Supporting Information

Fabrication of Symmetric Supercapacitor Device using MnO_2/Cellulose nanocrystals/Graphite electrode by Sputtering for Energy Storage

Nitesh Choudhary,[a,b] Akshay Tomar,[b] Shiva Singh,[a] Ramesh Chandra,[b] Pradip K. Maji[*a]

[a] *Department of Polymer and Process Engineering, Indian Institute of Technology Roorkee, Saharanpur Campus, Saharanpur- 247001, India*

[b] *Institute Instrumentation Centre, Indian Institute of Technology Roorkee, Roorkee-247667, India*

Corresponding author:* ***Pradip K. Maji*

Email: pradip@pe.iitr.ac.in; Contact No.: +91-7895965010

Supporting Information

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Table S1 *The sputtering parameters used for the deposition of highly ordered MnO_2 thin films.*

Sputtering parameters	
Target Material	Manganese (Mn)
Substrate	n-silicon (100)/Graphite sheet
Base pressure	3 μTorr
Working pressure	~8 mTorr
Deposition power	70W (RF)
Deposition time	60 minutes
Substrate temperature	Room temperature (RT) and Temperature variation
Target-substrate distance	5.5 cm
Gas used	Ar:O_2::20:20 (sccm)

Supplementary Section S1: Characterization details: Calculation of electrochemical parameters

S1.1 Electrodes in three-electrode configuration.

The areal capacitance of the electrode,

$$C_a = \frac{2 \times I \times dt}{V \times A} \tag{S1}$$

Where C_a is the areal capacitance, td is the discharge time, I is the current density, V is the voltage window and A is the area of the electrode.

The energy density (E) and power density (P) were calculated using equation (S2 and S3) [1].

$$Energy\ density = \frac{C_a \times \triangle V^2}{2 \times 3600} \times 1000 \tag{S2}$$

$$Power\ density = \frac{E \times 3600}{\triangle t} \tag{S3}$$

Where E and P represent the areal energy ($Whcm^{-2}$) and power density (Wcm^{-2}) of the electrode, V is the voltage window, C_a represents the areal capacitance, and $\triangle t$ is the discharging time.

All the calculated electrochemical results from the CV/GCD curve are tabulated in the tables.

The amount of material deposited on the graphite sheet was calculated by measuring the weight of the graphite sheet film before and after the CNC and MnO_2 deposition. The weight percentage of MnO_2 ($W_{MnO2}\%$) was estimated using the following equation:

Mass loss:

$$W_{MACNC\%} = \frac{W_f - W_i}{W_f} \times 100\% \qquad (S4)$$

Where W_i is the initial weight of the graphite sheet, and W_f is the final weight of the graphite sheet after CNCs/MnO_2 deposition.

$$W_{CNCs/MnO2} = w_f - w_i \qquad (S5)$$

$$CNCs/MnO2 = 0.002g$$

Hence, the deposition of on the surface of the graphite sheet is 2 mg.

