

## Electronic Supplementary Information

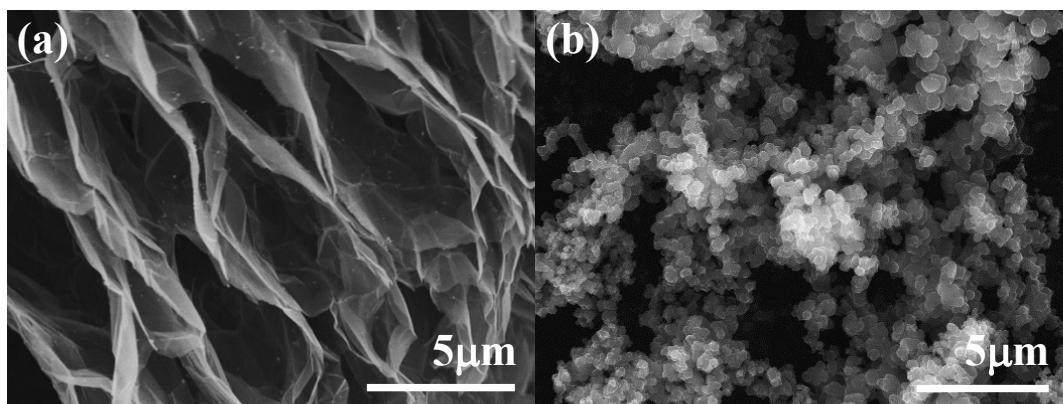
# Design and fabrication of polypyrrole/expanded graphite 3D interlayers nanohybrids towards highly capacitive performance

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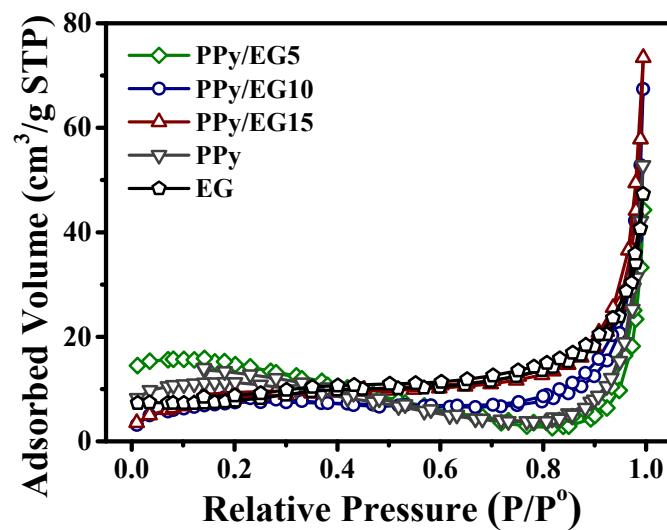
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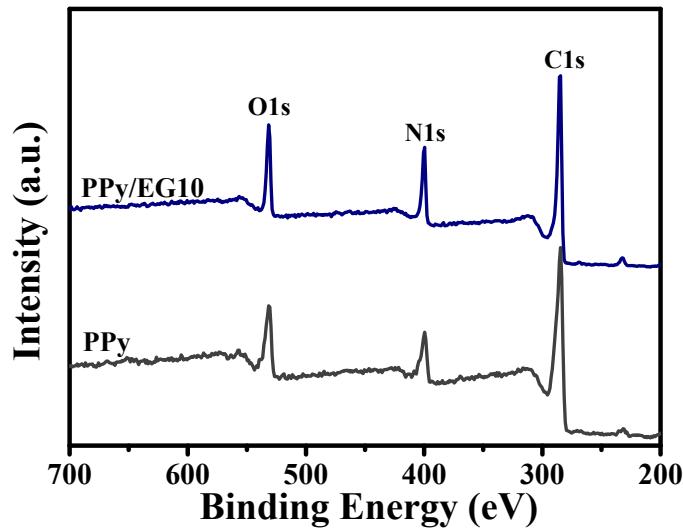
**Fig. S1** The SEM images of (a) EG and (b) PPy



