## **Supporting Information**

## Tuning the nanoporous structure of carbons derived from the composite of cross-linked polymers for charge storage applications

Farshad Barzegar\*, Vladimir Pavlenko, Muhammad Zahid, Abdulhakeem Bello, Xiaohua Xia, Ncholu Manyala, Kenneth I. Ozoemena, Qamar Abbas\*

\* Corresponding authors: <a href="mailto:qamar.abbas@tugraz.at">qamar.abbas@tugraz.at</a>, <a href="mailto:farshadbarzegar@gmail.com">farshadbarzegar@gmail.com</a>

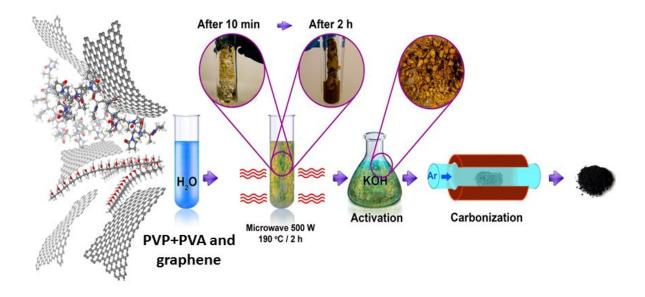


Figure S1. Various steps during the preparation of activated carbons with and without the use of graphene as additive. The reaction under microwave irradiation at 500 W (190°C, 2h) results in the production of hydrogel which possesses porous structure. The activation of hydrogel at high temperature ca. 700°C results in nanoporous carbon materials.

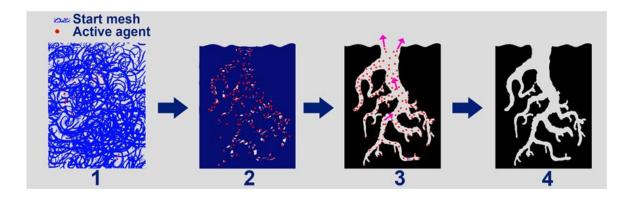


Figure S2. Model shows the gradual development of pores. 1 the formation of the polymer mesh with active agents, 2 is a temperature dependent process before carbonization that leads heat deformation of the mesh structure of polymer, 3 occurs during the carbonization process whereby the volatiles agents escape creating the pores within the carbon matrix and 4 is the final structure obtained after the process.

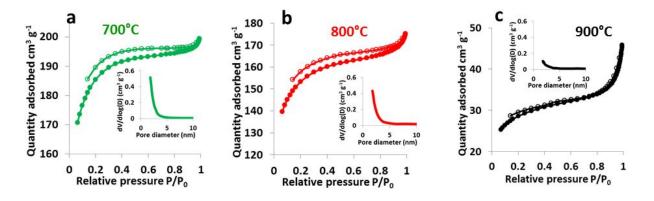


Figure S3. Nitrogen gas adsorption/desorption isotherm of the carbon sample obtained after activation of PVA (10 wt.%) alone at different temperature from 700°C to 900°C.

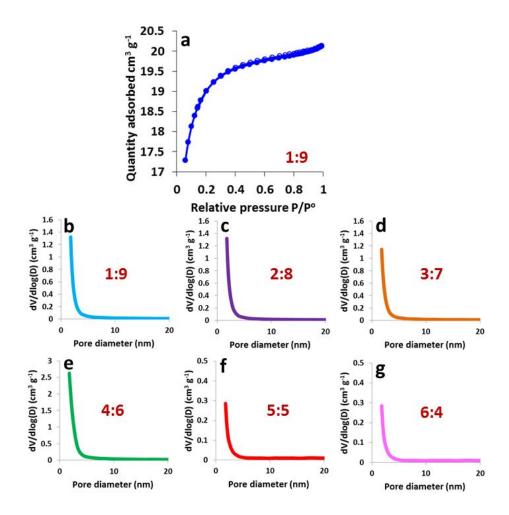


Figure S4. (a) Nitrogen gas adsorption/desorption isotherm of the nanoporous carbon sample obtained after activated of the hydrogel composite with PVP:PVA ratio of 1:9 at 700°C. (b-g) the pore size distribution for carbon materials obtained from composite with increasing PVP to PVA ratio 1:9 to 6:4.

