## **Supplementary Information**

## High performance NiO<sub>x</sub> nanoplatelet based films by scrape-coating method for bifunctional electrochromic and energy storage devices

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Fig.S1 Schematic diagram of the dynamics of electrochromic mechanism of  $NiO_x$  films in  $PC/LiClO_4$ .



Fig.S2 XPS spectra of  $NiO_x$  NPs: (a) O 1s and (b) Ni 2p



Fig.S3 SEM image of (a)  $NiO_x$ -0; (b)  $NiO_x$ -0.25 and (c)  $NiO_x$ -0.50 films.



Fig.S4 Diagram of the mechanism of formation of Ni vacancies (Ni atoms in green, O atoms in blue).



Fig.S5 Surface structures of NiO. Ball-and-stick model of the surface of NiO:  $\gamma$  (200) surface,  $\delta$  (200) surface with a Ni vacancy,  $\eta$  (220) surface,  $\lambda$  (220) surface with a Ni vacancy. Here red ball is Oxygen atom and gray ball is Nickel atom.



Fig.S6 (a) XRD and (b) FTIR spectra of NiO<sub>x</sub> NPs films with different PEG4000 contens



Fig.S7 The maximum transmittance spectra of NiO<sub>X</sub>-0.05 films after different number of cycles.



Fig.S8 Dynamic transmittance spectra and colouring efficiency plots of NiOx-0.05 films. (a) and (b) after 500 cycles; (c) and (d) after 1000 cycles.



Fig.S9 Cycling stability of NiOx-0, NiOx-0.05 and NiOx-0.50 films in 1 M PC/LiClO4 solutio.



Fig.S10 EPR spectra of NiO<sub>x</sub> NPs and Ar-NiO<sub>x</sub> NPs.



Fig.S11 Electrochromic properties of  $Ar-NiO_x$  films. (a) transmission spectra and (b) dynamic transmittance spectra.

Table S1 Values in Nyquist plots for different films at room temperature (25°C) and calculated results

Sample	$R_e(\Omega)$	$R_{ct}(\Omega)$	$C_{dl}(\Omega)$	$Z_w(\Omega)$	C <sub>l</sub> (Ω)	Result(Ω)
NiO <sub>x</sub> -0	12.68	65.45	5.88×10 <sup>-8</sup>	1.74×10 <sup>-2</sup>	5.28×10 <sup>-3</sup>	12.69
NiO <sub>x</sub> -0.05	10.35	51.90	8.44×10 <sup>-8</sup>	2.11×10 <sup>-2</sup>	4.37×10 <sup>-3</sup>	10.35
NiO <sub>x</sub> -0.25	15.87	63.44	6.42×10 <sup>-6</sup>	1.51×10 <sup>-2</sup>	4.26×10 <sup>-3</sup>	18.88
NiO <sub>x</sub> -0.50	18.52	66.94	8.29×10 <sup>-8</sup>	6.69×10 <sup>-3</sup>	4.14×10 <sup>-3</sup>	18.53

Table S2 Summary of the electrochromic properties of different films.

Films	Δ <b>Τ (%)</b>	t <sub>c</sub> (s)	t <sub>b</sub> (s)	CE (cm <sup>2</sup> /C)
NiO <sub>x</sub> -0	58.7	2.0	2.0	70.9
NiO <sub>x</sub> -0.05	63.1	1.7	1.5	83.8
NiO <sub>x</sub> -0.25	53.6	2.1	2.0	75.3
NiO <sub>x</sub> -0.50	46.2	2.1	1.9	69.0
NiO <sub>x</sub> -0.05 NiO <sub>x</sub> -0.25 NiO <sub>x</sub> -0.50	63.1 53.6 46.2	1.7 2.1 2.1	1.5 2.0 1.9	83.8 75.3 69.0



Fig.S12 Areal capacitance as a function of current density.

## **S13**

## Preparation of WO<sub>3</sub> electrochromic films

Highly transparent WO<sub>3</sub> films were prepared by a simple sol-gel method and spin-coating method, with Triton X-100 used as a surfactant to improve the switching speed of the films. Dissolve 1 g of WCl<sub>6</sub> powder in 20 ml of anhydrous ethanol and slowly add 2 ml of acetic acid, stirring mix thoroughly until the solution becomes pure black in colour; add 2ml of H<sub>2</sub>O<sub>2</sub>, at which point the solution turns pale yellow; Triton X-100 was added at a volume ratio of 1:50 and stirring was continued for 1 h to obtain the WO<sub>3</sub> precursor solution.

The WO<sub>3</sub> precursor solution was uniformly spin-coated onto the FTO substrate and annealed at 250°C for 30 min to obtain WO<sub>3</sub> electrochromic films. The FTO substrates were first treated in acetone, ethanol, DI water and a UV cleaner in sequence before use. The properties of the WO<sub>3</sub> films can be controlled by the number of spin coats, each of which needs to be individually annealed at 300°C for 30min. In order to obtain highly transparent WO<sub>3</sub> films, we spin-coated only three layers. The Fig.S11 shows the transmission spectra and dynamic transmittance spectra of the spin-coated 3-layer WO<sub>3</sub> film. The film exhibits >95% transmission of the bleached state and a fast switching speed (4.0s/0.8s), making it suitable for use in devices.



Fig. S13 Electrochromic properties of WO<sub>3</sub> films in 1M PC/LiClO<sub>4</sub> solution. (a) transmission spectra and (b) dynamic transmittance spectra.