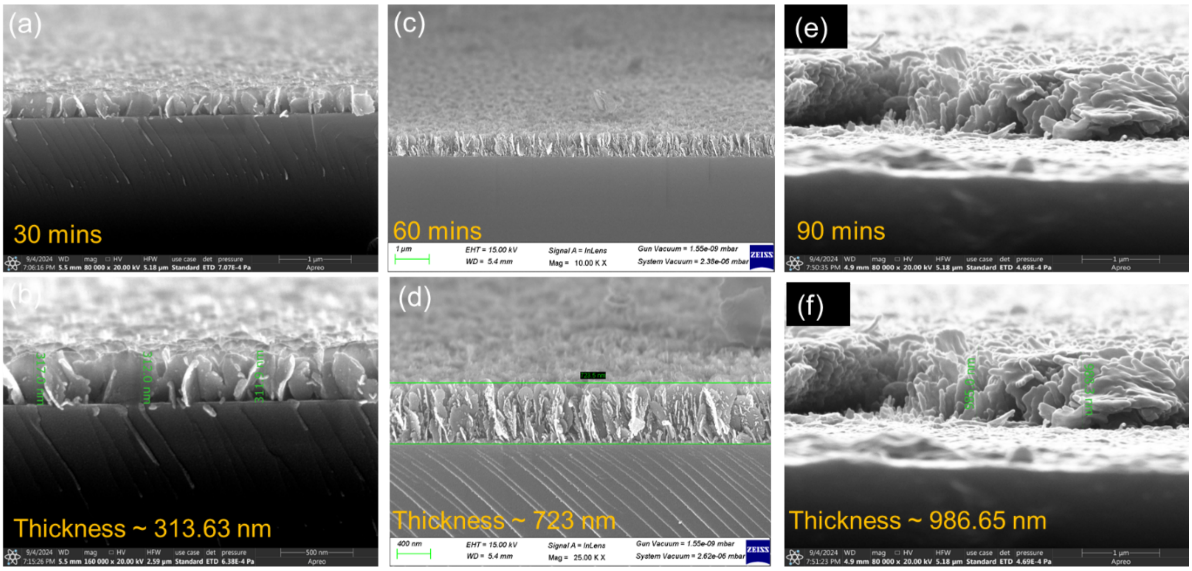

Fig. S1 *Cross-sectional view of MnO_2 thin film (a-b) at 30 minutes (c-d) at 60 minutes, and (e-f) at 90 minutes.*

Table S2 *shows the prepared electrode materials' Roughness, Skewness (Rsk), and Kurtosis values (Rku).*

S.No	Name of the electrode	Parameters	Values
1.	Graphite	Roughness	R_{max} 243 nm, 37.7 nm
		Skewness (R_{sk})	-0.352
		Kurtosis value (R_{ku})	2.96
2.	CNCS/silicon	Roughness	R_{max} 120 nm, 19.2 nm
		Skewness (R_{sk})	-0.175
		Kurtosis value (R_{ku})	2.58
3.	CNCS/Graphite	Roughness	R_{max} 224 nm, Rq 34.1 nm
		Skewness (R_{sk})	-0.304
		Kurtosis value (R_{ku})	2.78
4.	MnO_2/CNCS/Graphite	Roughness	R_{max} 427 nm, Rq 53.0 nm
		Skewness (R_{sk})	0.0955
		Kurtosis value (R_{ku})	3.44

