

## *Electronic Supplementary Information*

### **Nanostructured Mesophase Electrode Materials: Modulating Charge-Storage Behavior by Thermal Treatment**

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Table S1. Elemental analysis data of CGPNHs

Sample	Element %			N/C
	N	C	H	
2CGPNH500	12.269	74.423	1.421	0.165
4CGPNH500	13.134	73.310	1.806	0.179
6CGPNH500	15.240	70.484	2.361	0.216
8CGPNH500	15.329	69.932	1.930	0.219
10CGPNH500	15.895	69.376	2.341	0.229
2CGPNH600	12.206	76.657	1.465	0.159
4CGPNH600	12.827	76.696	1.504	0.167
6CGPNH600	14.993	72.810	1.709	0.206
8CGPNH600	15.474	72.231	2.130	0.214
10CGPNH600	15.523	71.702	1.828	0.216
2CGPNH800	5.954	84.637	0.735	0.070
4CGPNH800	9.059	80.653	1.042	0.112
6CGPNH800	10.141	78.310	1.146	0.129
8CGPNH800	10.340	77.168	1.272	0.134
10CGPNH800	10.965	76.796	1.460	0.143
2CGPNH1000	2.804	86.988	0.780	0.032
4CGPNH1000	3.871	83.805	0.687	0.046
6CGPNH1000	4.191	83.069	0.853	0.050
8CGPNH1000	4.830	85.729	1.510	0.056
10CGPNH1000	5.379	84.017	1.769	0.064

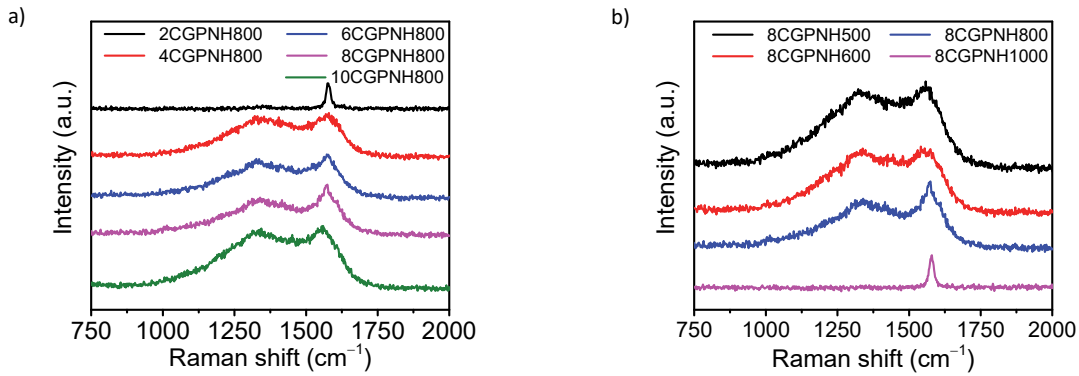


Figure S1. Raman spectra of CGPNHs prepared with (a) different  $P_w/G_w$  ratios at a  $T_H$  of 800 °C and (b) different  $T_H$  conditions at a  $P_w/G_w$  ratio of 8:1 (532 nm excitation).

Table S2. Intensity ratios of G peak to D peak calculated from the data in Figure S1a.

Sample	$I_D/I_G$
2CGPNH800	0.03
4CGPNH800	0.94
6CGPNH800	0.91
8CGPNH800	0.79
10CGPNH800	0.96

Table S3. Intensity ratios of G peak to D peak calculated from the data in Figure S1b.

Sample	$I_D/I_G$
8CGPNH500	0.92
8CGPNH600	0.97
8CGPNH800	0.79
8CGPNH1000	0.34

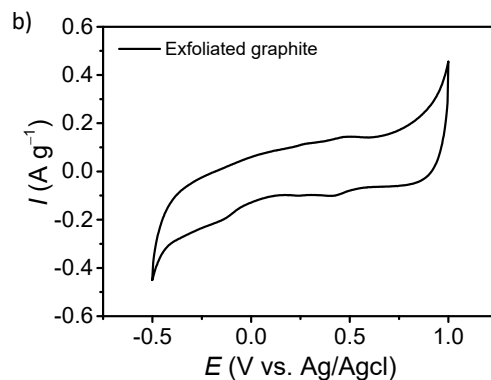
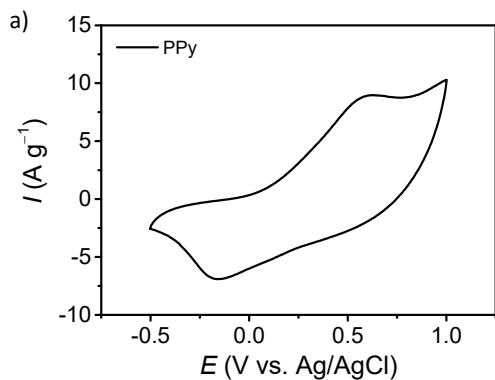


Figure S2. CV curves of control samples recorded at a scan rate of 25 mV s<sup>-1</sup>: (a) PPy nanoparticles\_only and (b) exfoliated graphite\_only.

