Electronic Supplementary Material (ESI) for Sustainable Energy & Fuels.

Supporting Information

A bottom-up fabrication for Sulphur (S), Nitrogen (N) co-Doped two-dimensional Microporous Carbon Nano-sheets for high-performance Supercapacitor and H_2, CO_2 storage

Prajnashree Panda [a,b,c] and Sudip Barman[a,b,c]*

[a]School of Chemical Sciences, National Institute of Science Education and Research (NISER), HBNI, Bhubaneswar, Orissa-752050, India, Tel.: +91 6742494183,

[b]Homi Bhabha National Institute, Training School Complex, Anushakti Nagar, Mumbai 400094, India

[c]Centre for Interdisciplinary Sciences, National Institute of Science Education and Research (NISER), Bhubaneswar, Orissa-752050, India

E-mail address: sbarman@niser.ac.in.

Materials

Formamide was purchased from Merck, India. Thiourea was purchased from Spectrochem PVT. LTD. Mumbai (India). Potassium Hydroxide (KOH), Hydrochloric acid (HCl), and Sulphuric acid (H_2SO_4) were purchased from CDH chemicals, India. PTFE binder was purchased from MTI corporation. Deionized water was used to carry out all experiments.

Material Characterization

The powder x-ray diffraction patterns (p-XRD) of samples were measured by Bruker X-ray diffractometer (DAVINCI D8 ADVANCE equipped with Cu Kα source of wavelength 0.154 nm). The morphological characterization was investigated by a Field-emission scanning electron microscope (FESEM) system (Model-∑igma, Carl Zeiss, Germany) and Transmission Electron Microscopy (TEM) instrument operated at 200 kV (F200, JEOL). XPS measurement was done using AXIS ULTRA (Kratos) instrument where a monochromated Al-kα source was used. XPS was taken from the sample deposited on the silicon wafer. The CO_2 and N_2 physisorption isotherms were collected using AUTOSORB-1 (Quantachrome). The pore size distribution (PSD) was calculated using the nonlocal density functional theory (NLDFT) model while the micropore analysis was carried out using the t-plot method. LabRAM HR Evolution, Horiba Scientific, and Raman Spectrometer were used for Raman analysis using a 532 nm laser source. Electrochemical measurements were performed using CS310 Electrochemical Workstation (Corrtest Instruments).

Electrochemical Characterization

Fabrication of electrodes was carried out by mixing PTFE binder, conductive carbon, and p-CNS-X with a mass ratio of 10:10:80 (in ethanol) followed by coating over Nickel foam (1*1 cm^2) and Ti foil (current collector) for the base and acid medium respectively. The electrodes were dried at 85 ℃ in a vacuum. The mass loading of each electrode was close to 1 mg. The single electrode Performance was measured in the electrolyte of 6M KOH and 1M H_2SO_4 in a three-electrode system configuration with Hg/HgO reference electrode (Ag/AgCl in 1M H_2SO_4) and platinum mesh as counter electrode while the symmetric device was tested in the two-electrode system. The symmetric device was prepared using glass microfibre filter paper as a separator in a CR2032 coin cell configuration. The thickness of the working electrode was measured from FESEM cross sectional area and is calculated to be 75-100 µm. 1M Na_2SO_4 was used as an electrolyte for the test in a neutral medium. From a single electrode, the specific capacitance (C_s) Calculation was carried out from Galvanostatic Charge discharge (GCD) curves using the following equation

$$C_s = \frac{I\Delta t}{m(V_f - V_i)} \quad \text{(S1)}$$

Specific capacitance (C_s) can be calculated from the CV curve by using the following equation

$$C_s = \frac{\int IdV}{2m\Delta V\upsilon} \quad \text{(S2)}$$

Where C_s = specific capacitance

I = current applied (A)

Δt = discharge time (sec)

m = mass of the active material (g)

$V_f - V_i$ = Voltage window (V)

$\int Idv$ $= area\ under\ the\ CV\ curve$

υ = scan rate (mV s^{-1}).

