Supporting Information

Superior Magnetocapacitance in Ferro/Ferrimagnetic Fe₃O₄/Fe/Fe₃C integrated Ndoped Carbon Hybrid Nanostructures under Mild Magnetic Fields

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Keywords: Supercapacitor, hybrid nanostructures, Ferro/Ferrimagnetic, Magnetocapacitance, Magnetohydrodynamic

Pyrolysis condition number	Temperature and time	
COND1	600 °C \rightarrow 1 h, 800 °C \rightarrow 2 h	-
COND2	600 °C \rightarrow 2 h, 800 °C \rightarrow 2 h	-
COND3	600 °C \rightarrow 2 h, 800 °C \rightarrow 3 h	-
COND4	600 °C \rightarrow 1 h, 900 °C \rightarrow 2 h	-
COND5	600 °C \rightarrow 1 h, 900 °C \rightarrow 3 h,	-
COND6	600 °C \rightarrow 0.5 h, 850 °C \rightarrow 2.5 h	Suitable condition
COND7	$600 ^{\circ}\text{C} \rightarrow 2 \text{ h}, 900 ^{\circ}\text{C} \rightarrow 3 \text{ h}.$	-

Table S1. Pyrolysis conditions of catalysts synthesis

Mechanistic study of Fe@NCNS hybrid nanostructure formation

The controlled pyrolysis of a precursor mixture containing biowaste, melamine, and iron nitrate through thermal condensation resulted in the formation of the Fe@NCNS hybrid nanostructure. Melamine first undergoes direct polycondensation, resulting in the formation of graphitic carbon nitride nanosheets at 600 °C, while biowaste creates a carbon skeleton within the interlayers of the carbon nitride. In the following higher temperatures, the Fe₃O₄/Fe/Fe₃C nanoparticles generated from the thermal decomposition of iron nitrate (the reaction process involves a series of reactions: $Fe^{3+} \rightarrow Fe_3O_4 \rightarrow Fe \rightarrow Fe_3C \rightarrow \alpha$ -Fe above 700 °C) will get confined within these sandwich layers through the Fe-N coordination. Continued heating leads to the thermal decomposition of carbon nitride, while iron nanoparticles cause the graphitization of biowaste, yielding 2D carbon nanosheets and resulting in the formation of hybrid nanostructure at the end of the pyrolysis process. Therefore, the hybrid nanostructure was formed through the simultaneous in situ catalytic graphitization effect of metal nanoparticles. Graphitic carbon nitride served not only as a carbon source for the formation of distinct thin carbon shells but also as a nitrogen source incorporated into both the carbon shells and the layered 2D carbon nanosheets. This strategy is especially appealing for practical applications since both the synthesis process and synthesis materials are feasible for large scale production. The variation

in reaction temperature and duration produced hybrid nanostructures with distinct physical structures and chemical compositions.

Synthesis process of nitrogen doped porous carbon nanosheets (PNCNS)

Biowaste camachile fruit peel was initially dried and grounded into a fine powder form and is precarbonized at 300 °C for 1 hr in an inert atmosphere. This pulverized biowaste precursor is mixed with sodium bicarbonate (NaHCO₃), and melamine in a 1:1:0.5 mass ratio and is carbonized in a tubular furnace at 850 °C for 2 h in argon atmosphere. The collected sample was washed simultaneously with ethanol and DI water. The dried final product is called as PNCNS.

Materials characterization

High resolution transmission electron microscopy (TECNAI F20) and field emission scanning electron microscopy (Quanta 200 (FEG)) were used to observe the morphological properties of the prepared materials. X-ray powder diffraction (XRD) patterns were recorded on a D8 Advance x-ray diffractometer (Bruker). Raman spectra were taken on a WiTec Alpha spectrometer, with an Nd:YAG laser at an excitation wavelength of 532 nm. The X-ray photo electron spectroscopy (K- α^+ , Thermo fisher scientific) experiments were carried out with a monochromatic Al-K α source. The surface area and pore size distribution analyses were measured by N₂ adsorption-desorption technique in an automated surface area and porosity analyzer (ASAP 2020, Micromeritics) at 77 K. Brunauer-Emmett-Teller (BET) method was used to measure the total surface area using N₂ adsorption desorption isotherms. Pore size distribution was determined from the differential desorption isotherms using the Barrett-Joyner-Halenda (BJH) models. Temperature dependent magnetization measurements (5-300 K) were carried out in quantum design MPMS SQUID VSM. Zero field cooled/field cooled (ZFC/FC) magnetization measurements were carried out by applying constant filed of 100 Oe.

