

Electronic Supplementary Information

Redox additive Aqueous Polymer-gel Electrolyte for Electric Double Layer Capacitor

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Fig. S1

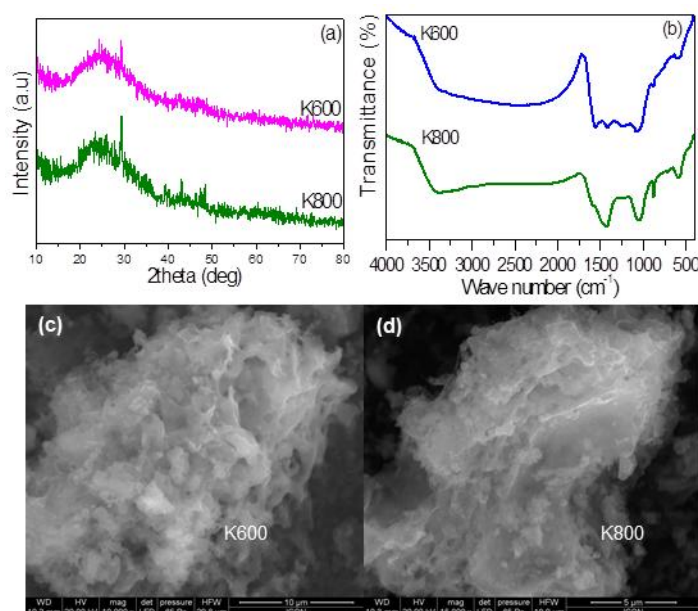


Fig. S1 XRD pattern (a), FTIR spectra (b) and (d) SEM image (c-d) of activated carbons (K600 and K800)

The absence of peaks in K800 indicates the reduction of functional groups by increasing the temperature. Notably, the peak around 1562 cm⁻¹ is not observed in K800. This peak belongs to the quinone functional group, so this sample exhibits the lower intrinsic resistance and pseudocapacitive interaction, resulting in lower capacitance.

Fig. S2

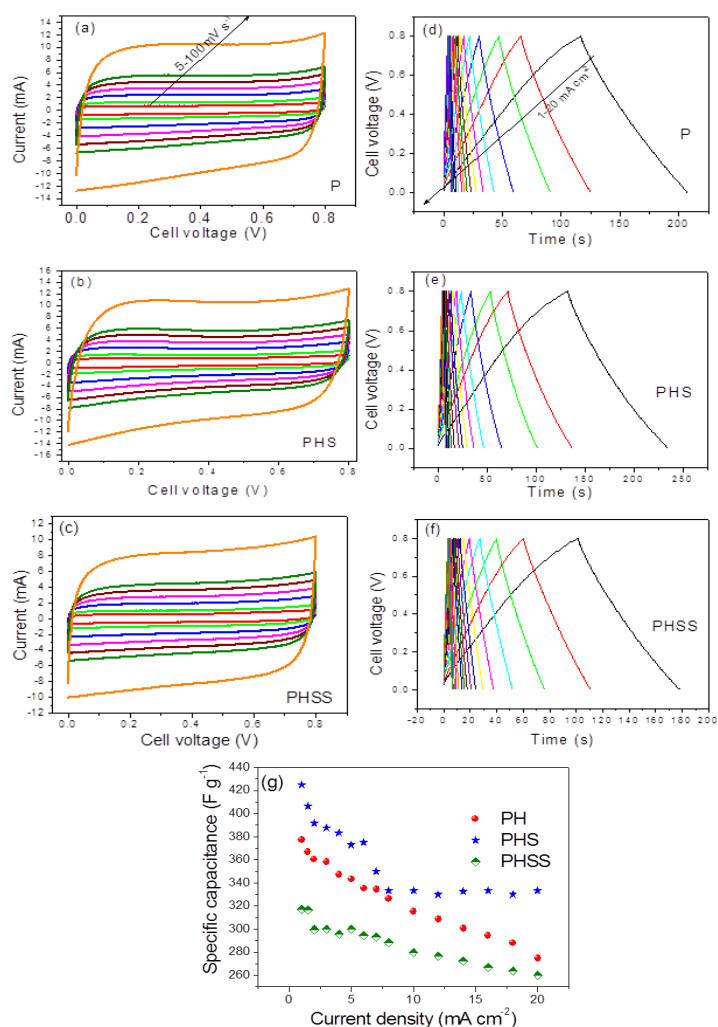


Fig. S2 CV curves at different scan rates (5-100 mV s⁻¹) (a-c), Charge-discharge curves at different current densities (1-20 mA cm⁻²) (d-f), current density vs. specific capacitance (g) of P, PHS and PHSS

The calculated specific capacitance (C_{sp}) from charge-discharge curves are 377, 425 and 317 F g⁻¹ for P, PHS and PHSS respectively at 1 mA cm⁻². Here, PHS shows the higher C_{sp} than the P and PHSS. Also PHS maintains the constant C_{sp} from 8-20 mA cm⁻² (330-333 F g⁻¹) which reveals its excellent individual electrochemical properties.