For a symmetric supercapacitor device, the calculation was done using the following equations.

$$C_s = \frac{2I\Delta t}{m(V_f - V_i)} \quad \text{(S3)}$$

$$ED = \frac{C_{cell}\Delta V^2}{2 \times 3.6} = \frac{C_s \Delta V^2}{8 \times 3.6} \quad \text{(S4)}$$

$$PD = \frac{E \times 3600}{t} \quad \text{(S5)}$$

Where C_s = specific capacitance, ED = Energy density, and PD = power density.

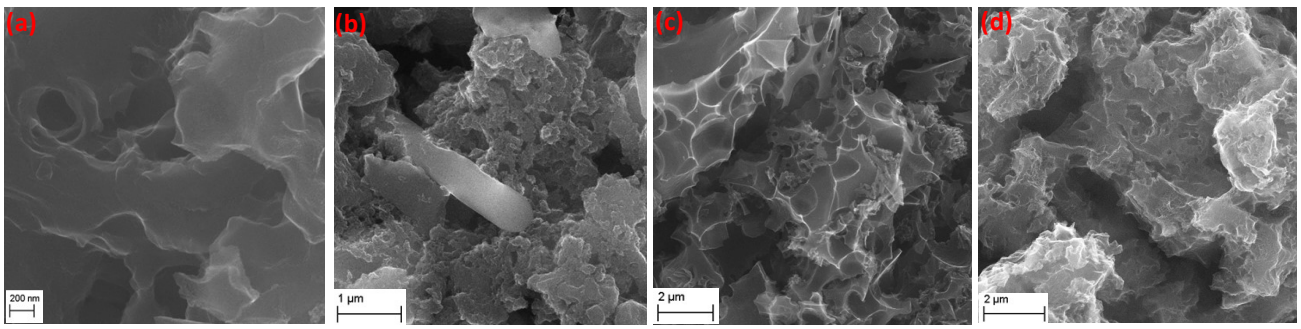

Figure S1. FESEM image of (a) p-CNS-600, (b) p-CNS-700, (c) p-CNS-800, and (d) p-CNS-900 samples.

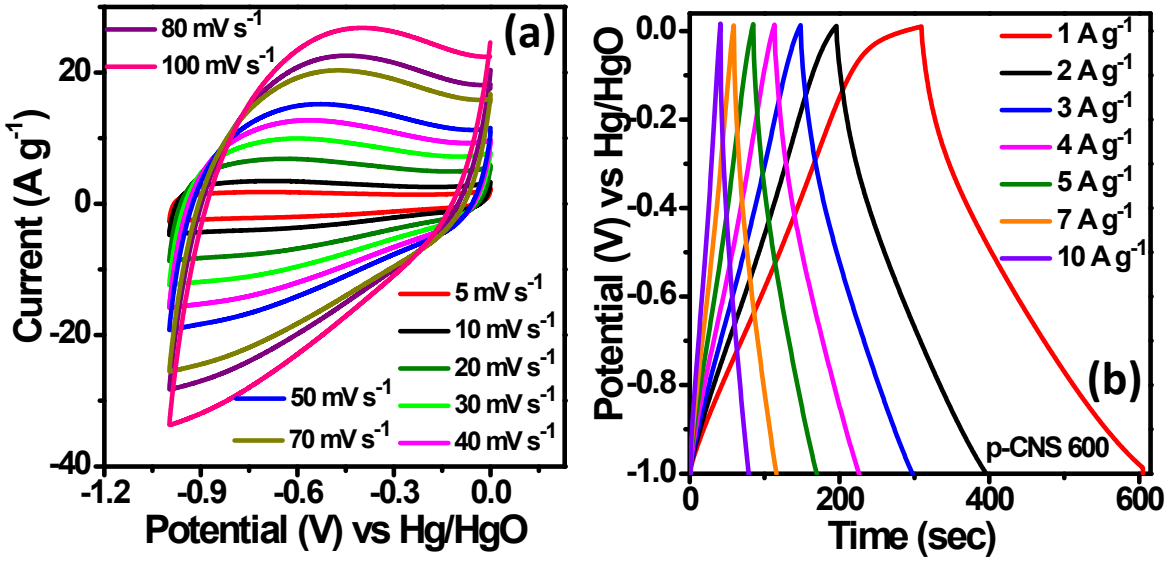

Figure S2. CV and GCD profiles of p-CNS-600.