**Fig. S2**  $\text{N}_2$  adsorption-desorption isotherms of EG, PPY, PPY/EG15, PPY/EG10 and PPY/EG5 samples

**Table S1** The specific surface area of EG, PPY, PPY/EG15, PPY/EG10 and PPY/EG5 samples

Sample	$S_{\text{BET}} (\text{m}^2 \text{ g}^{-1})$
PPY	15.8
PPY/EG5	22.5
PPY/EG10	28.7
PPY/EG15	30.9
EG	26.3



**Fig. S3** XPS full-scan spectra of PPy and PPy/EG10 samples

**Table S2** Elemental composition of the EG, PPy and PPy/EG10 samples

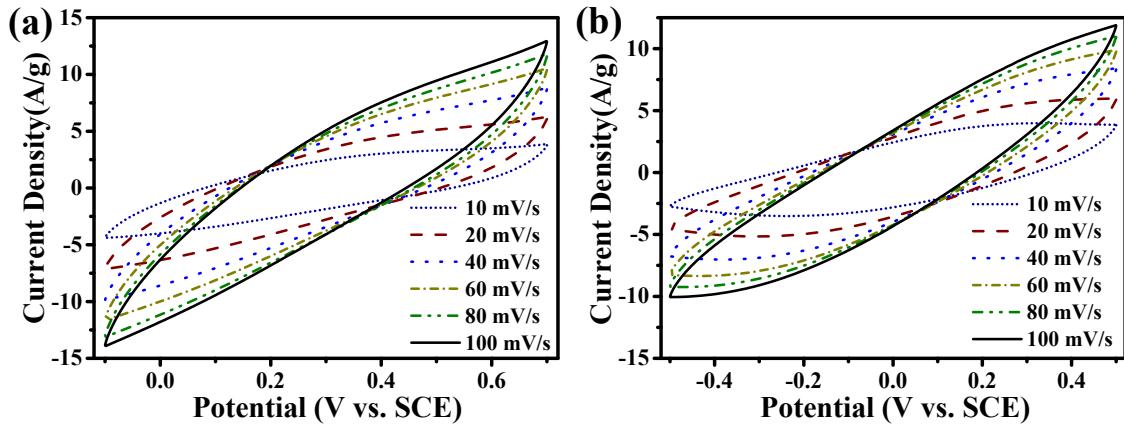
Sample	C1s(at.%)	O1s(at.%)	N1s(at.%)
PPy	71.89	12.49	15.62
EG	97.26	2.48	0.27
PPy/EG10	76.90	11.18	11.92

**Table S3** The calculated values of  $R_s$ ,  $R_{ct}$ ,  $Z_w$ ,  $C_{dl}$  and  $C_{ps}$  through CNLS fitting of the experimental impedance spectra based on the proposed equivalent circuit displayed in Fig. 6a

Samples	$R_s$ ( $\Omega \text{ cm}^{-2}$ )	$R_{ct}$ ( $\Omega \text{ cm}^{-2}$ )	$Z_w$ ( $\Omega \text{ cm}^{-2}$ )	$C_{dl}$ ( $\mu\text{F cm}^{-2}$ )	$C_{ps}$ ( $\text{mF cm}^{-2}$ )
EG	0.5516	0.071	0.1215	145.5	0.7768
PPy/EG5	0.5365	1.908	0.0614	20.94	1543
PPy/EG10	0.3699	1.839	0.1249	35.18	2839
PPy/EG15	0.8563	1.464	0.1904	23.04	1680
PPy	0.8665	2.944	0.1177	34.64	3254

**Table S4** The calculated values of  $R_s$ ,  $R_{ct}$ ,  $Z_w$ ,  $C_{dl}$  and  $C_{ps}$  through CNLS fitting of the experimental impedance spectra based on the proposed equivalent circuit displayed in Fig. 7a

