Supporting Information

Developing Solid State Electrochromic-supercapacitor Prototype through DFT-guided Charge Transfer Engineering in 2D-Carbon Doped NiO

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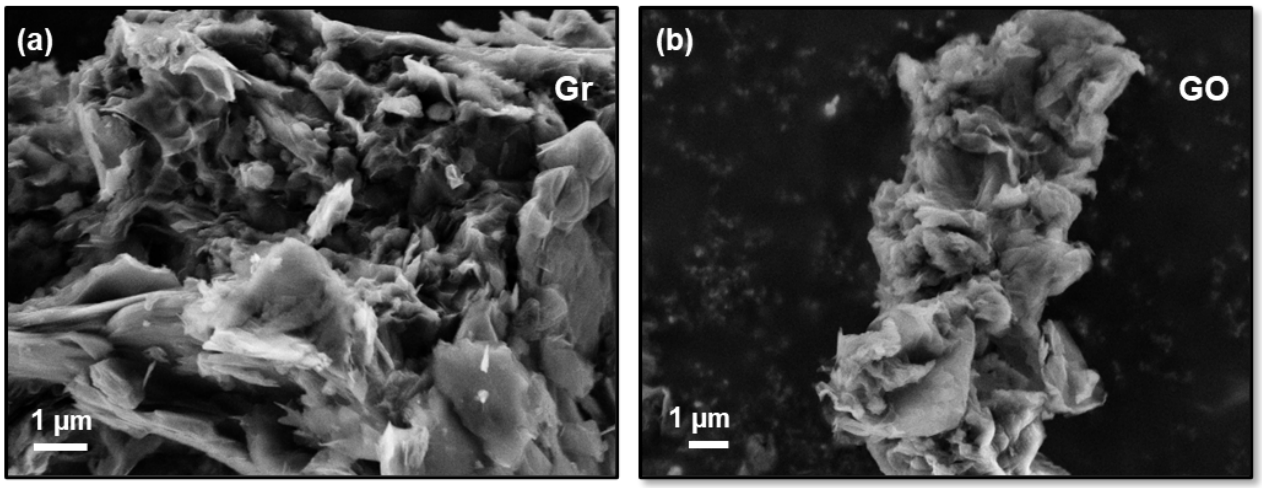

Figure S1: The FESEM micrographs of (a) Gr and GO (b).

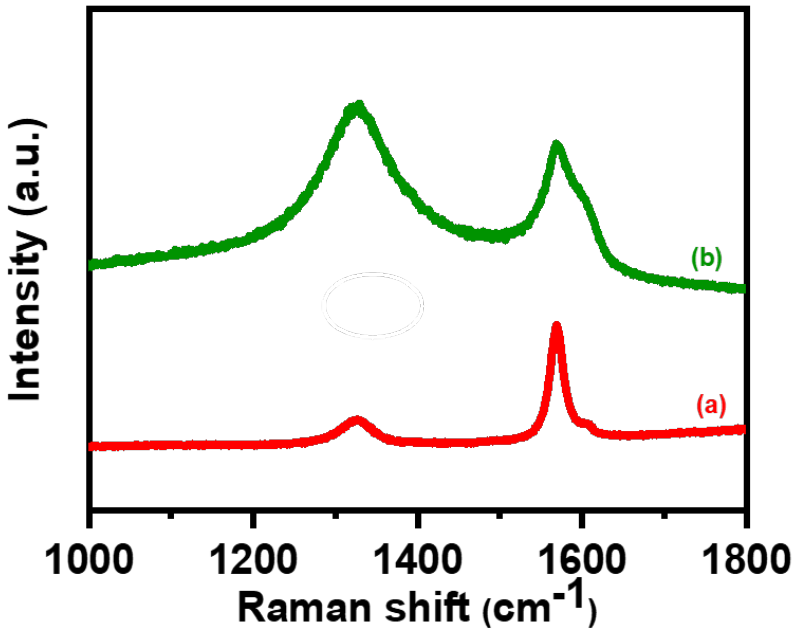

Figure S2: The Raman spectra of (a) Gr and GO (b).