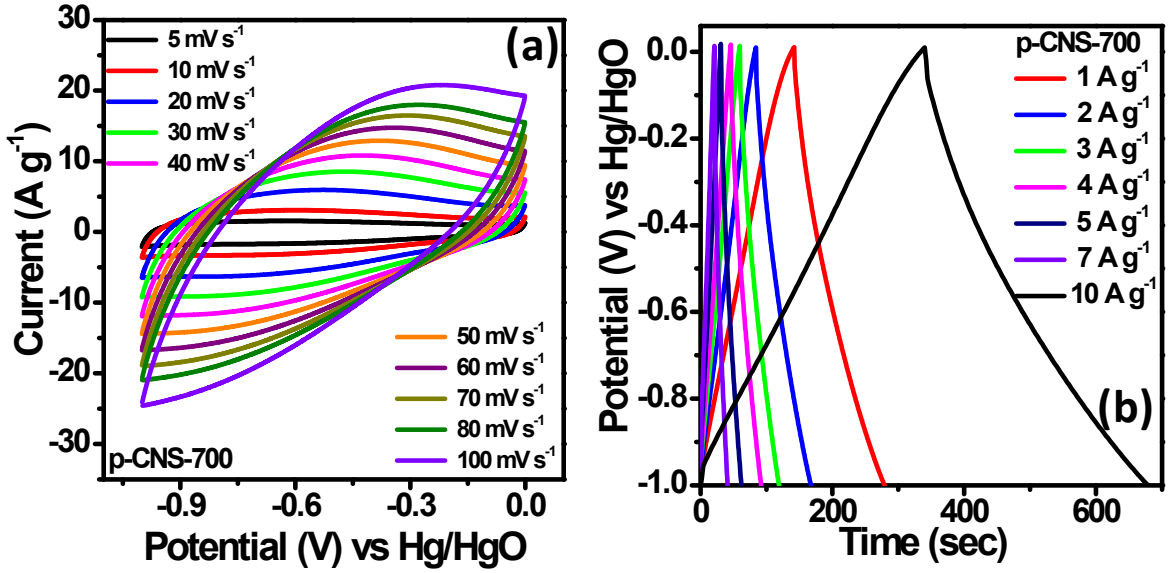

Figure S3. CV and GCD profiles of p-CNS-700.

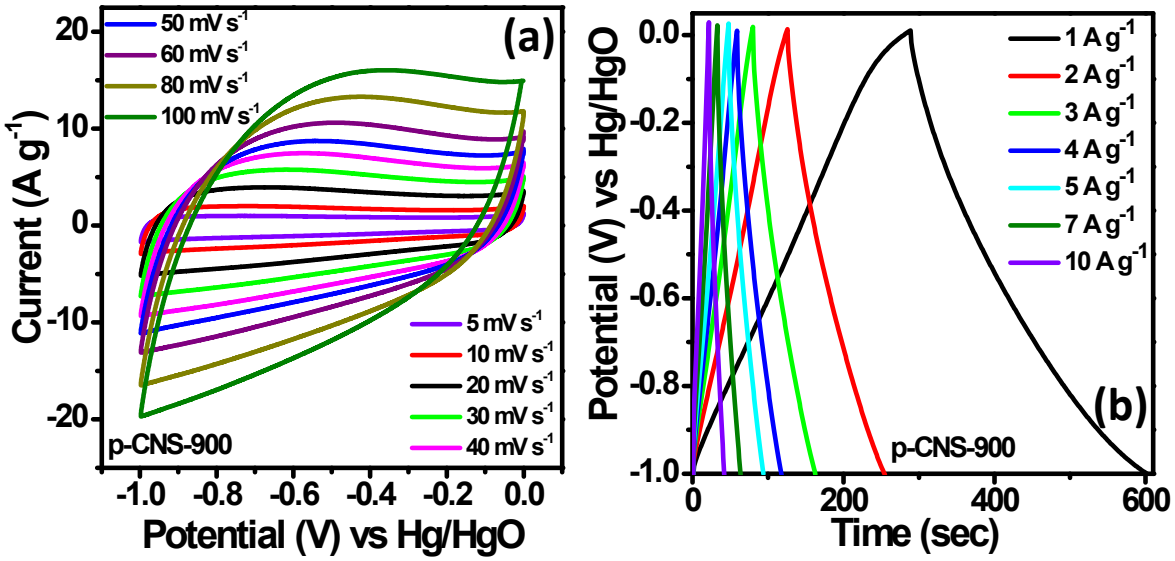

Figure S4. CV and GCD profiles of p-CNS-900.