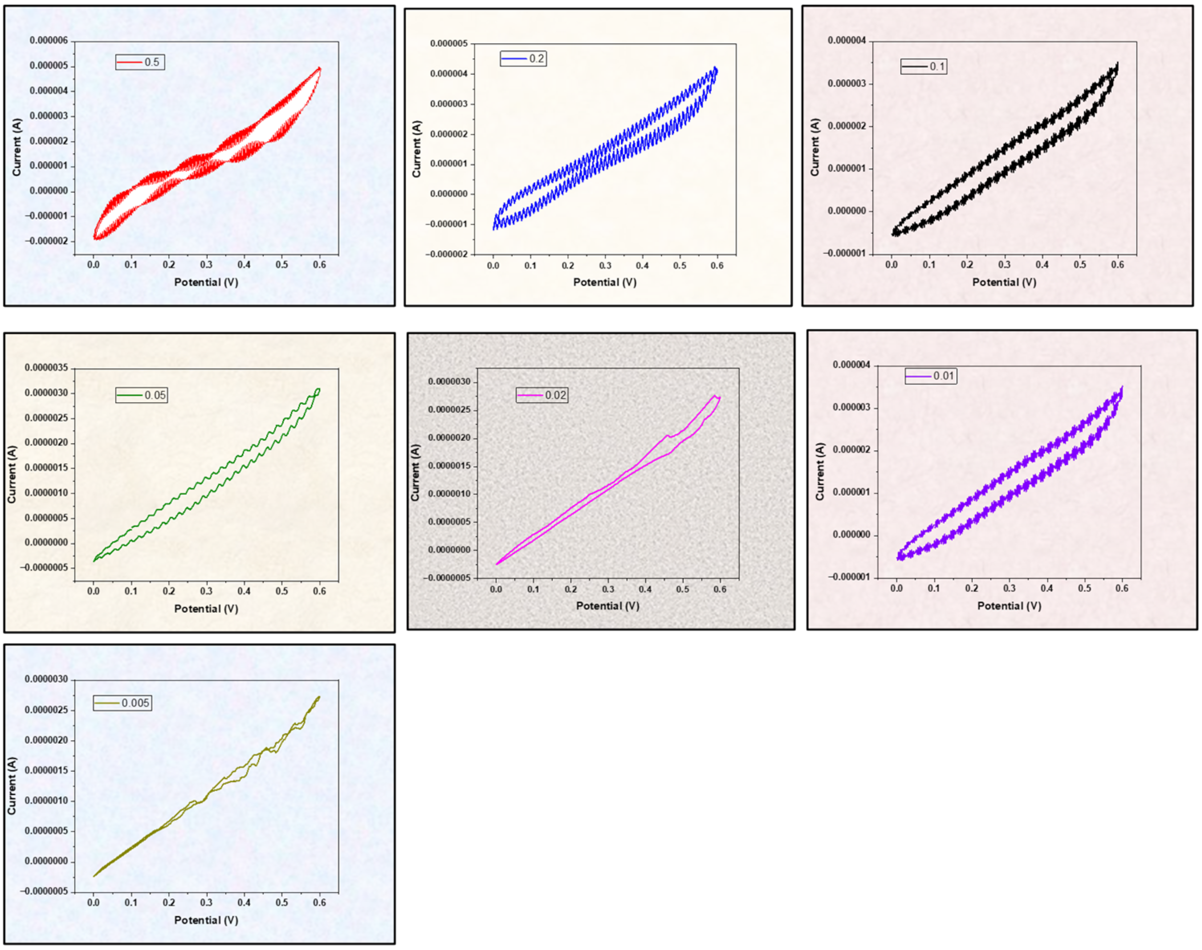

Fig. S2 *shows the CV of cellulose nanocrystals (CNCs) on the silicon substrate at different scan rates which ranges from 0.5 to 0.005 V/s.*

Table S3 *Calculated areal capacitance at various scan rates with energy density and integrated area from the CV curve of the CNCs/Silicon electrode.*

Scan rate (V/s) CNCs	Area	Volatge window	Areal capacitance (mF/cm^2)
0.5	4.26014E-7	0.6	0.00142
0.2	3.46192E-7	0.6	0.00288
0.1	2.83334E-7	0.6	0.00472
0.05	1.63674E-7	0.6	0.00546
0.02	8.47866E-8	0.6	0.00707

0.01	2.83334E-7	0.6	0.04722
0.005	2.67289E-8	0.6	0.00891

Table S4 *Calculated areal capacitance at various scan rates with energy density and integrated area from the CV curve of the CNCs/Graphite electrode.*

Scan rate (CNCs/Graphite)	Area	Voltage window	Areal capacitance (mF/cm^2)
0.5	0.00239	0.8	7.46059
0.2	0.00178	0.8	13.86841
0.1	0.00142	0.8	22.22567
0.05	0.0012	0.8	37.51883
0.02	0.00103	0.8	80.108