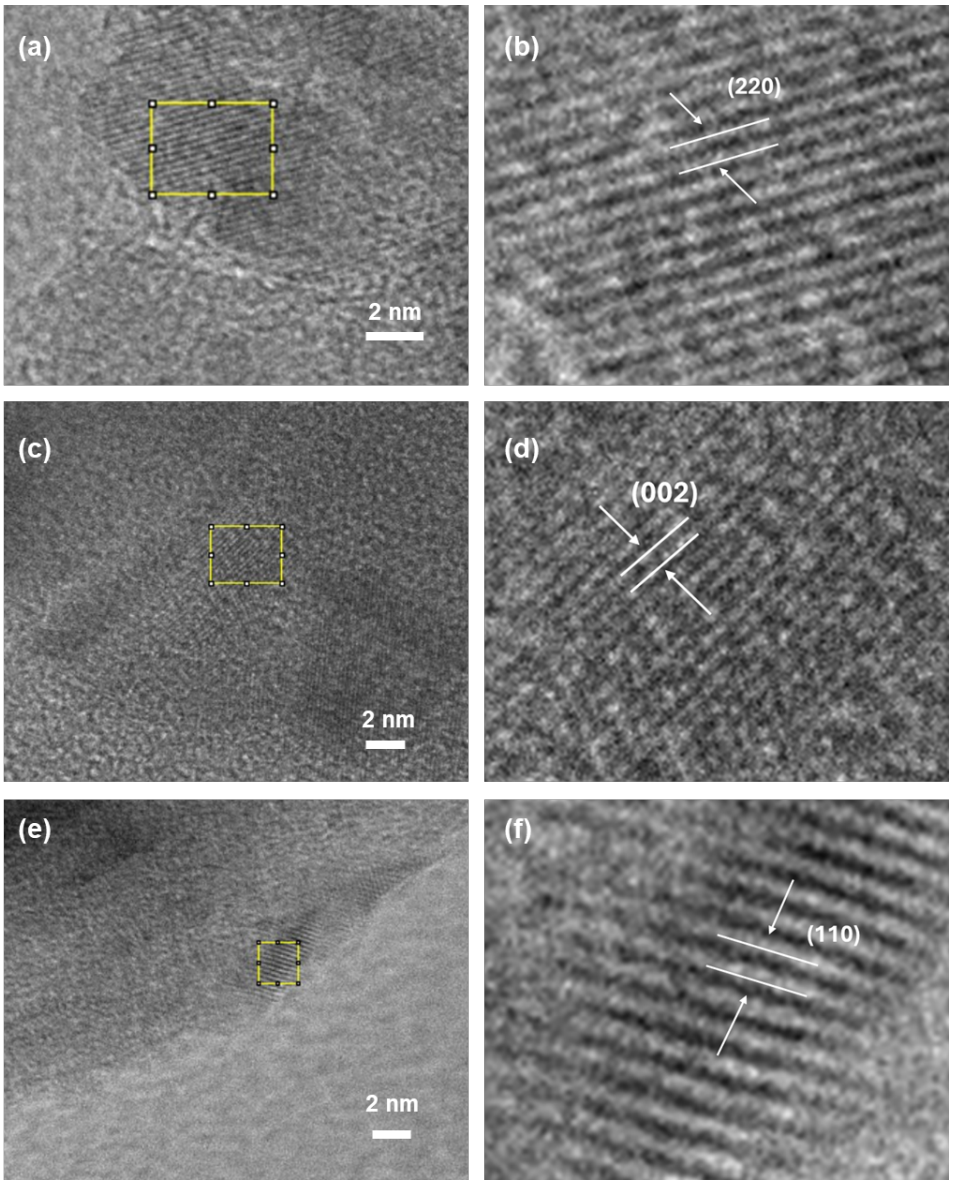

Figure S3: Higher magnification HRTEM images with lattice indexing (a, b) NiO, (c, d) Gr-NiO and GO-NiO (e, f).

Text S1: Electrochemistry of Electrodes and Devices

The electrochemistry of the electrodes was performed in three electrode configurations using 2M KOH as electrolyte. The electroactive electrodes namely NiO, Gr-NiO and GO-NiO as working electrode (W.E.), Ag/AgCl as reference electrode (R.E.) and Platinum wire as counter electrode (C.E.). The cyclic voltammetry was performed in the potential range of -0.1 to 0.7 V at various

scan rates of 5 to 50 mV/s. The electrochemical impedance spectroscopy was carried out in the frequency range of 10kHz to 10mHz at 0.35V. The Galvanostatic charging discharging was performed at various current densities varying from 0.2 mA/cm^2 to 1 mA/cm^2. The GCD profile of NiO and Gr-NiO was taken in broad potential range of 0 to 0.47 V. The GCD profile of GO-NiO electrode was taken in a potential range of 0 to 0.44 V.