Fig. S3

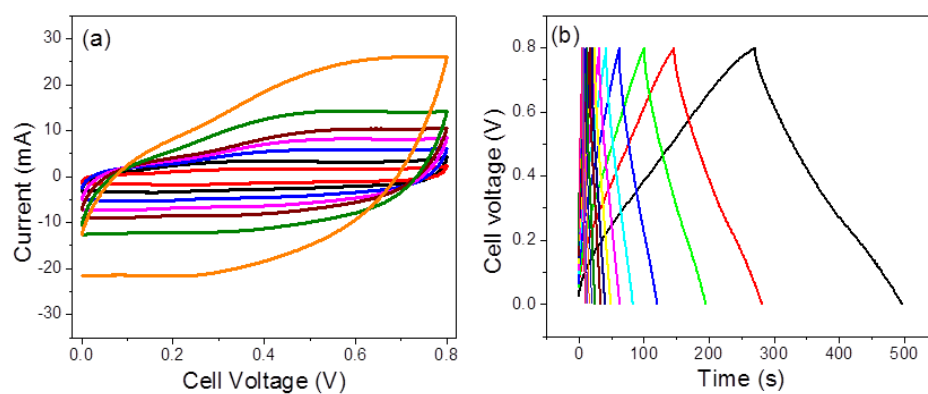


Fig. S3 CV curves (a) and Charge-discharge curves (b) of PHHQS at different current densities (1-20mA cm⁻²)

Fig. S4

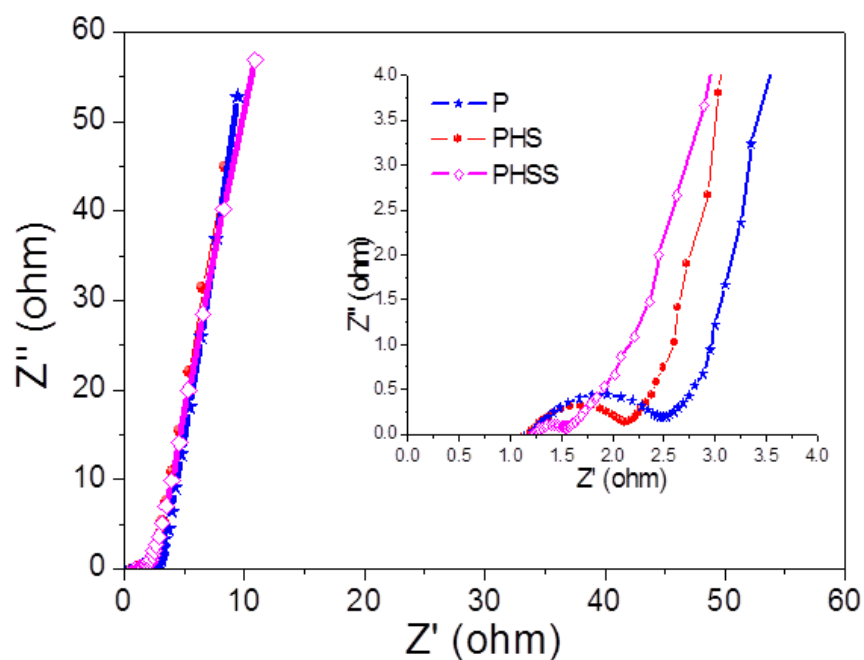


Fig. S4 Nyquist plots of P, PHS, PHSS

Here, the size of the semicircle loops decreased which indicates the decrease of the intrinsic resistance and pseudocapacitive interaction. These intrinsic resistance and pseudocapacitive interaction are strongly attributed to the presence of functional groups on the AC. In the present case based on ref. (1a, b), we conclude that the reason for decreasing intrinsic resistance is due the reduction of functional groups by increasing the carbonization temperature. Interestingly, it was confirmed via FTIR results (Fig. S1).

