03	1.27	-1.1
PCN400	350	400	27.4	62	15	47	0.6	-0.23	1.07	-0.9
PN400	350	400	0	61	21	40	0.51	-0.01	1.29	-0.99
PCN550	350	550	47.8	55	11	44	0.69	-0.23	1.07	-0.81
PN550	350	550	0	55	9	46	0.64	-0.13	1.17	-0.86
PCN900	350	900	56.3	57	11	46	0.79	-0.62	0.68	-0.71
PN900	350	900	0	56	11	45	0.77	-0.56	0.74	-0.73
1.5V ~ -1.5V										
PN0	350	0	0	45	44	1				
PCN200	350	200	15.9	48	13	35	0.35	-0.28	1.22	-1.15
PN200	350	200	0	44	13	31	0.38	-0.18	1.32	-1.12
PCN400	350	400	27.4	62	14	48	0.6	-0.53	0.97	-0.9
PN400	350	400	0	60	16	44	0.49	-0.12	1.38	-1.01
PCN550	350	550	47.8	55	9	46	0.66	-0.53	0.97	-0.84
PN550	350	550	0	55	8	47	0.6	-0.33	1.17	-0.9
PCN900	350	900	56.3	57	11	46	0.79	-0.78	0.72	-0.71

PN900	350	900	0	56	11	45	0.8	-0.78	0.72	-0.7
1.5V ~ -1.8V										
PN0	350	0	0	44	42	2				
PCN200	350	200	15.9	48	13	35	0.3	-0.57	1.23	-1.2
PN200	350	200	0	43	12	31	0.35	-0.47	1.33	-1.15
PCN400	350	400	27.4	62	14	48	0.54	-0.74	1.06	-0.96
PN400	350	400	0	60	14	46	0.48	-0.28	1.52	-1.02
PCN550	350	550	47.8	55	8	47	0.7	-0.74	1.06	-0.8
PN550	350	550	0	55	8	47	0.6	-0.56	1.24	-0.9
PCN900	350	900	56.3	57	11	46	0.8	-1.06	0.74	-0.7
PN900	350	900	0	57	10	47	0.83	-1.04	0.76	-0.67

Table S4. The EC property compared with nickel oxide based darkish electrochromic devices.

Material	**Cell type (half/full cell)**	**ΔT (%)**	**T_c (%)**	**T_b (%)**	**$\tau_{c0.95}$ (s)**	**$\tau_{b0.95}$ (s)**	**$\lambda_{measured}$ (nm)**	***Color***	**Voltage Range (V)**	***Ref.***
PCN400(this work)	Full cell	**70**	**4**	**74**	**0.4**	**0.8**	**580**	**Black**	**1.5 to -1.5**	This paper
NiO-pyridine	Half cell	41.5	46.5	87	1.77	2.91	550	Black	1.5 to -1.5	(1)
Bi/Cu(Electrodeposition mechanism)	Full cell	69	11	80	30	3	600	black	0.8 to -0.6	(2)
cross-linked poly(4-vinyltriphenylamine) (480nm)/Cu	Full cell	82.9	5.1	88	8.5	68.7	700	black	2 to -2	(3)
Sputtered Nickel oxide (Ta. Ni, W)	Full cell	67.1	11.9	79	14.9	1.5	600	Brown-black	1.8 to -1.8	(4)
Si-Li codoping Nickel oxide coating with sputtered Zinc tin oxide	Half cell	35.5	50.0	85.5	10.2	7.8	550	Brown	1.2 to -1.2	(5)
Ni-Co mixed hydroxide	Half cell	38.2	37.7	75.9	1	1	635	Brown-black	0.6 to 0	(6)

Table S5. Electrochemical properties and carrier concentration of N200 ~ 900 and CN200 ~ 900.

Electrode	[A] C_{CoPW} ($\mu g/cm^2$)	[B] C_{CoPW} ($\mu g/cm^2$)	$E^{P.1st}_{ox}$ (V)	Current density (mA/cm2)	$E^{P.2nd}_{ox}$ (V)	Current density (mA/cm2)	$E^{P.1st}_{red}$ (V)	Current density (mA/cm2)	$E^{P.2nd}_{red}$ (V)	Current density (mA/cm2)	[C] $E^{1st}_{1/2}$ (V)	[C] $E^{2nd}_{1/2}$ (V)	Areal charge (mC/cm2)	% increase in charge amount	[D] *Carrier concentration* *($/cm^{-3}$)*
N200	0	0	0.58	0.17	1.02	0.21	0.87	-0.19	0.25	-0.19	0.42	0.95	5.6	-	-
CN200	15.9	15.9	0.57	0.15	1.13	0.31	0.88	-0.22	0.37	-0.2	0.47	1.01	5.9	5	-
N400	0	0	0.54	0.37	1.06	0.41	0.87	-0.37	0.19	-0.44	0.37	0.97	11.8	-	3.06E+19
CN400	27.4	30.8	0.55	0.37	1.18	0.72	0.98	-0.59	0.3	-0.42	0.43	1.08	14.4	22	3.69E+19
N550	0	0	0.51	0.77	1.13	0.87	0.85	-0.74	0.21	-0.93	0.36	0.99	23.5	-	1.584E+20
CN550	47.8	40.7	0.42	0.87	1.19	1.41	1.01	-1.26	0.32	-0.87	0.37	1.1	28.5	21	1.851E+20
N900	0	0	0.61	1.03	1.33	1.32	0.71	-0.99	0.18	-1.13	0.4	1.02	29.3	-	-
CN900	56.3	48.2	0.66	1.23	1.26	1.87	0.86	-1.44	0.26	-1.34	0.46	1.06	34.5	17	-

C_{CoPW} is the contents of CoPW determined by [A] weighing the mass of the film and [B] calculating according to the weight percentage of elements obtained from the TOF-SIMS. E^P_{red} and E^P_{ox} (V vs vs Ag/AgCl) were determined from the cyclic voltammetry in 1 M Li^+ containing BMIM TFSI, which was used as the electrolyte. [C] Half-wave potential. [D] Carrier concentration was obtained from the Hall effect measurement.

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