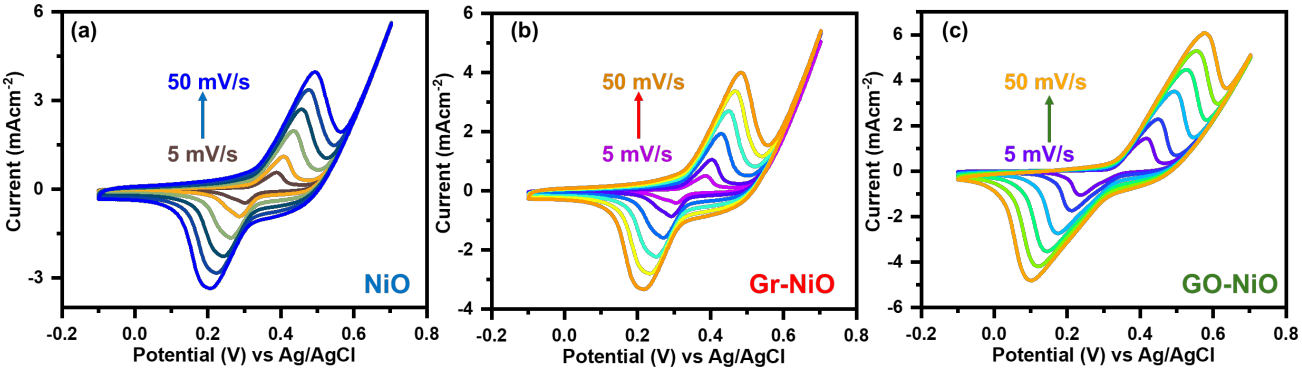

Figure S4: Scan rate dependent CV curve of electrodes (a) NiO, (b) Gr-NiO and GO-NiO (c).

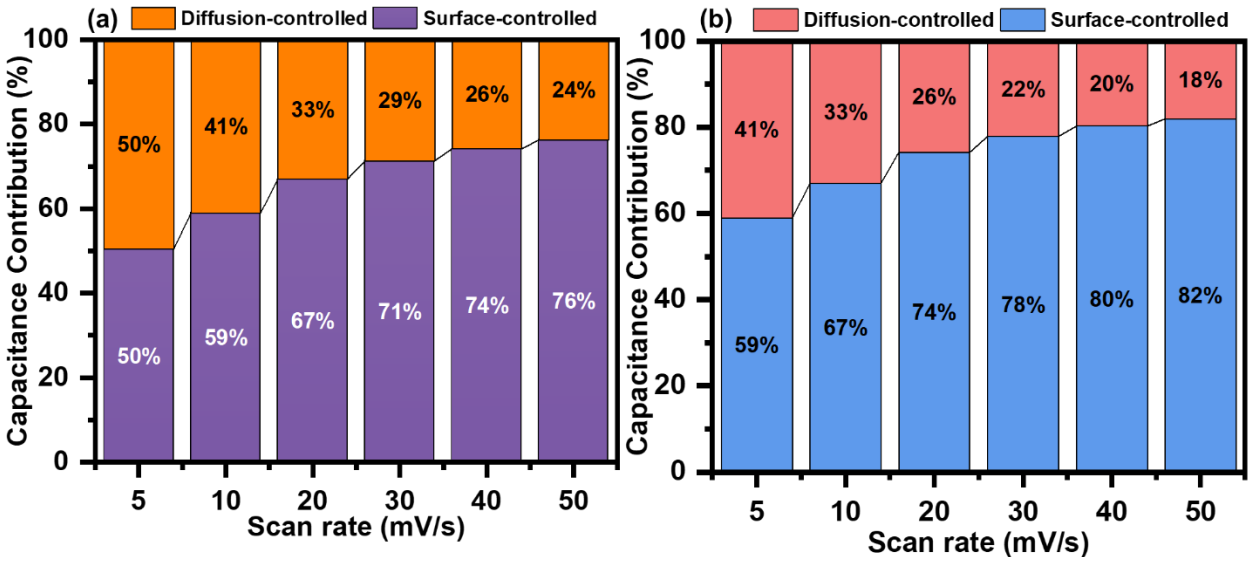

Figure S5: Capacitive contribution plot of electrodes (a) NiO and Gr-NiO (b).