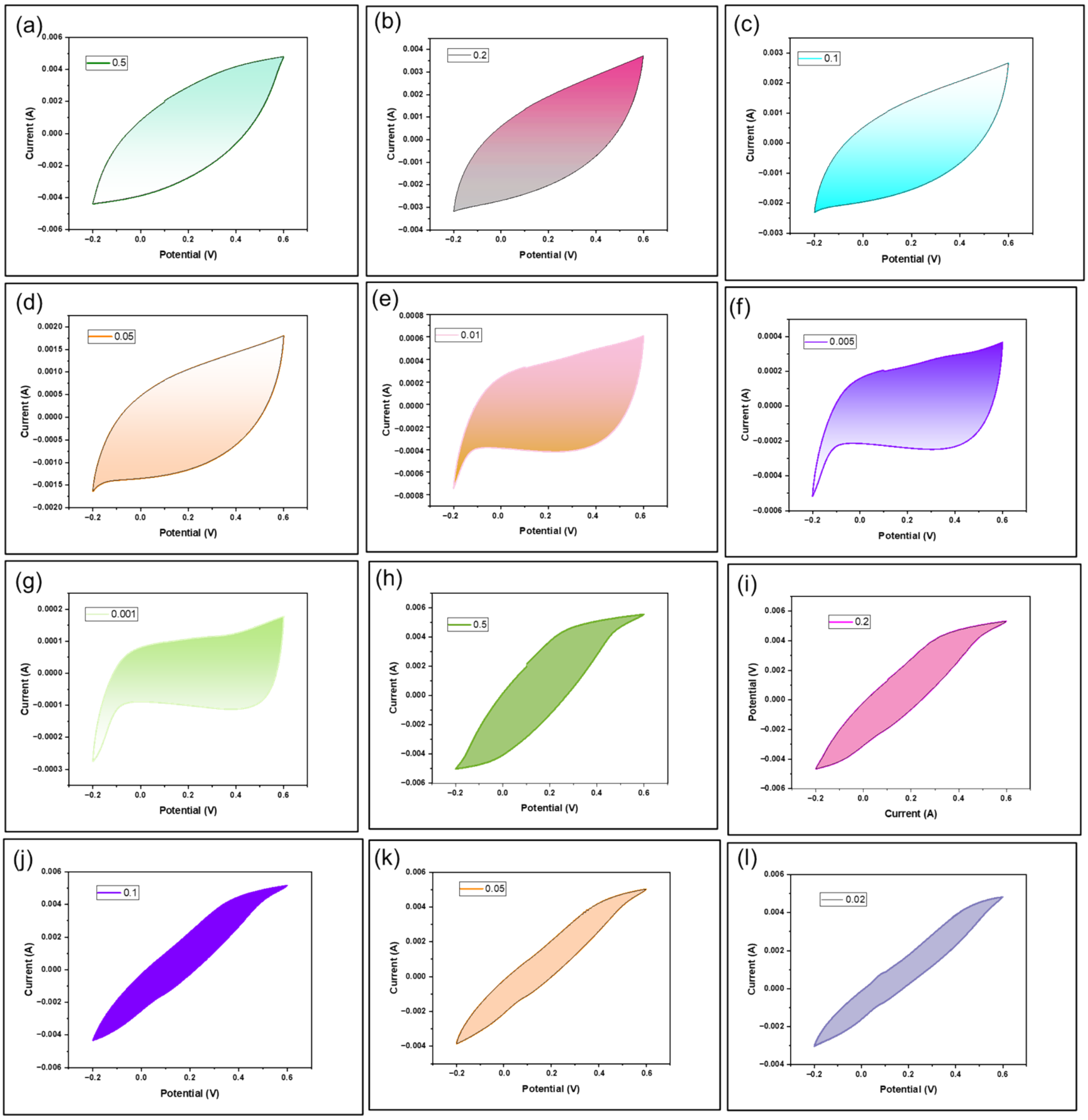

Fig. S3 *shows (a-g) the CV of pristine Graphite (h-l) CV of CNCs/Graphite at different scan rates spanning from 0.5 to 0.005 V/s.*

Table S5 *Calculated areal capacitance at various scan rates with energy density and integrated area from the CV curve of the MnO_2 on CNCs/Graphite (20:20, 1h, 70W, 7.9mTorr, 75^0C) electrode.*

Scan rate	Area	Voltage window (-0.2 to +1 i.e., 1.2V)	Areal capacitance (mF/cm^2)

0.005	7.05455E-4	1.2	97.97988
0.5	0.00576	1.2	7.99858
0.2	0.00466	1.2	16.17431
0.1	0.00361	1.2	25.05605
0.05	0.00272	1.2	37.81669
0.02	0.00169	1.2	58.53999
0.01	0.0011	1.2	76.15694
0.005	7.05455E-4	1.2	97.97988
0.002	3.75121E-4	1.2	130.25027