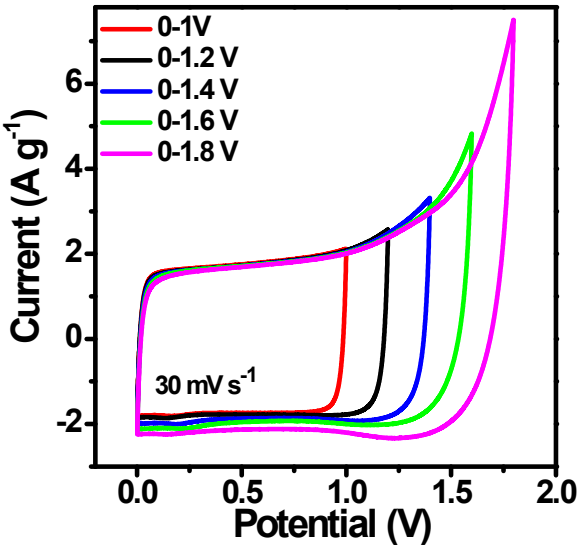

Figure S5. CV curves of a symmetric capacitor at the different potential range of 1 to 1.8 V in 1M Na_2SO_4.

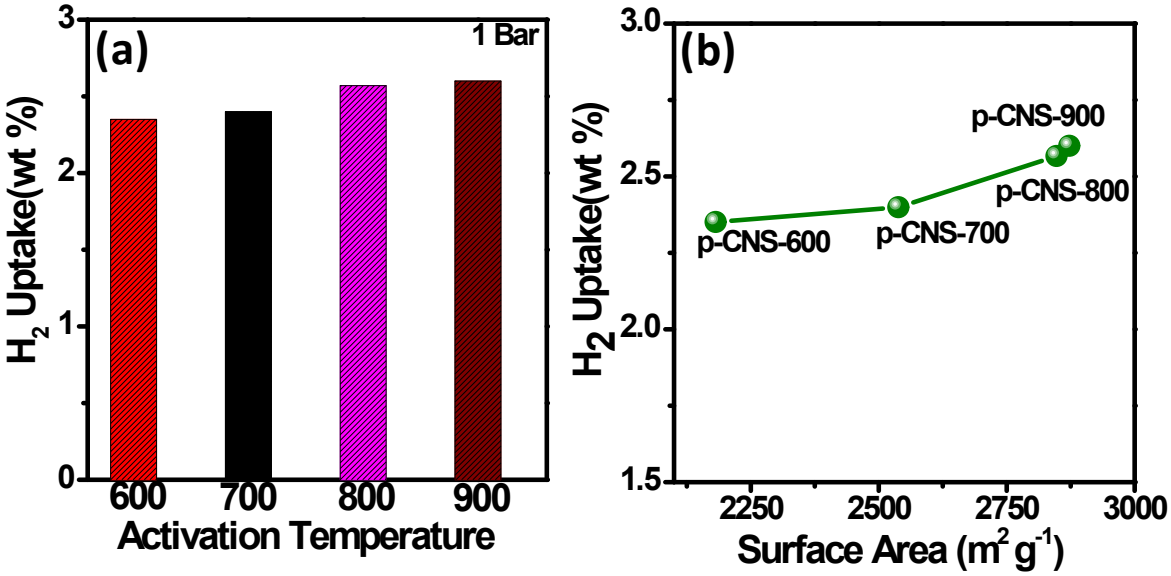

Figure S6. (a) H_2 uptake of all the samples at 77K and 1 bar pressure. (b) Change in H_2 uptake as a function of surface area