Fig. S5

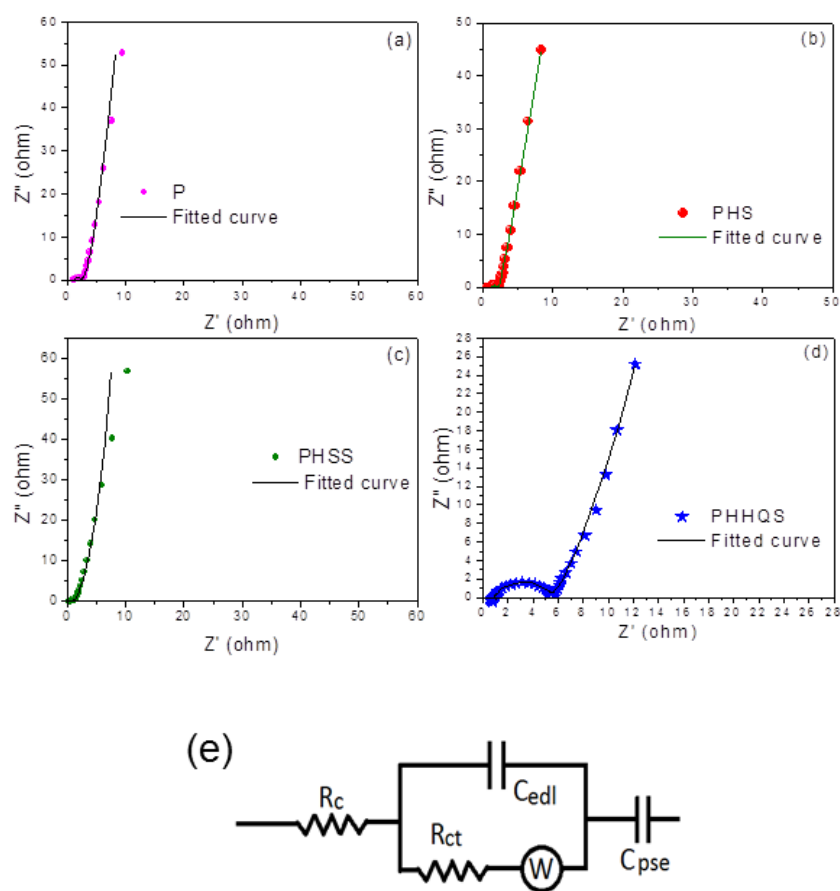


Fig. S5 Nyquist plots and experimental fitted curves of P, PHS, PHSS (a-c) and PHHQS (d) and used equivalent circuit for fitting the Nyquist plots (e)

Here, the EIS was measured in the frequency range of 30 mHz to 1 MHz for applied a.c voltage of 10 mV. Also, the EC-Lab V10.12 software was used for fitting the EIS data. The equivalent circuit includes bulk solution resistance, charge transfer resistance, electrical double layer element (C_{edl}), pseduocapacitance element (C_{pse}) and Warburg impedance (diffusive resistance, is the result of the frequency dependence of ionic transport in the electrolyte) (W). The calculated values of R_c and R_{ct} are given in table S1.

Table.S1

	Specific capacitance at 1 mA cm ⁻¹ (C _{sp}) (F g ⁻¹)	Energy density at 1 mA cm ⁻¹ (E) (Wh kg ⁻¹)	R _c (Ω)	R _{ct} (Ω)	W (Ω)
P	377	8	1.2	1.4	1.6
PHS	425	9	1.2	1	0.75
PHSS	317	7	1.1	0.6	2.5
PHHQS	941	20	1	4	1.9

The Specific capacitance and Energy density, Power density is calculated by following equations:

Specific capacitance for cell:

$$C_{cell} = \frac{I\Delta t}{MV} (F \cdot g^{-1})$$

Specific capacitance for single electrode:

$$C_{SP} = \frac{4I\Delta t}{MV} (F \cdot g^{-1})$$

Specific capacitance from EIS:

$$C_{SP} = \frac{2}{2\pi f Z''m} (F \cdot g^{-1})$$

Energy density for cell:

$$E = \frac{ItV}{7.2M} (Wh \cdot kg^{-1})$$

Power density for cell:

$$P = \frac{3.6E}{t} (W \cdot g^{-1})$$

Where, I is the applied current, M is the total mass of the active material (AC) in both electrodes, Δt is the discharge time, V is the potential range, 2πf=ω is a angular frequency, Z'' is a impedance imaginary values and m is a single electrode mass.

References:

- 1 (a) D. Hulicova, M. Kodama and H. Hatori, *Chem. Mater.*, 2006, **18**, 2318; (b) S. Roldan, I. Villar, V. Ruiz, C. Blanco, M. Granda, R. Menendez and R. Santamaria, *Energy Fuels*, 2010, **24**, 3422; (c) S. A. Hashmi, R. J. Latham, R. G. Linford and W. S. Schlindwein, *J. Chem. Soc. Faraday Trans.*, 1997, **93**, 4177.
- 2 R. Liu, S. I. Cho and S. B. Lee, *Nanotechnology*, 2008, **19**, 215710.
- 3 H. Yu, J. Wu, L. Fan, Y. Lin, K. Xu, Z. Tang, C. Cheng, S. Tang, J. Lin, M. Huang and Z. Lan, *J. Power Sources*, 2012, **198**, 402.
- 4 H. Yu, J. Wu, L. Fu, K. Xu, X. Zhang, Y. Lin and J. Lin, *Electrochim. Acta*, 2011, **56**, 6881.