Table S4. BET surface areas of the representative samples\*

Sample	BET Surface Area (m <sup>2</sup> g <sup>-1</sup> )
8CGPNH500	345
8CGPNH600	384
8CGPNH800	388
8CGPNH1000	303

\*Nitrogen sorption experiments were performed at 77.4 K with Micromeritics ASAP 2020.

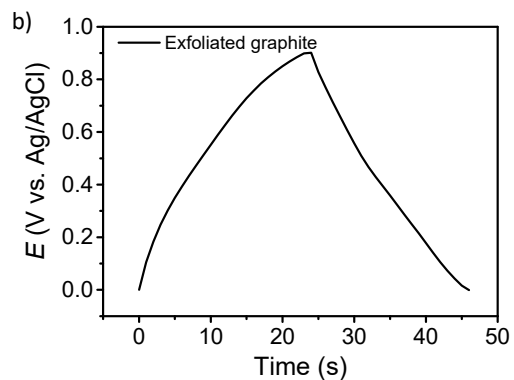
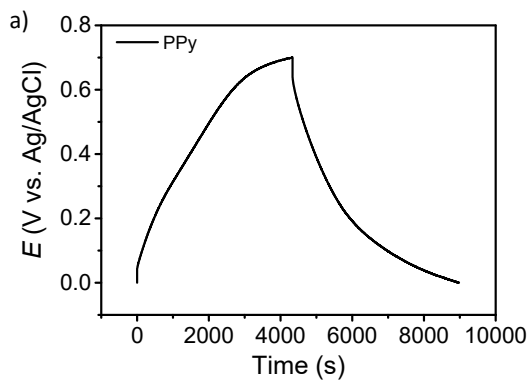


Figure S3. Charge/discharge curves of control samples recorded at a current density of  $0.1 \text{ A g}^{-1}$ : (a) PPy nanoparticles\_only and (b) exfoliated graphite\_only.

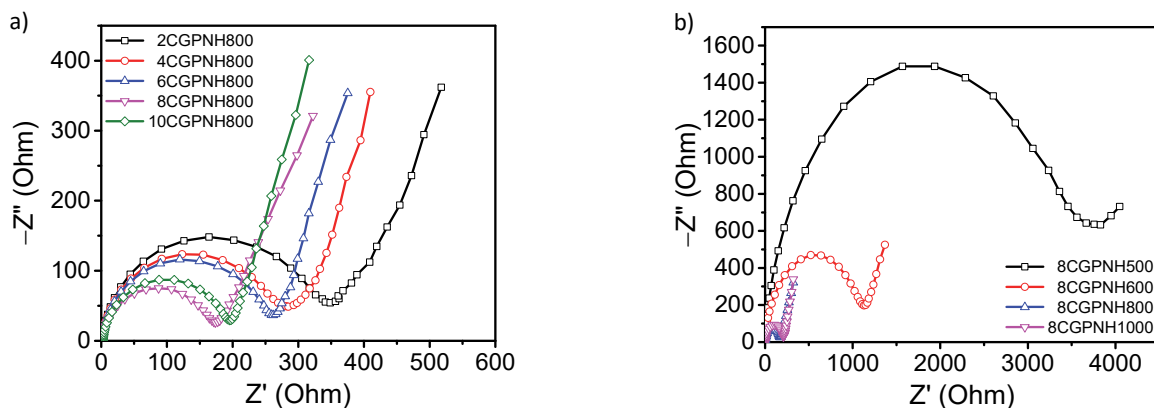


Figure S4. Electrochemical impedance spectroscopy Nyquist plots of the representative samples: CGPNHs prepared with (a) different  $P_w/G_w$  ratios at a  $T_H$  of 800 °C and (b) different  $T_H$  conditions at a  $P_w/G_w$  ratio of 8:1. A similar trend was observed in the *IR* drop data.

Table S5.  $R_{ct}$  values calculated from the data in Figure S4a.

Sample	$R_{ct}$ ( $\Omega$ )
2CGPNH800	347
4CGPNH800	280
6CGPNH800	257
8CGPNH800	172
10CGPNH800	193

Table S6.  $R_{ct}$  values calculated from the data in Figure S4b.