Electrochemical characterizations

Electrochemical properties of the prepared electrode materials were studied in three electrode and full cell assemblies. Electrode were prepared by mixing the electrode material and polytetrafluoroethylene (PTFE) in a 95:5 mass ratio in N-methyl-2-pyrrolidone (NMP) solution to form a thick slurry. The slurry was coated onto a carbon paper (1cm x 1cm) and dried them at 90 °C for 12 h. These electrodes were used as working electrodes with a mass loading of

approximately ~ 3 mg in a three electrode supercapacitor assembly system with platinum (Pt) wire as a counter electrode and Hg/HgO as a reference electrode in 3 M KOH electrolyte. Cyclic voltammograms (CVs), galvanostatic charge-discharge curves (GCDs), and electrochemical impedance spectra (EIS) studies were conducted by applying an AC signal of amplitude 5 mV in the frequency range of (10 mHz-100 kHz) for the assembled supercapacitor devices using the Biologic SP-300 electrochemical workstation. For the full cell ASC assembly, the working electrodes were prepared by coating the slurry on copper foil and then dried them at 90 °C for 9 hr in a vacuum oven. The dried electrodes were cut in the form of circular discs of 12 mm diameter. For this ASC assembly a negative electrode (PNCNS) mass was maintained as 2.2 mg and positive electrode (Fe@NCNS) mass was maintained as 0.9 mg. 2032 coin cells were assembled in an argon filled glovebox, where the oxygen and moisture levels were maintained below 0.1 ppm. Polypropylene sheet was used as a separator. The electrolyte used was 3 M KOH. To examine the effect of the external magnetic fields, the three electrode and two electrode cell configurations were placed between the two magnetic coils connected to the DC power supply (current range 0-5 A). From the given equation below the calculated magnetic field B at the center was 2 mT, 4 mT and 6 mT, respectively.

B= μ_0 . N. I/b(1+(x/b)^2)^{3/2}

Where, b is the radius of the magnetic coil (b=12 cm), N is the number of turns (N=400), μ_0 is the permeability of free space =1.25x10⁻⁶ T. mA⁻¹, x is the coil distance on axis to the inflection point (x=4 cm), and I is supplying electric current from a DC power supply. The magnetic flux density was adjusted through tuning the magnitude of the current and the exact amount of MF is measured using a tesla meter. An electric current of approximately ~0.56 A, ~1.12 A, and ~1.68 A were supplied to provide a magnetic field strength of 2mT, 4 mT and 6 mT, respectively.

The specific capacitance (F/g) of the electrodes was measured from the galvanostatic charge discharge curves following the equation:

$$C_{s} = \frac{\Delta t \times I}{\Delta V \times m} - \dots - (S1)$$

For half-cell measurements
$$C_{cell} = \frac{t_{discharge \times I}}{\Delta V \times M} - \dots - (S2)$$

For full cell supercapacitor

$$E = \frac{1 \times C_{cell} \times (\Delta V)^2}{2 \times 3.6} - \dots - (S3)$$
$$P = \frac{E_{cell} \times 3600}{t_{discharge}} - \dots - (S4)$$

Where I, ΔV , m, and Δt are discharge current (A), potential window (V), mass of the active material (g), and discharge time (s), respectively. The specific capacitance, energy density E(Wh/Kg), power density P(W/Kg) of the asymmetric supercapacitor were measured following the equations (2)-(4).

Where C_{cell} is the gravimetric specific capacitance of the device (F/g), M is the total active mass of both electrodes (g), I is the discharge current (A), ΔV is the voltage window (V), and t_{discharge} is the discharge time (s).

Using the following equation, we balanced the mass loading of the positive and negative electrodes to assembled the asymmetric supercapacitor.