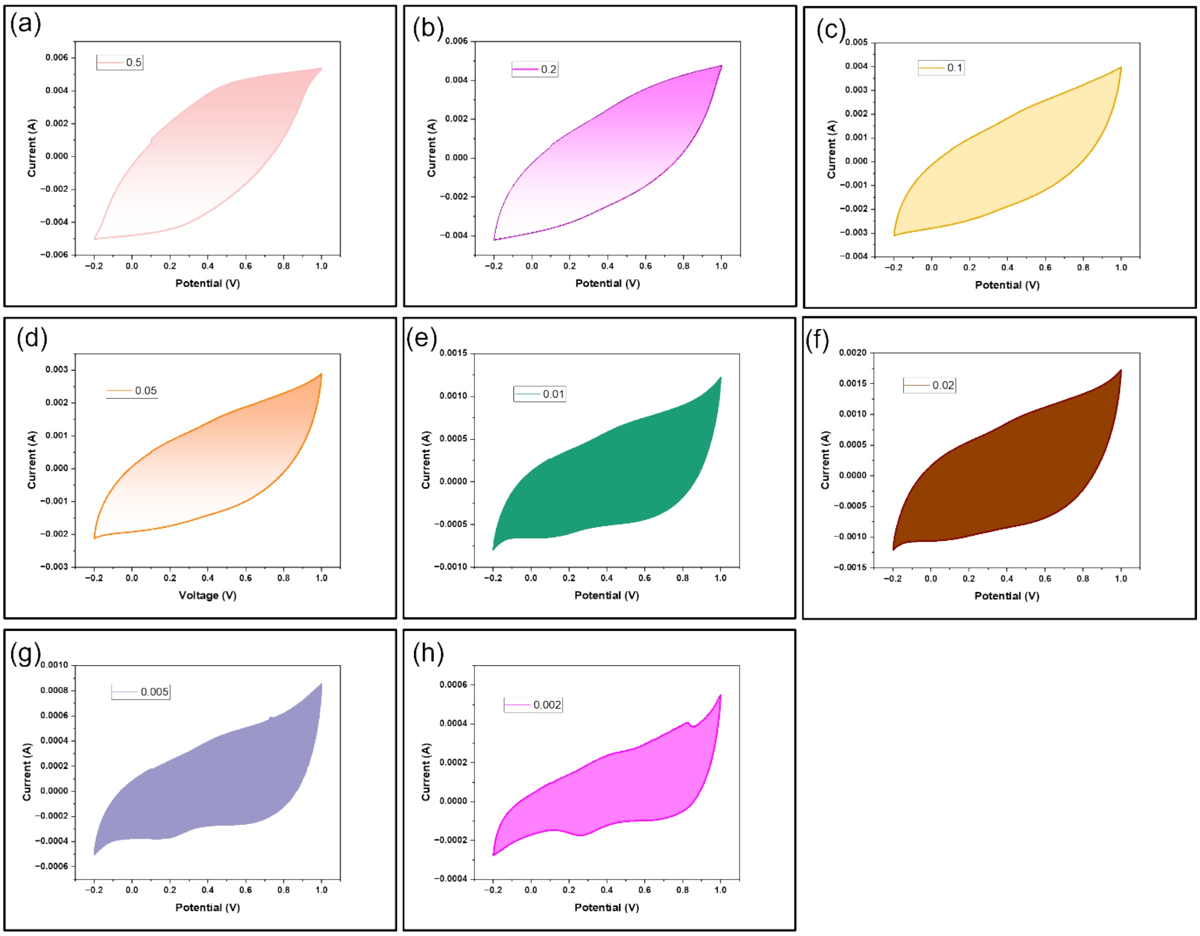

Fig. S4 *Cyclic voltammetry curves of MnO_2/CNCs/Graphite-75^{0}C at various scan rates.*