Sample	$R_{ct}$ ( $\Omega$ )
8CGPNH500	3784
8CGPNH600	1128
8CGPNH800	172
8CGPNH1000	208

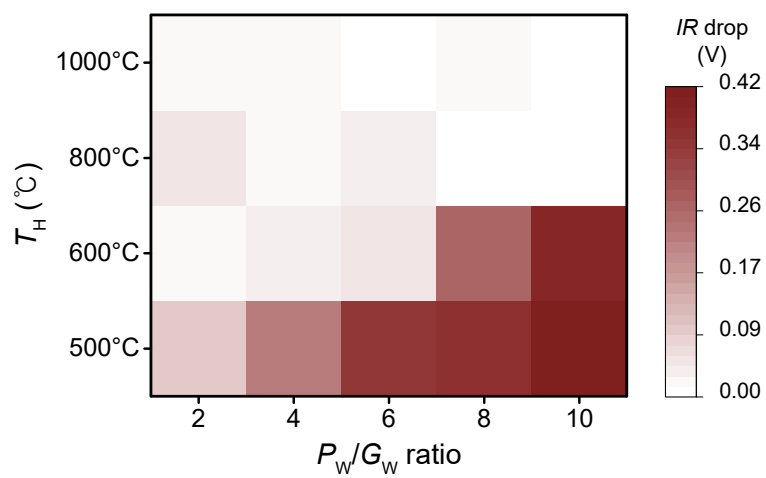


Figure S5. *IR* drop values calculated from the data in Figure 6a.



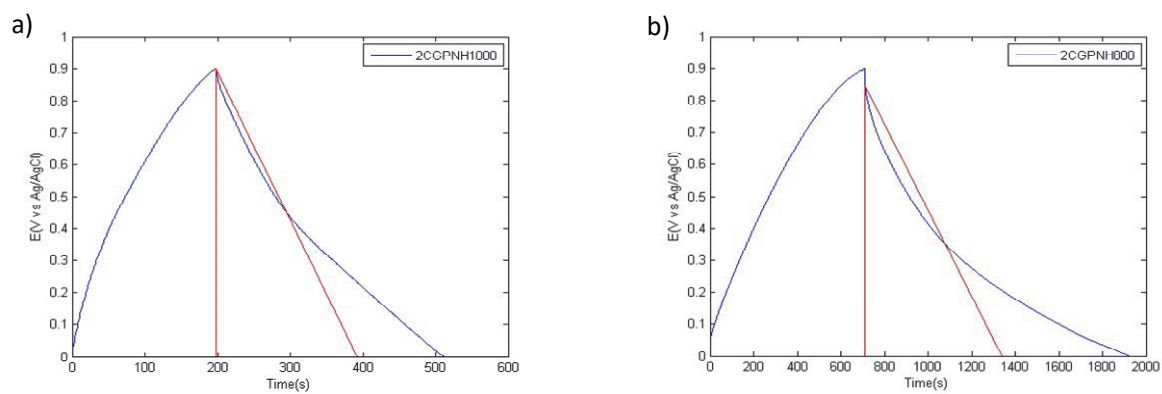


Figure S6. Representative examples of calculating EPR index values from the charge/discharge curve using the algorithm.

Table S7. Full data of calculated EPR index values

Sample	<i>I</i>	<i>E</i>	<i>P</i>	EPR	Error
2CGPNH500	6.69±3.81	151.58±37.64	72.76±15.70	67.32 : 32.67	±4.63
4CGPNH500	8.25±3.02	88.28±14.92	63.05±10.95	58.28 : 41.72	±3.96
6CGPNH500	3.50±1.43	21.43±8.08	30.37±10.98	41.04 : 58.96	±1.71
8CGPNH500	2.55±0.68	10.17±3.55	16.01±3.18	37.88 : 62.12	±4.36
10CGPNH500	1.01±0.27	2.79±0.92	5.54±1.44	33.21 : 66.79	±2.66
2CGPNH600	20.54±9.19	282.31±70.53	127.22±23.30	68.44 : 31.56	±5.24
4CGPNH600	14.57±6.65	229.11±41.10	114.05±12.88	66.32 : 33.68	±5.16
6CGPNH600	16.00±8.01	268.21±50.19	165.66±16.80	61.43 : 38.57	±3.06
8CGPNH600	4.85±1.81	128.87±26.33	96.30±15.45	57.04 : 42.96	±1.36
10CGPNH600	4.16±0.32	26.20±2.38	20.20±3.15	56.65 : 43.35	±3.98
2CGPNH800	17.53±5.97	218.55±18.19	84.42±5.51	72.05 : 27.95	±2.29
4CGPNH800	25.46±6.96	285.74±15.59	107.49±14.96	72.72 : 27.28	±3.09
6CGPNH800	20.64±7.74	304.96±30.23	165.26±22.41	64.91 : 35.09	±2.29
8CGPNH800	38.16±13.00	428.38±39.67	273.19±41.44	61.20 : 38.80	±1.68
10CGPNH800	17.22±3.30	359.47±65.94	227.74±42.25	61.23 : 38.77	±0.20
2CGPNH1000	3.17±0.87	86.14±23.33	28.94±7.23	74.61 : 25.39	±2.35
4CGPNH1000	11.06±3.19	150.16±9.31	55.42±1.08	73.00 : 27.00	±1.08
6CGPNH1000	7.39±2.69	144.84±43.29	73.85±23.99	66.37 : 33.63	±0.73
8CGPNH1000	17.11±4.92	261.48±35.09	147.14±8.67	63.78 : 36.22	±2.34
10CGPNH1000	21.83±2.30	324.60±66.91	192.53±47.69	62.92 : 37.08	±1.76