Table S1: Collective values of solution resistance (R_s) and charge transfer resistance (R_{ct}) for all three electrodes.

Electrodes	R_s (Ω)	R_{ct} (Ω)
NiO	25	0.3
Gr-NiO	22.9	0.3
GO-NiO	45.8	2.1

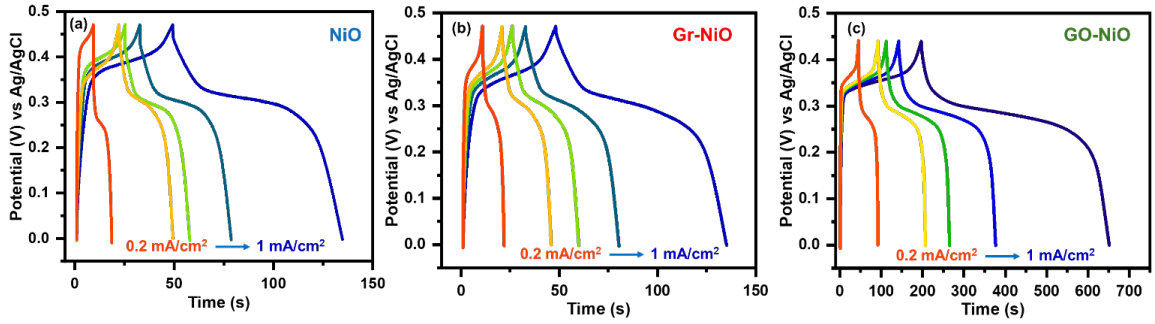

Figure S6: The GCD profile at various current densities (a) NiO, (b) Gr-NiO and GO-NiO (c).

Text S2: Electrochemical calculations

The specific capacitance for all the electrodes has been calculated using the Equation (S1)

$$Cs = \frac{I \times \Delta t_d}{A \times \Delta V}, \quad \text{(S1)}$$

where I/A repersents the current density (mA/cm^2), Δt_d is the discharging time of the electrode and ΔV is the potential window taken to perform the GCD.

The cyclic stability of the electrodes NiO, Gr-NiO and Go-NiO were recorded at 1mA/cm^2, 1mA/cm^2 and 3mA/cm^2. The retention of the electrode was calculated using Equation (S2) to check its perfromance for real life applications.

$$Retention = \frac{Cs\ value\ of\ n^{th}\ cycle}{Cs\ value\ of\ 1^{st}\ cycle} \times 100\%, \quad \text{(S2)}$$

The specific capacitance value from the CV curve has been calculated using the Equation (s3).

$$Cs\ (from\ CV\ curve, mF/cm^2) = \frac{Area\ under\ the\ CV\ curve}{2 \times v \times \Delta V \times active\ area\ of\ the\ device}, \quad \text{(S3)}$$

here v and ΔV denotes the scan rate and potential window, respectively.

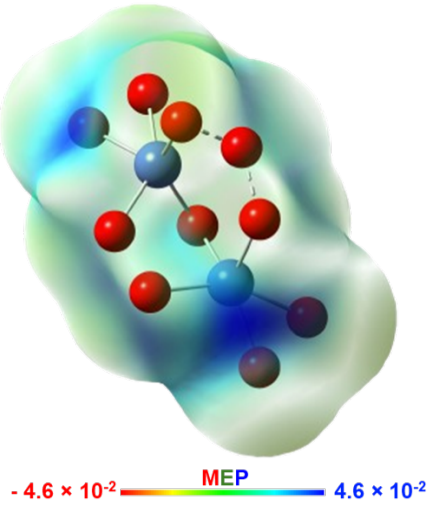

Figure S7: MEP plot of NiO.