 $\frac{M_{+}}{M_{-}} = \frac{C_{-} \times \Delta V_{-}}{C_{+} \times \Delta V_{+}}$, the variables M₋ and C₋ represent the mass and specific capacitance of the negative electrodes and M₊ and C₊ represent the mass and specific capacitance of the positive electrodes, respectively. The optimal mass ratio between the two electrodes, M_{+}/M_{-} can be adjusted to achieve balanced charge storage.

The degree to which surface capacitive and diffusion processes influence capacitance was assessed utilizing the power law equation¹

 $i = a\nu^b - \dots - \dots - \dots - (S6)$

The charge storage process is governed by surface control when b=1, diffusion control when b=0.5, and a combination of both surface and diffusion-control when b is $0.5 \le b \le 1$. The calculated b values derived from the linear fitting of log (ν) versus. log(i) at 0 mT and 6 mT are found to be 0.60, and 0.48, respectively. The observations suggest that at 0 mT, the capacitor storage is mainly dominated by both physical capacitive effect and ion diffusion process. However, as the MF strength increases to 6 mT, the diffusion-controlled process begins to significantly contribute to energy storage. The capacity contribution ratio was determined using the equation²

 $i(V) = k_2 v^{\frac{1}{2}} + k_1 v$ ------ (S7), where i denote the current response at a specific potential V, and $k_1 v$ and $k_2 v^{\frac{1}{2}}$ represents the surface capacitive behavior and diffusion contribution, respectively. The current values of CV at varying scan rates can determine the k_1 and k_2 values.



Figure S1. FE-SEM images of the Fe@NCNS at (a) COND1, (b) COND2, (c) COND3, (d) COND4, (e) COND5, and (f) COND7.



Figure S2. Elemntal mapping of (a) Overlay, (b) Carbon, (c) Oxygen, (d) Nitrogen, and (e) Iron, (F) EDS spectra for the Fe@NCNS hybrid electrode.



Figure S3. (a) N_2 adsorption-desorption isotherms, (b) BJH pore size distribution plot of Fe @NCNS.



Figure S4. (a) FE-SEM image, (b) EDS spectra, (c) XRD patterns, (d) Raman spectra, (e) BET surface area, and (f) Pore size distribution of the PNCNS.

Figure S4a illustrates the interconnected crumpled graphene like porous carbon nanosheets, forming a three-dimensional nanostructure. The EDS spectra **Figure S4b** shows the presence of surface nitrogen functional groups. **Figure S4c** presents the XRD pattern featuring a prominent diffraction peak 26° and a weak intensity diffraction peak at 43°. These peaks are indicative of graphitic nanostructures, cofirming the formation of 3D graphene like carbon

nanosheets and suggesting a high degree of graphitization. The Raman spectra for the PNCNS is shown in **Figure S4d**, displaying the characteristic Raman peaks of the D, G and 2D bands. **Figure S4e** shows the BET N₂ adsorption-desorption isotherms, exhibiting characteristics of type-I, type-IV isotherms. The measured specific surface area is $2057 \text{ m}^2/\text{g}$. Figure S4f presents the analysis of pore size distribution for PNCNS, revealing the existence of micro, meso and macro pore structure.



Figure S5. XPS survey spectrum of the Fe@NCNS.



Figure S6. Nyquist plots under under varied magnetic fields for the Fe@NCNS with the equivalent circuit.



Figure S7. (a) GCD curves under 2 mT, (b) GCD curves under 4 mT for the Fe@NCNS based ASC.

The electrochemical performance of the negative PNCNS electrode was evaluated in a three electrode assembly

The electrochemical performance of the negative PNCNS electrode was evaluated in a three electrode assembly using a 3 M KOH electrolyte. **Figures S8a-b** show the CV and GCD curves. The extensive integral area under the CV curves signifies a substantial specific capacitance. The nearly rectangular CV shape suggests contribution from both EDLC capacitance and pseudocapacitive energy storage. The surface functional groups plays a significant role in contributing to the pseudo capacitance. The GCD curves exhibit a linear behavior along with excellent reversibility. A capacitance of 581.5 F/g is obtained at a current density of 1.5 A/g, with a capacitance retention of 81.5% % at 45 A/g, indicating excellent rate capability (81%). The **Figure S8c** illustrates the Nyquist plot for the PNCNS electrode. The values of $R_s=2.6 \Omega$ and $R_{ct}= 5.6 \Omega$ indicate low electrode and electrolyte resistances, which facilitates rapid ion diffusion at the electrode-electrolyte interface and enhances charge storage capabilities. The presence of wrinkles and edges in carbon nanosheets enhances the accessible electrode surface area to the electrolyte, promoting rapid ion diffusion and resulting in superior electrochemical performance. The high conductivity, high surface area and hierarchical porous structure of the PNCNS electrode material effectively mitigate the capacitance decay.