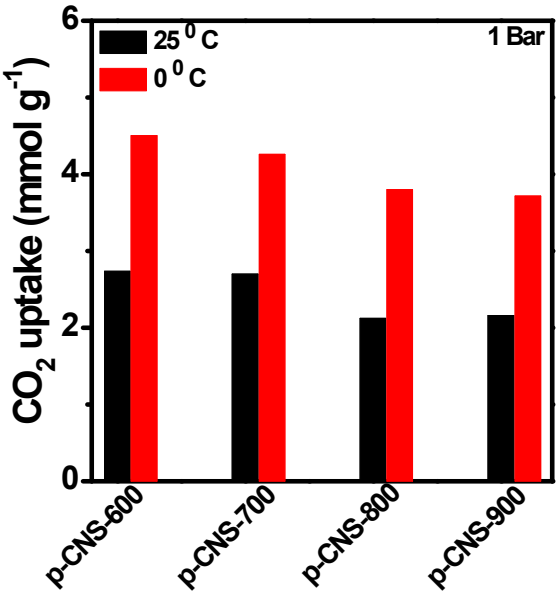

Figure S7. CO_2 uptake capacities of p-CNS-600 to 900 at 1 bar pressure.

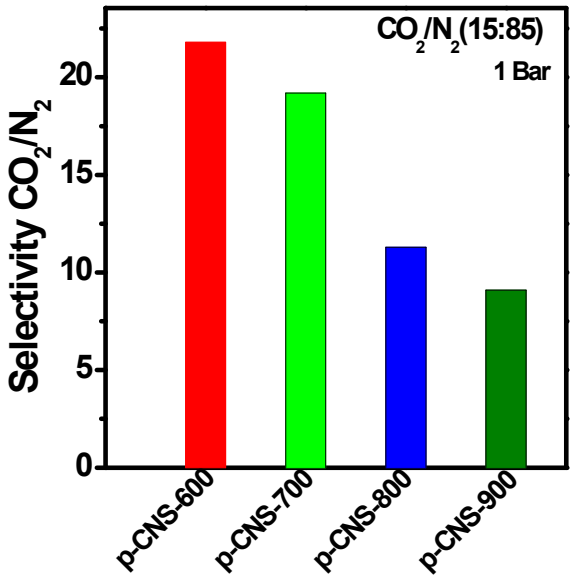

Figure S8.CO_2/N_2 selectivities of SPCs for binary mixture of ratio (15:85) under pressure of 1 bar.

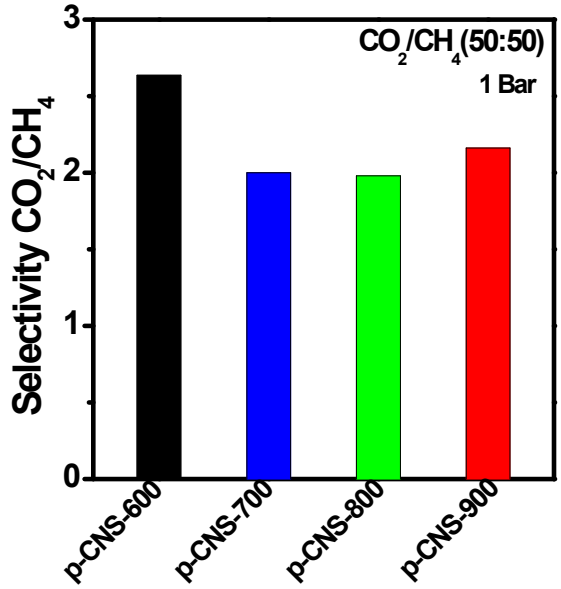

Figure S9. CO_2/CH_4 selectivities for binary mixture of ratio (50:50) under pressure of 1 bar

Table S1. Weight % of C, S, N, and O present in p-CNS-X samples calculated from SEM EDX analysis.

Sample Name	C (%)	S (%)	N (%)	O (%)
p-CNS-600	60.60	7.40	10.80	21.20
p-CNS-700	82.70	3.10	3.20	11.00
p-CNS-800	85.10	1.1	3.70	10.09
p-CNS-900	89.00	0.20	3.50	7.30

Table S2**.** Comparison of electrochemical performance of p-CNS-X samples in 6M KOH electrolyte.

Sample Name	**Specific capacitance (F g^{-1}) at 1 A g^{-1} current density (6M KOH electrolyte)**
p-CNS-600	299
p-CNS-700	343
p-CNS-800	375
p-CNS-900	315