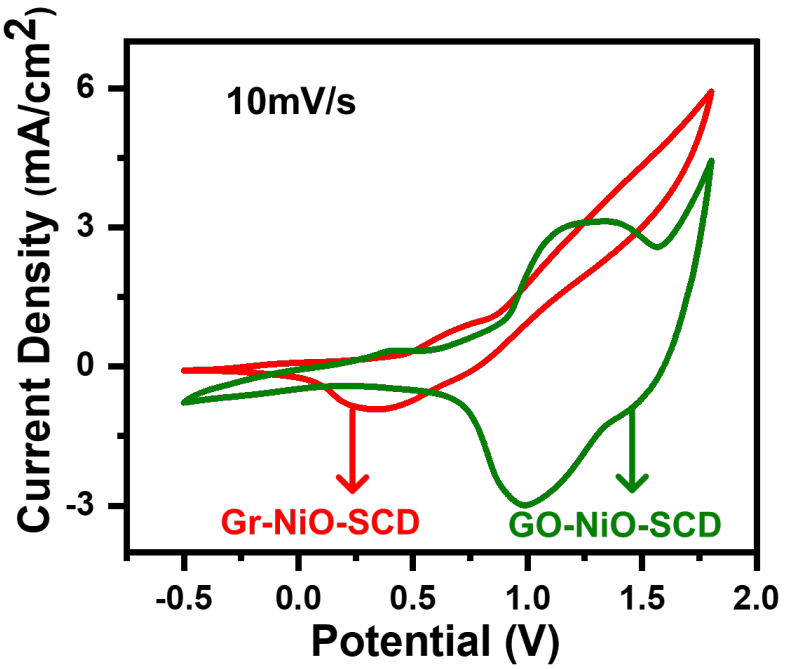

Figure S8: Comparative CV curve of Gr-NiO-SCD and GO-NiO-SCD.

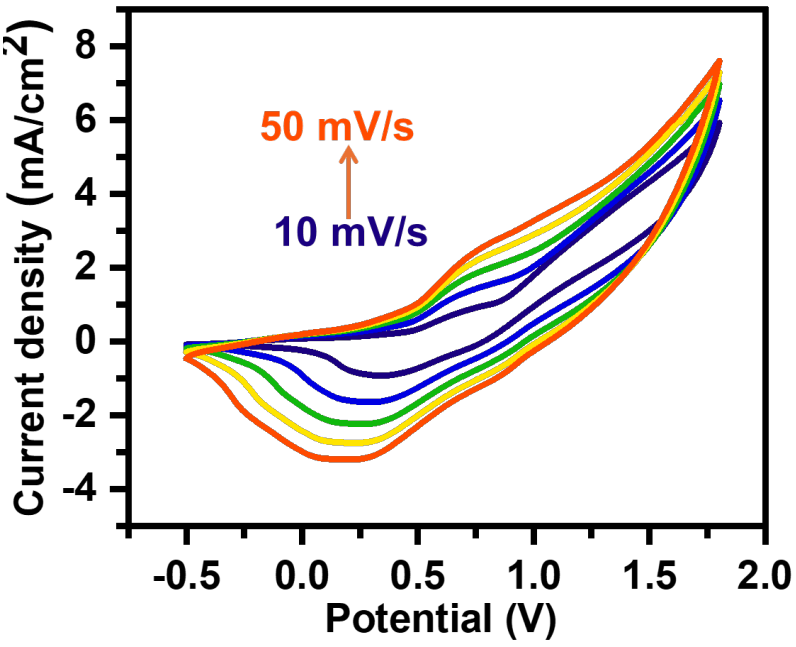

Figure S9: The scan rate dependent CV curve of the Gr-NiO-SCD.