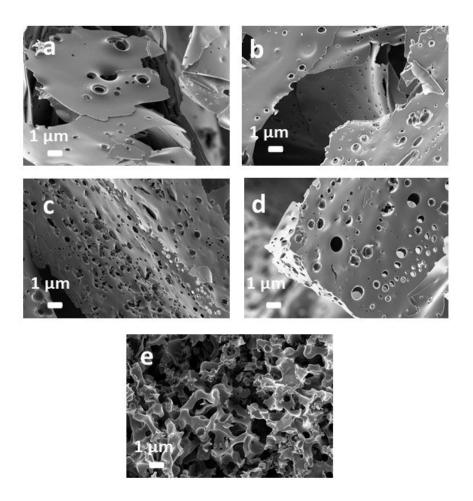


Figure S5. Scanning electron microscopy (SEM) images of the activated carbons obtained from the hydrogel composite material with PVP:PVA ratio (a) 1:9, (b) 2:8, (c) 3:7, (d) 4:6 and (e) 5:5. The composite material is firstly activated at 700°C and then after washing and cleaning it is post-treated again at 700°C to obtain carbon samples for electrode preparation for constructing supercapacitor cells.

Sample	1:9	2:8	3:7	4:6	5:5	6:4
BET (m <sup>2</sup> g <sup>-1</sup> )	1230	1249	1347	1846	606	440
micropore volume <sup>a</sup>	0.47	0.48	0.48	0.49	0.29	0.15
(cm <sup>3</sup> /g) <sup>a</sup>						
Total pore volume <sup>b</sup>	2.25	2.27	2.26	2.63	2.30	2.89
(cm <sup>3</sup> /g) <sup>b</sup>						
Pore diameter <sup>c</sup>	2.67	2.64	2.66	2.36	3.80	5.64
(nm) <sup>c</sup>						

<sup>&</sup>lt;sup>a</sup> t-Plot micropore volume

Table S1. BET surface area and porous textural data obtained for carbon materials after activation of hydrogel composite at 700°C for various increasing PVP to PVA ratio from 1:9 to 6:4. Highest surface area of 1846 m<sup>2</sup> g<sup>-1</sup> is achieved for sample 4:6 with microporous structure and an average pore diameter of 2.36 nm.

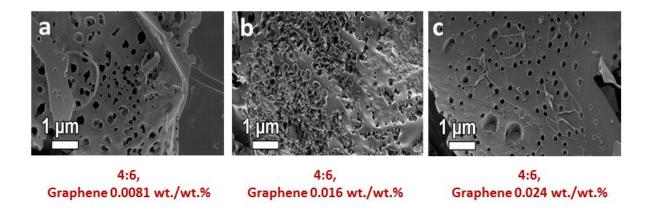


Figure S6. SEM images of carbon samples obtained after activation of three-component mixture of PVP:PVA (4:6) with graphene at varying mass proportion.

<sup>&</sup>lt;sup>b</sup> Single point adsorption total pore volume of pores less than 131 nm diameter at  $P/P_o = 0.98$ 

<sup>&</sup>lt;sup>c</sup> BJH Desorption average pore diameter (4V/A)

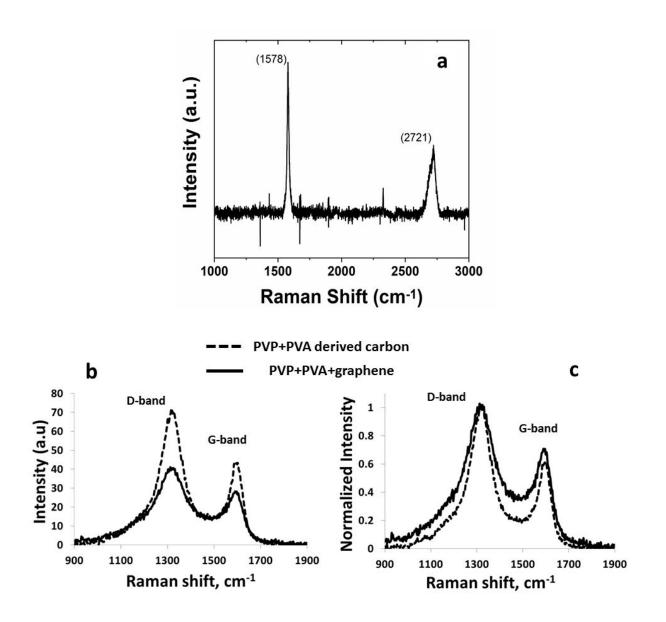


Figure S7. Raman spectra of (a) lab-scale prepared graphene in the range 1000 cm<sup>-1</sup> to 3000 cm<sup>-1</sup>, and (b) the carbon materials PVP+PVA and PVP+PVA+graphene in the wavenumber 900 cm<sup>-1</sup> to 1900 cm<sup>-1</sup> showing the D-band and G-band, (c) shows the comparison of D- and G-bands between two carbon samples with normalized y-axis.