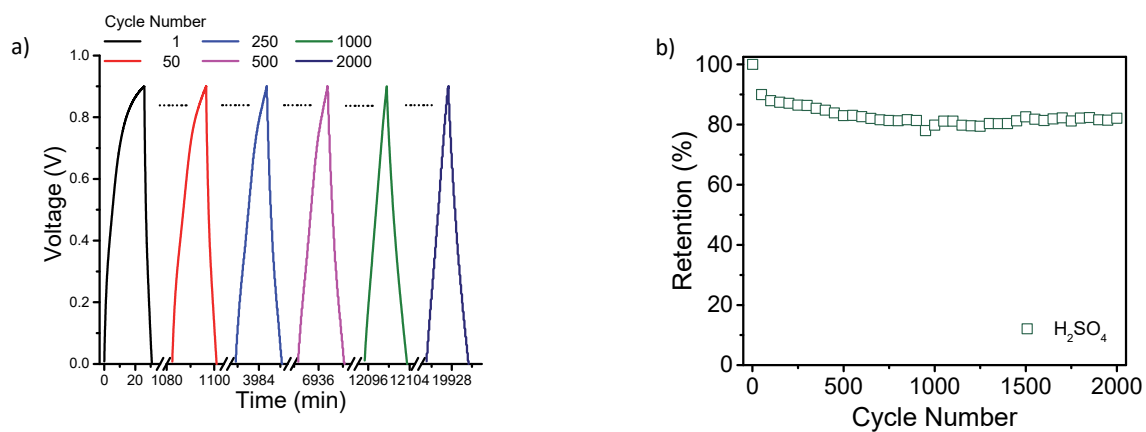


Figure S7. Long-term cycling stability of the 8CGPNH800 capacitor cell assembled with sulfuric acid electrolyte, cellulose membrane, and stainless steel current collector:

a) Representative galvanostatic charge/discharge curves recorded at a current density of  $0.1 \text{ A g}^{-1}$  and b) plot of capacitance retention against cycle.

Table S8. Summary of the data used for Figure 9c.

Sample	Electrolyte	Energy density (Wh kg <sup>-1</sup> )	Power density (kW kg <sup>-1</sup> )
1	1M H <sub>2</sub> SO <sub>4</sub>	22.4–15.0	0.18–9.00
2	1M H <sub>2</sub> SO <sub>4</sub>	13.3–10.0	0.18–9.00
3	1M Na <sub>2</sub> SO <sub>4</sub>	5.7–2.5	0.18–9.00
5	6M KOH	15.2–10.0	0.18–9.00
6	6M KOH	9.1–5.0	0.18–9.00
Ref. [26]	2M KOH	4.8–2.7	0.03–0.96
Ref. [27]	5M KOH	8.5–6.9	0.63–20.00
Ref. [28]	1M Na <sub>2</sub> SO <sub>4</sub>	2.8–1.9	0.1–2.6
Ref. [29]	6M KOH	9.4–5.2	1.1–103.0
Ref. [30]	0.5M NaCl	12.50–0.25	0.50–0.18
7	HMIM-PF <sub>6</sub>	74.4–10.5	0.6–30.0
Ref. [27]	EMIMBF <sub>4</sub>	80.0–65.6	0.44–9.10
Ref. [31]	PYR <sub>14</sub> TFSI	22.5–7.5	1.9–5.2
Ref. [32]	EMIMBF <sub>4</sub>	57.0–49.0	0.4–18.0