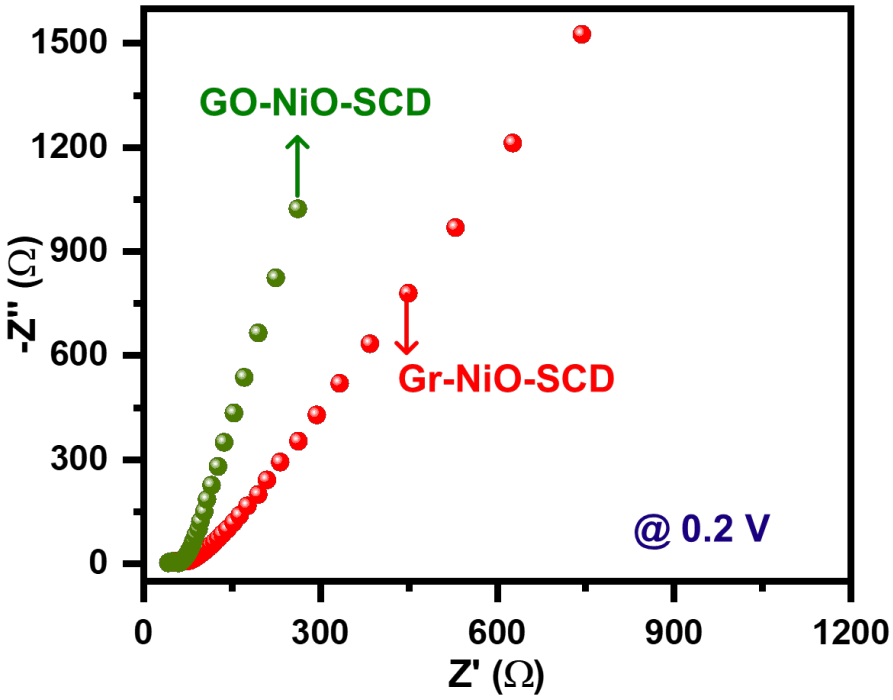

Figure S10: Comparative Nyquist plot of Gr-NiO-SCD and GO-NiO-SCD recorded at 0.2V

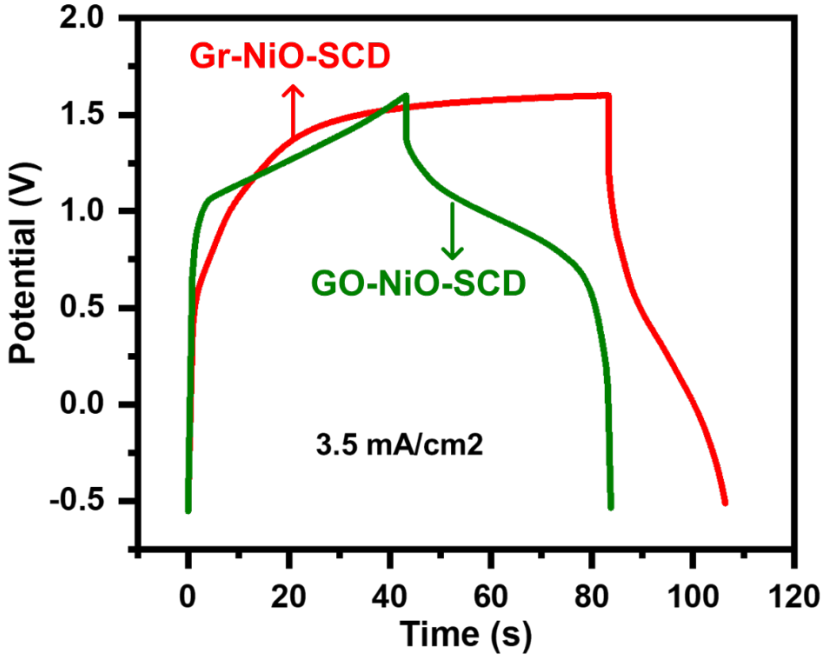

Figure S11: Comparative GCD profile of Gr-NiO-SCD and GO-NiO-SCD recorded at 3.5 mA/cm^2.

Table S2: Comparison table of NiO based supercapacitor devices.

S.No.	Device	Specific Capacitance (mF/cm^2)	Current density (mA/cm^2)	References.
1.	Co-doped NiO Device	10.8	0.4	Xue et al. [1]
2.	NiO/Ag NWs/Viologen	35.5	2.5	Mysoon et al. [2]
3.	Cu-doped NiO Device	14.9	0.1	Kim et al. [3]
4.	Fe2O3/ rGO-NiO	24.1	0.5	Xue et al. [4]
5.	**GO-NiO-SCD**	**103.7**	**3.5**	**This work.**