Figure S8. (a) CV curves at different scan rates, (b) GCD curves at different current densities, and (c) Nyquist plot for the N-doped porous carbon nanosheets derived from biowaste (PNCNS) in three electrode system with 3 M KOH electrolyte.



Figure S9. (a) Percentage ratio of capacity contribution at various scan rates at 0 mT, (b) Percentage ratio of capacity contribution at various scan rates at 6 mT, (c) Linear plot between peak current and scan rate to determine the b value.

Pyrolysis	D band position	G band position	2D band	I_D/I_G ratio
condition	(cm^{-1})	(cm ⁻¹)	position	
			(cm ⁻¹)	
COND1	1335.8	1563.7	2664.5	1.08
COND2	1338.4	1569.3	2673.3	1.12
COND3	1340.7	1571.6	2681.7	1.20
COND4	1341.0	1569.8	2673.3	1.08
COND5	1341.2	1577.6	2676.6	1.05
COND6	1334.8	1564.2	2667.7	0.90
COND7	1341.2	1569.8	2682.1	0.93

Table S2. Raman analysis data for the Fe@NCNS

Sample Name	Surface	Total pore	Micro pore	Pore Width
	area (m ² /g)	volume (cm ³ /g)	volume (cm^3/g)	(nm)
Fe@NCNS	632	0.48	0.45	3.5

Table S3. The BET surface area and pore size distribution of the prepared electrode material.

Electrode material	Specific capacitance	Current density (A/g) or	Electrolyte	Reference
	(F/g)	Scan rate (mV/s)		
Fe@NCNS	1327.3	1.5 A/g	3M KOH	This work
Fe ₃ O ₄ /carbon	139	0.5 A/g	1 M KOH	[3]
Fe ₂ O ₃ /VACNT	248	8 A/g	2 M KOH	[4]
Fe ₂ O ₃ QDs/FGS	347	10 mV/s	1 M Na ₂ SO ₄	[5]
Fe ₃ O ₄ /graphene	661	0.5 A/g	1MKOH	[6]
Fe ₃ O ₄ /rGO	96	1mA/cm ²	1MKOH	[7]
FeOOH/GNS/CNS	267	0.5 A/g	1 M Li ₂ SO ₄	[8]
Mesoporous carbon/Fe ₂ O ₃	235	0.5 A/g	1M Na ₂ SO ₃	[9]
pyrrole treated Fe ₃ O ₄ film	106	1 A/g	1M Na ₂ SO ₃	[10]
Fe ₃ O ₄ -M-CNT	145.4	1 A/g	1M Na ₂ SO ₄	[11]
Fe ₃ O ₄ /rGO	480	1 A/g	1 M KOH	[12]
Fe ₃ O ₄ /C	386	1 A/g	2 M KOH	[13]
NCSs@Fe ₃ O ₄	206	1 A/g	6 M KOH	[14]
Fe ₃ O ₄ /rGO	455	3.6 A/g	2 М КОН	[15]
Fe ₃ O ₄ /CNFs	135	0.42 A/g	1M Na ₂ SO ₃	[16]

 Table S4. Comparision of the electrochemcial performance of the reported materials for supercapacitors

Electrode material	Supercapacitor	Electrolyte	Magnetic	R_s	R_{ct}	W_i
	assembly		field	(Ω)	(Ω)	(Ω)
			strength			
Fe@NCNS	Three electrode	3 M KOH	0 mT	0.52	4.65	2.5
			2 mT	0.46	4.11	2.2
			4 mT	0.42	3.46	1.85
			6 mT	0.30	2.93	1.56
Fe@NCNS	ASC	3 М КОН	0 mT	0.62	5.99	3.2
			2 mT	0.50	4.70	2.53
			4 mT	0.41	4.10	2.20
			6 mT	0.30	3.20	1.71
PNCNS	Three electrode	3 М КОН	0 mT	2.6	5.6	3.96

 Table S5. A list of resistance measurement values obtained for the design supercapacitor systems.