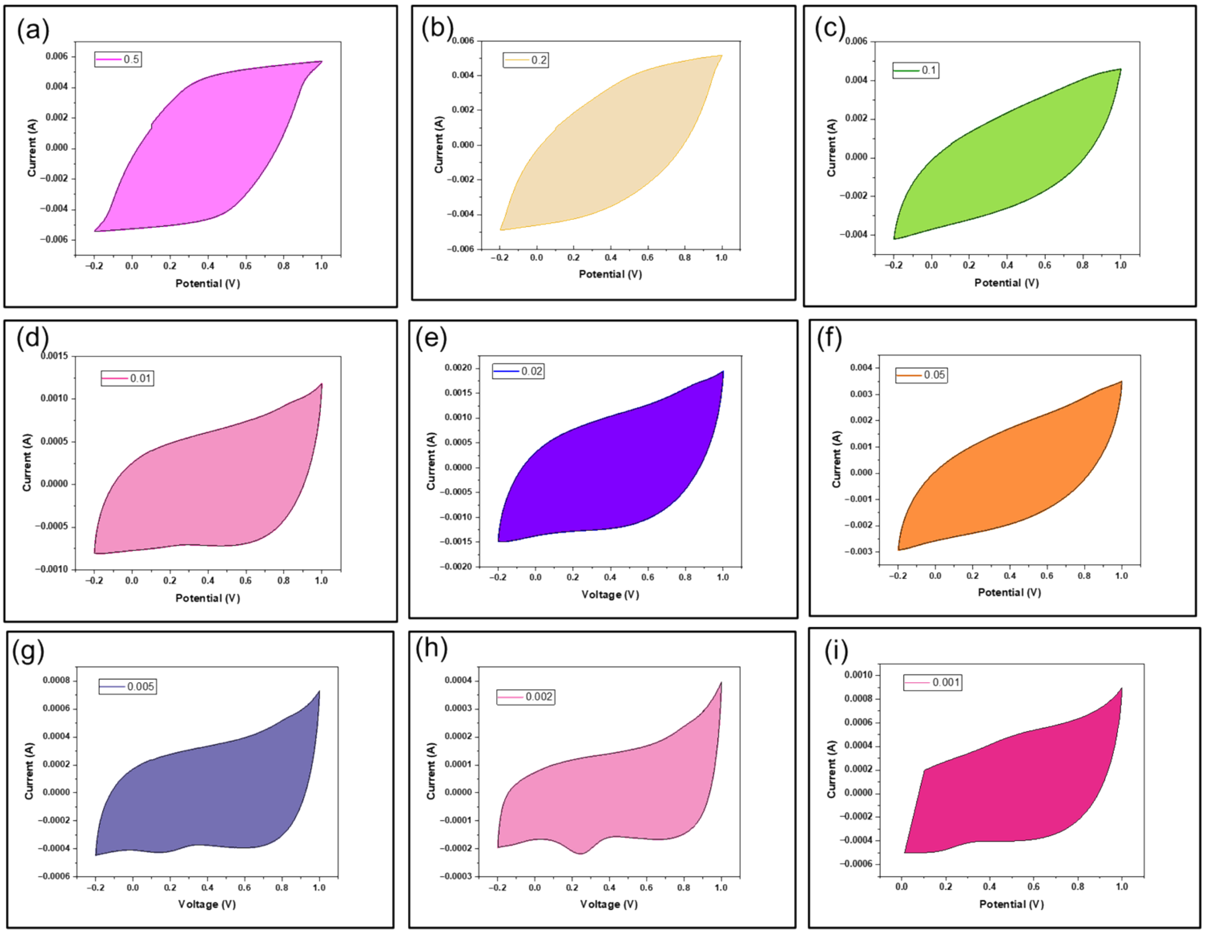

Fig. S5 *Cyclic voltammetry of MnO_2/CNC/Graphite@150^{0}C and @250^{0}C at various scan rates (0.5, 0.2, 0.1, 0.01, 0.02, 0.05, 0.005, 0.002, 0.001 V/s).*