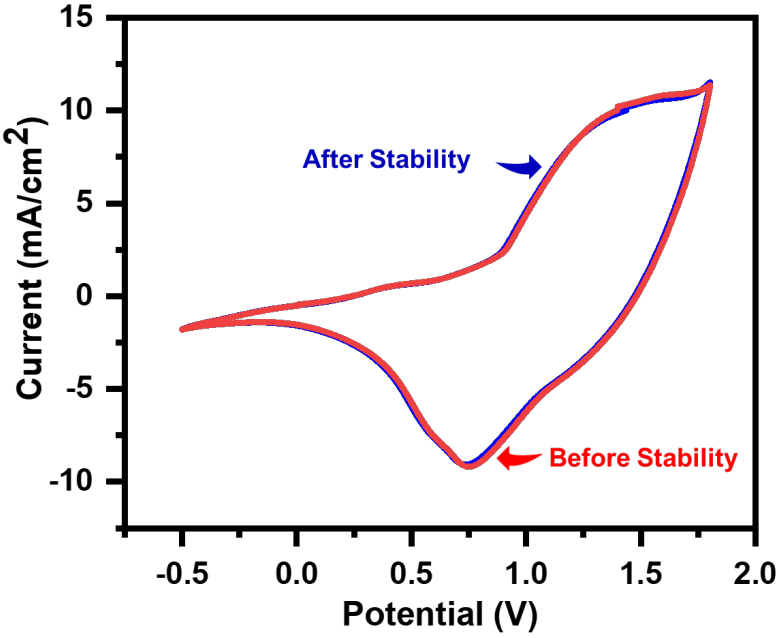

Figure S12: The CV curve of the GO-NiO-SCD before and after stability measurements.

Text S3: Optical study of the electrodes

The optical study of all the three electrodes is conducted in three electrode configurations consisting of Ag/AgCl and Platinum wire as R.E. and C.E. respectively. The electroactive electrodes (NiO, Gr-NiO and GO-NiO) were used as W.E. The in-situ bias dependent transmittance spectra for NiO were recorded at various potentials (-0.5V to 1V) in the wavelength range of 400 to 1100 nm.

The color contrast (CC) of the electrodes was evaluated using the Equation S4.

$$CC = \frac{T_{Off} - T_{On}}{T_{Off}} \times 100\%, \quad \text{(S4)}$$

here T_{Off} and T_{On} represents the Off (-0.5V) and ON (1V) state transmittance value of the electrodes.

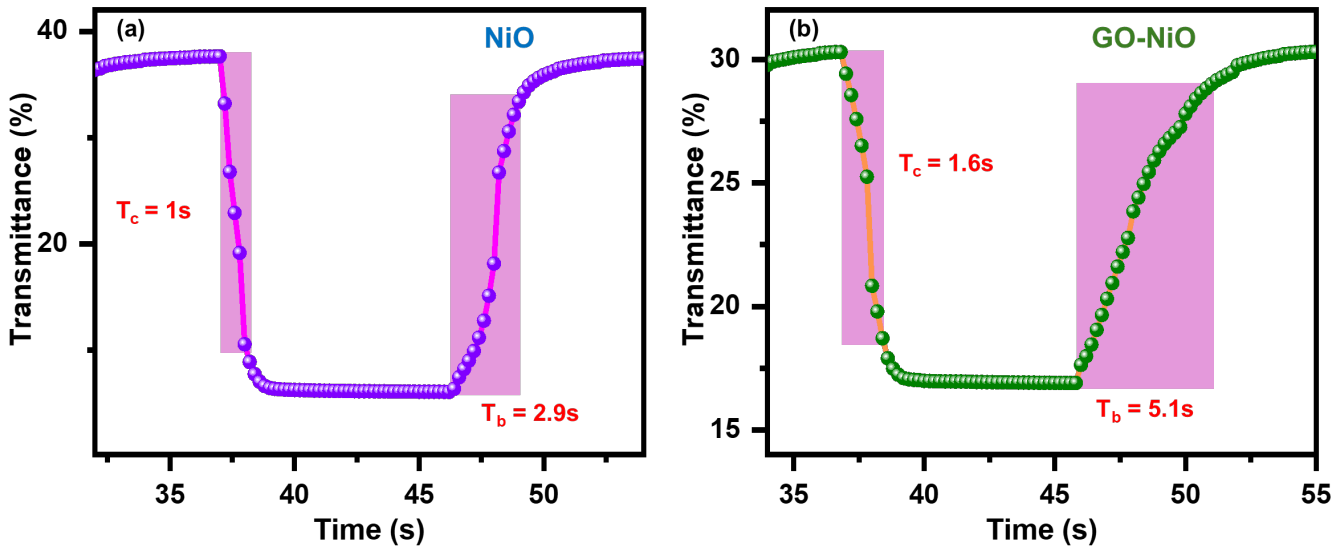

Figure S13: The single switching cycle of the (a) NiO and GO-NiO electrodes (b).

