Highly Porous Graphitic Carbon and Ni₂P₂O₇ for High Performance Aqueous Hybrid Supercapacitor

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SEM images of waste writing paper and product obtained after hydrothermal treatment



Figure S1: SEM images of (a) waste writing paper and (b) product obtained after hydrothermal treatment at different magnification



SEM image of HPGC

Figure S2: Low magnification SEM images of HPGC

TEM image of HPGC



Figure S3: (a) *TEM image* (b) & (c) *HR-TEM image of HPGC. High magnified image* (c) *shows small pores in HPGC*

Raman spectra of HPGC and commercial activated carbon

Raman spectroscopy is the best tool to characterize graphitic carbon. Usually, in Raman spectra two band named D band and G band appears. The D band, commonly known as defect band, is related to breathing mode of sp² hybridized carbon rings which is will active if ring is adjacent to defect. However, the G band is characteristic of the in plane vibration of sp²-bonded carbon atoms. The second-order 2D band is typical of graphitic carbon and its broadening gives impression about number of layer.^{1,2} Along with this, the intensity ratio between G and D band is also directly proportional to the degree of graphitization.^{3,4} Thus based on above points we have also calculated I_G/I_D for our HPGC and compared it with the commercial available activated carbon (Fig. S4). For HPGC sample I_G/I_D was 1.38 however, for commercial activated carbon this value is ~0.86 which suggest that HPGC is graphitized. Additionally, in comparison to commercial activated carbon we also observed more prominent broad 2D band around 2760 cm⁻¹ which also suggest that HPGC has been graphitized.



Figure S4: Raman spectra of HPGC and commercial activated carbon

Cyclic voltammetry of commercial activated carbon



Figure S5: Cyclic voltammetry of commercial activated carbon at different scan rates

SEM image of HPGC electrode before and after half-cell charge-discharge



Figure S6: SEM images of HPGC (a) & (b) before and (c) & (d) after 1500 charge discharge cycle



TGA plot and XPS of Ni₂P₂O₇

Figure S7: (a) TGA and DTA of as prepared $Ni_2P_2O_7$ without calcination (b) (b) survey scan, (c) Ni 2p, (d) O 1s and (e) P 2p XPS spectra of $Ni_2P_2O_7$

Element	Atom (%)	Wt (%)
Ni	12.8	24.73
Р	15.9	16.07
О	57.2	29.96
Cu	14.1	29.24
Total	100	100

Table S1: Table showing presence of element in atom (%) and weight (%) in $Ni_2P_2O_7$

* Presence of Cu element is due to the Cu grid

Redox behavior of Ni₂P₂O₇

To confirm the redox behavior, the Ni₂P₂O₇ electrodes were studied using CV measurements at various scan rates of $2 - 10 \text{ mV s}^{-1}$ which is given in main text Fig. 6 (b). The peak current values increased with increase in scan rate and follow Randles-sevick equation. The voltammetric response of the electrode was studied using the power law $i = av^b$, where v is the scan rate, and both a & b are adjustable parameters.⁵⁻⁸ The parameter b is determined from the slope of the linear plot of log i versus log v (Fig S8) and its value will provide information about capacitive (b = 1) or battery behavior (b = 0.5) of electrode. As the b-value approaches 0.5 (0.508 calculated from Fig. S8), the charge storage of Ni₂P₂O₇ electrode is mainly controlled by the diffusion controlled process. Thus, the charge storage performance of Ni₂P₂O₇ electrode is mainly by OH⁻ ion intercalation/deintercalation process.



Figure S8: Peak current versus scan rate of $Ni_2P_2O_7$

SEM image of Ni₂P₂O₇ electrode after half-cell charge-discharge



Figure S9: *SEM images of* Ni₂P₂O₇ *after 750 charge discharge cycle*

Cyclic voltammetry of as prepared sample



Figure S10: Cyclic voltammetry of as prepared sample (without calcination) at different scan rates

Formula for Specific capacitance, energy density and power density calculations

Specific capacitance calculation

$$C = \frac{I\Delta t}{m\Delta V}$$

Energy density calculation

$$E = \frac{I\Delta V\Delta t}{m}$$

Power density calculation

$$P = \frac{I\Delta V}{m}$$

where C, E, P, $I \Delta V$, Δt and m are specific capacitance, energy density, power density, current, potential difference, discharging time and mass of electroactive material.

Mass balance equations

It is well known that the positive and negative charge should be balanced $(q_+=q_-)$ for fabrication of the full cell.⁸⁻¹⁰ The mass balancing will follow equation (1),

$$\frac{m_{+}}{m_{-}} = \frac{c_{-}}{c_{+}} \times \frac{\Delta E_{-}}{\Delta E_{+}}$$
 ----- (1)

where, m is mass of the active material in the electrode and ΔE is the working potential of the material. The specific capacitance calculated from the discharge curve is 384 Fg⁻¹ for AC at a current density of 4 A g⁻¹. In the same current density, Ni₂P₂O₇ delivered a capacitance of 1554 Fg⁻¹. On the basis of the specific electric quantity values, the mass ratio of negative to positive electrode is calculated to be ~ 5:2 (2.5:1) mg. So the mass loading of the negative electrode material was adjusted to 2.5 mg and positive electrode is 1 mg. Total mass of the two electrode is 3.5 mg. The working potential of the cell is optimized as 1.6 V which is higher than that of the AC based symmetric supercapacitor (1.0 V) in alkali aqueous electrolyte.

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