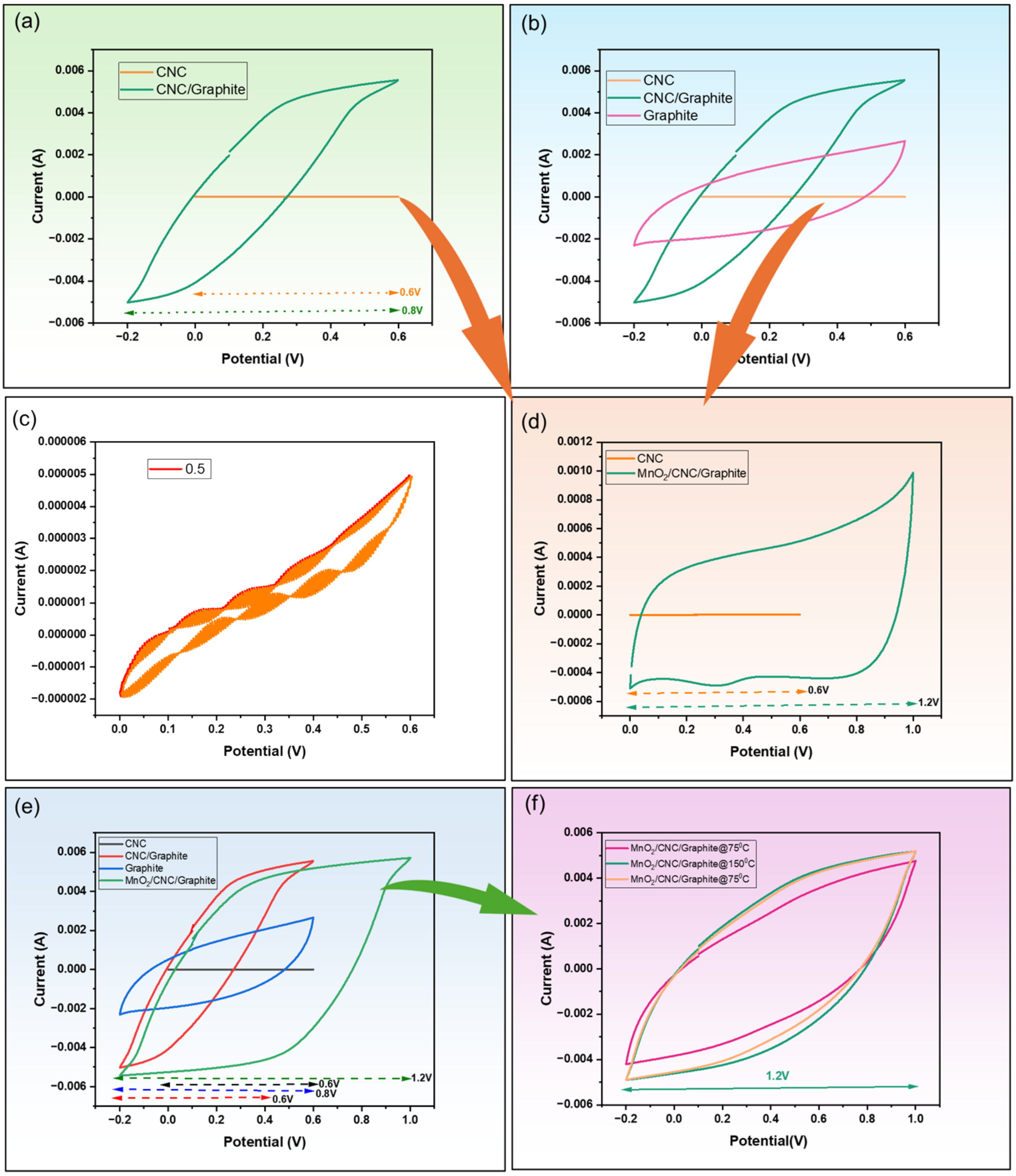

Fig. S6 *shows the comparative graph (a) the CV of CNCs, CNCS/Graphite, (b)the CV of CNCs, CNCS/Graphite, and Graphite, (c) shows CV of only CNCs, (d) CV curve of CNCs and composite, (e) Combine CV curve, and (f) the CV of MnO2/CNCs/Graphite at different temperatures.*

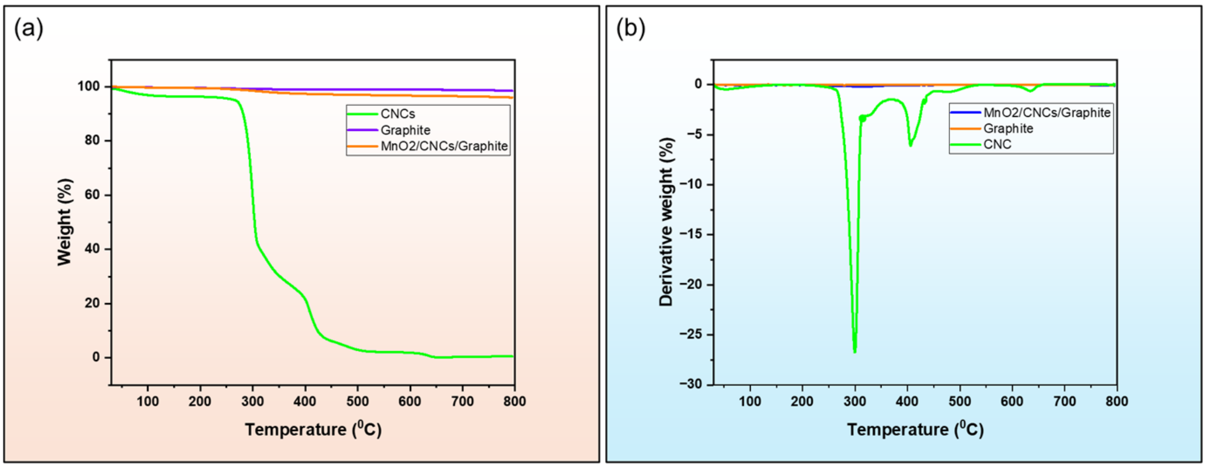

***Fig. S7** Thermal analysis of CNCs, Graphite, and MnO_2/CNCs/Graphite (a) Thermogravimetry graph and, (b) Differential thermal analysis graph.*