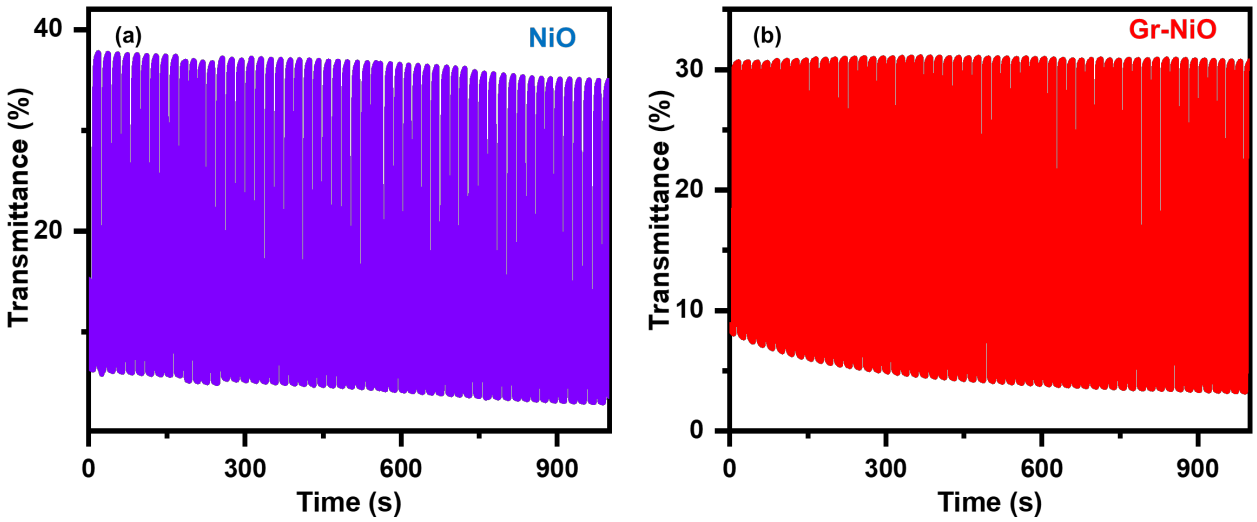

Figure S14: The switching stability of the (a) NiO and Gr-NiO electrodes (b).

Table S3: The electrochemical and optical properties of electrodes and devices.

Electrode/ Device	Redox potential @10mV/s		Resistance		Specific capacitance	Switching time T_c/ T_b	Color contrast
	Oxidation	Reduction	Solution	Charge transfer			
NiO	0.28V	0.4V	25 Ω	0.3 Ω	38 mF/cm^2 @ 0.2mA/cm^2	1/2.9 s	77.4%
Gr-NiO	0.28V	0.4V	22.9 Ω	0.3 Ω	38.5 mF/cm^2 @ 0.2mA/cm^2	0.8/0.9 s	68.2%
GO-NiO	0.21V	0.44V	45.8 Ω	2.1 Ω	210.8 mF/cm^2 @ 0.2mA/cm^2	1.6/5.1 s	37.4%
GO-NiO-SCD	1.3 V	1V	42 Ω	17 Ω	103.7 mF/cm^2 @ 3.5 mA/cm^2	NA	NA

References

[1] J. Xue, W. Li, Y. Song, Y. Li, and J. Zhao, Visualization electrochromic-supercapacitor device based on porous Co doped NiO films, Journal of Alloys and Compounds **857**, 158087 (2021).

[2] R. Mysoon, M. Ojha, D. Maity, M. Pal, and M. Deepa, Switchable electrochromic supercapacitor with vivid colors using a phenolic-viologen paired with an Ag nanowires/NiO film, Journal of Energy Storage **114**, 115747 (2025).

[3] S. Y. Kim, T. Y. Yun, K. S. Yu, and H. C. Moon, Reliable, High-Performance Electrochromic Supercapacitors Based on Metal-Doped Nickel Oxide, ACS Appl. Mater. Interfaces **12**, 51978 (2020).

[4] J. Xue, H. Xu, S. Wang, T. Hao, Y. Yang, X. Zhang, Y. Song, Y. Li, and J. Zhao, Design and synthesis of 2D rGO/NiO heterostructure composites for high-performance electrochromic energy storage, Applied Surface Science **565**, 150512 (2021).