Samples	$R_s$ ( $\Omega \text{ cm}^{-2}$ )	$R_{ct}$ ( $\Omega \text{ cm}^{-2}$ )	$Z_w$ ( $\Omega \text{ cm}^{-2}$ )	$C_{dl}$ ( $\mu\text{F cm}^{-2}$ )	$C_{ps}$ ( $\text{mF cm}^{-2}$ )
EG	1.201	0.257	0.2211	478.7	43.7
PPy/EG5	1.058	2.847	0.2526	22.23	1532
PPy/EG10	1.008	1.926	0.2461	30.84	2353
PPy/EG15	1.251	1.653	0.1883	25.49	1230
PPy	1.047	2.352	0.1373	19.64	3441



**Fig. S4.** The CV curves of PPy/EG10 electrode at the scan rate from 10~100  $\text{mV s}^{-1}$  in (a) 1 M  $\text{H}_2\text{SO}_4$  and (b) 1 M  $\text{KCl}$  electrolytes

**Table S5** The electrochemical properties of EG, PPy, PPy/EG15, PPy/EG10 and PPy/EG5 samples

Sample	1 M $\text{H}_2\text{SO}_4$ electrolyte			1 M $\text{KCl}$ electrolyte		
	$C_g$ ( $\text{F g}^{-1}$ )	Rate Capability (%)	Capacitance Retention (%)	$C_g$ ( $\text{F g}^{-1}$ )	Rate Capability (%)	Capacitance Retention (%)
PPy	293.6	33.8%	67.6%	266.9	33.4%	69.4%
PPy/EG5	376.4	54.7%	---	353.6	55.1%	---
PPy/EG10	454.3	75.9%	83.9%	442.7	73.3%	86.3%
PPy/EG15	402.5	78.4%	---	399.8	76.2%	--
EG	49.6	52.7%	95.9%	42.2	51.3%	96.8%

**Table S6** Specific capacitance of different carbon based PPy nanohybrids in three-electrode system

Material	Electrolyte	Current density (A g <sup>-1</sup> )	Specific capacitance (F g <sup>-1</sup> )	Ref.
Graphene/PPy nanotube aerogel	1 M H <sub>2</sub> SO <sub>4</sub>	0.5	253	[13]
PPy/sulfonated graphene	1 M H <sub>2</sub> SO <sub>4</sub>	0.3	310	[14]
Cellulose carbon aerogel/PPy	1 M H <sub>2</sub> SO <sub>4</sub>	0.5	387.6	[57]
PPy/modified GO	2 M NaNO <sub>3</sub>	1.0	202	[15]
Activated Carbons/PPy	1M Na <sub>2</sub> SO <sub>4</sub>	1.0	82.3	[24]
rGO/PPy aerogel	6 M KOH	0.5	304	[36]
PPy/EG	1 M H <sub>2</sub> SO <sub>4</sub>	1.0	454.3	
	1 M KCl	1.0	442.7	This work

**Table S7** Specific capacitance, power density and energy density of different carbon based PPy nanohybrids in two symmetric electrode system

Material	Electrolyte	Current density (A g <sup>-1</sup> )	Specific capacitance (F g <sup>-1</sup> )	Power density (kW kg <sup>-1</sup> )	Energy density (Wh kg <sup>-1</sup> )	Ref.
PPy/sulfonated graphene	1M KCl	0.3	253	0.1089 3.6	4.3 2.8	[14]
PPy/modified GO	2 M NaNO <sub>3</sub>	1.0	87	0.825	10	[15]
Graphene oxide/carbon dots/PPy	1M LiCl	0.5	216.8	0.25	30.1	[29]
PPy/EG	1 M Et <sub>4</sub> NBF <sub>4</sub> -PC	1.0	342.4	1.0 20	47.5 23.9	This work