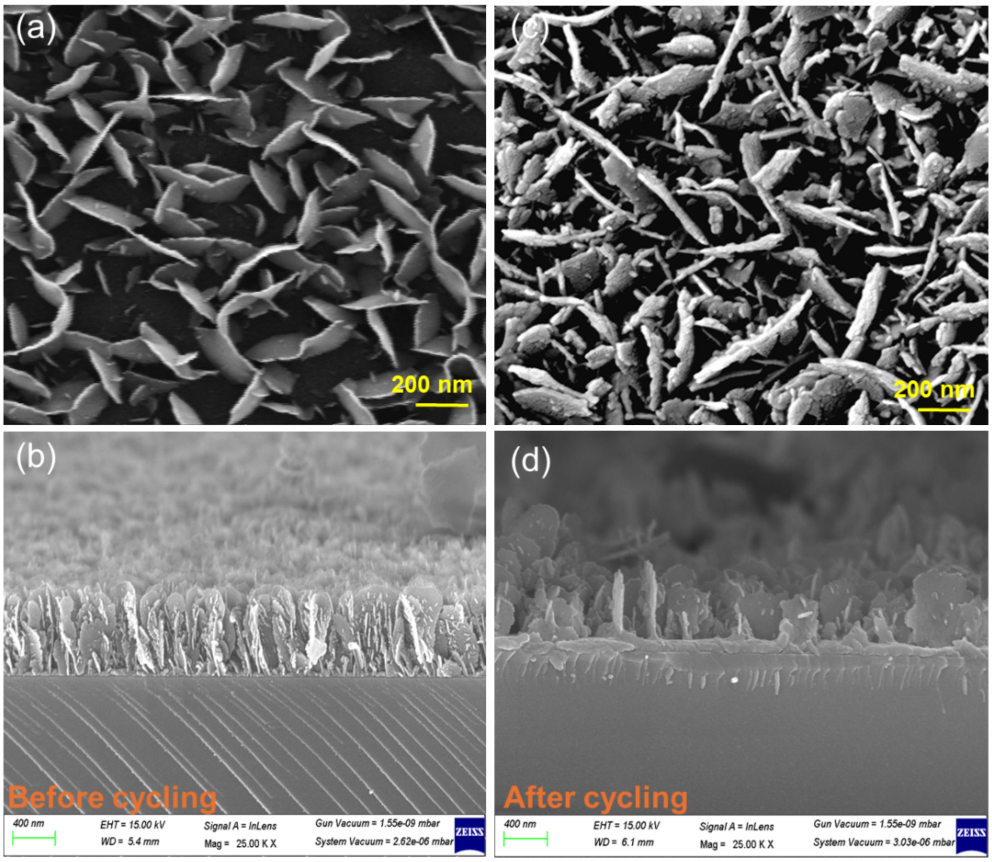

***Fig. S8** (a) Before cycling FESEM image, (b) Cross-section image before cycling, (c) After Cycling FESEM image, and (d) EDX image after cycling of symmetric supercapacitor device.*

Table S6 *Comparison of electrode materials used, areal specific capacitance, energy density, power density, scan rate or current denisties with cyclic efficiency after several cycles with the present study and other reported materials.*

Electrode materials used	**Areal capacitance (mF/cm^2)**	**Energy density (mWh/cm^2)**	**Power density (µW/cm^2)**	**Current density or scan rate**	**Cyclic efficiency(%)**	**Cycles**	**Reference**
Graphene/CNT/BC	495	0.0328	1200	35 mA/cm^2	86	5000	[2]
CNFs/Mxene/PC film (KOH)	143	-	17.5	0.1 mA/cm^2	90	10,000	[3]
Graphene-cellulose H_2SO_4	80	9	-	80 mA/cm^2	-	-	[4]
Expanded graphite foil (EGF)	65	0.163	1900	20 mA/cm^2	95	10,000	[5]
CNF/Mxene (H_2SO_4)	25.3	80	145	2 mV/s	-	-	[6]
Assymetric (MnO_2@CFCBSCA)	130	-	-	2.3 mA/cm^2	80.8	10,000	[7]
MnO_2/Cotton-derived carbon cloth	202	30.1 mu W h/cm^2	-	0.15 mW/cm^2	87.7	5000	[8]
polypyrrole (PPy)-coated cellulose paper	58	-	600	2 mA cm^2	91	10,000	[9]
Wheat straw cellulosic biochar 1M KOH	0.3	-	-	0.5 A/g	-	-	[10]
Cellulosic biomass/PAN membrane	11.5	-	-	0.5 mA/cm^2	92	5000	[11]
CNF/CNT/RGO carbon aerogels	109.4	-	-	-	83.8	100	[2]
Mno_2/CNCs/Graphite 1M Na_2SO_4 1.2V, Voltage window	148.9	20.69	2677	2 mV/s	85.27	15,000	**Present work**

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