Materials	Applied	Specific	Current density	References
	Magnetic field	capacitance	(A/g) or Scan	
	strength	increment (%)	rate (mV/s)	
MMNS	72 mT	70 %	5 mV/s	[17]
NG/Cdots/Fe ₃ O ₄	8.98 mT	53.8%		[-,]
NiO/NG	8.98 mT	6%	5 mV/s	[18]
Co ₃ O ₄ /NG	8.98 mT	28%	5 mV/s	[19]
MnO ₂ /ECNFs	1.34 mT	19 %	5 mV/s	[20]
Fe ₃ O ₄ /G	125 mT	92%	5 mV/s	[21]
МСО	3 mT	5%	3 mA/cm2	[22]
FCO	3 mT	40%	10 mV/s	[23]
NiCo ₂ O ₄ (NCO)	5 mT	25%	10 mV/s	[24]
NiO	5 mT	15%	10 mV/s	[25]
Co_3O_4 (CCO)	5 mT	7%	10 mV/s	[26]
FeCo ₂ O ₄	3 mT	40%	0.25 A/g	[27]
MnCo ₂ O ₄	3 mT	5%	3 mV/s	[28]
CCO	3 mT	4%	3 mV/s	[29]
NG/Cdots/Fe ₃ O ₄	8.98 mT	53.8%	5 mV/s	[30]
Fe@NCNS	6 mT	55%	1.5 A/g	This work

Table S6. Comparision of the electrochemcial performance of the reported materials for supercapacitors under external magnetic fields

Electrode	Supercap	Magnetic	specific	Rate	energy	cycle
material	acitor	field	capacitance	capability	density	stability
	assembly		(F g ⁻¹)/ at 1	(%)	(Wh/kg)	(%)
			A g ⁻¹	at 45 A g ⁻¹	at 1.5 A g ⁻¹ /	
					45 A g ⁻¹	
Fe@NCNS	Three	0 mT	1327.3	88.3	-	-
	electrode	2 mT	1606.1			
		4 mT	1924.5			
		6 mT	2057.3			
Fe@NCNS	ASC	0 mT	297.0	81.7	181.9/148.5	94.3
		2 mT	341.5		209.2/170.5	
		4 mT	395.0		241.9/197.5	
		6 mT	430.6		263.7/215.3	96.7
PNCNS	Three	0 mT	581.5	81.5	-	-
	electrode					

Table S7. A list of electrochemcial performance values obtained for the design supercapacitor systems.

Energy storage	Electrolyte	Magnetic	Energy	Power	cycle life	Ref
device		field	density	density		
		Strength				
Ti ₃ C ₂ T _x /NiCo- LDH-3T//AC	PVA-KOH	3 T	0.134 mW h/cm ²	1.61 mW/cm ²	82.3% after 6000 cycles	[27]
NNA@MnO ₂ // NNA@PPy	ЗМ КОН		48.88 Wh/kg	1275 W/kg	106.6% after 20000 cycles	[28]
NCS-12T//AC	PVA-KOH	12 T	$280 \ \mu Wh/cm^2$	1.71 mW/cm ²	84.1% after 5000 cycles	[29]
NiCoFe/NiCoF e-OH- G4//GCA	ЗМ КОН	0.15 T	61.4 Wh/kg	974.6 W/kg	92.7% after 20000 cycles	[30]
NCZP ^{6T} //FeS	PVA-KOH	6 T	106.9 Wh/cm ²	1.71 mW/cm ²	86.6% after 5000 cycles	[31]
CZ55S ^{6T} /G//AC	6М КОН	6 T	65.6 Wh/kg	903 W/kg	83.6% after 5000 cycles	[32]
Co ₃ V ₂ O ₈ //RG O//RGO		0.5 T	28.36 Wh/kg	400 W/kg	97.75% after 10000 cycles	[33]

 Table S8. Electrochemical performance of device in the presence of the magnetic field

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