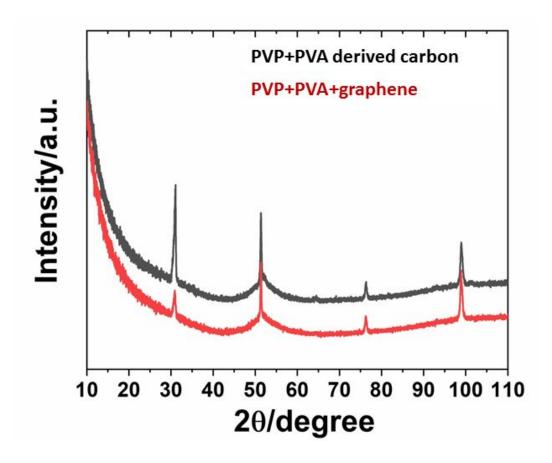


Figure S8. X-ray diffraction of carbon materials with and without graphene as labeled in the main text. XRD patterns for the synthesized samples match with graphite peaks. (JCPDS 01-089-7213) as amorphous sample.

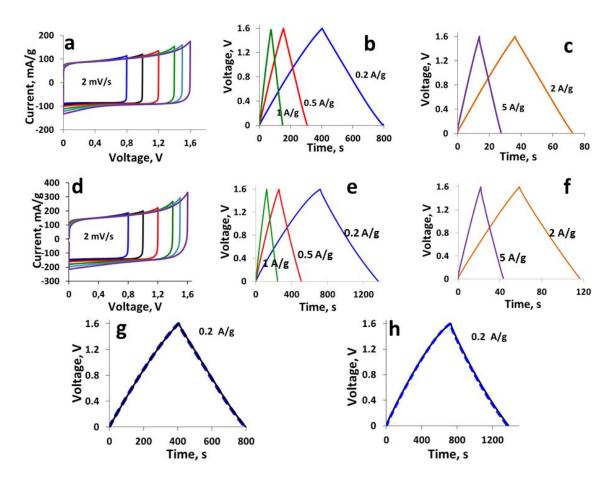


Figure S9. Electrochemical performance of symmetric supercapacitor using PVP+PVA (a-c) and PVP+PVA+graphene (d-f) based electrodes in 5 mol L<sup>-1</sup> NaNO<sub>3</sub> (a) cyclic voltammograms at 2 mV s<sup>-1</sup>, (b-c) galvanostatic charge/discharge curves at various specific currents from 0.2 A g<sup>-1</sup> to 5 A g<sup>-1</sup> (d) CVs at scan rates of 2 mV s<sup>-1</sup> up to 1.6 V, (e-f) galvanostatic charge/discharge curves at various specific currents from 0.2 A g<sup>-1</sup> to 5 A g<sup>-1</sup>, comparison of galvanostatic charge discharge curves (0.2 A g<sup>-1</sup>) for PVP+PVA (g) and PVP+PVA+graphene (h) based supercapacitors before (solid line) and after (dashed line) 10000 galvanostatic charge/discharge cycles up to 1.6 V.

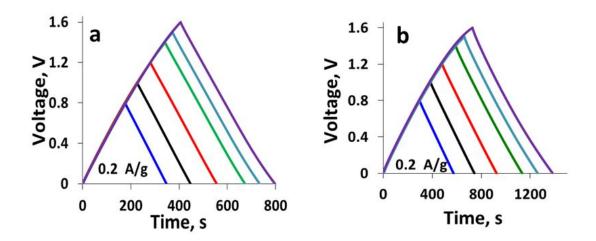


Figure S10. Galvanostatic charge/discharge curves (0.2 A  $g^{-1}$ ) of symmetric supercapacitors in 5 mol  $L^{-1}$  NaNO<sub>3</sub> up to various voltage from 0.8 V to 1.6 V for (a) PVP+PVA and (b) PVP+PVA+graphene.

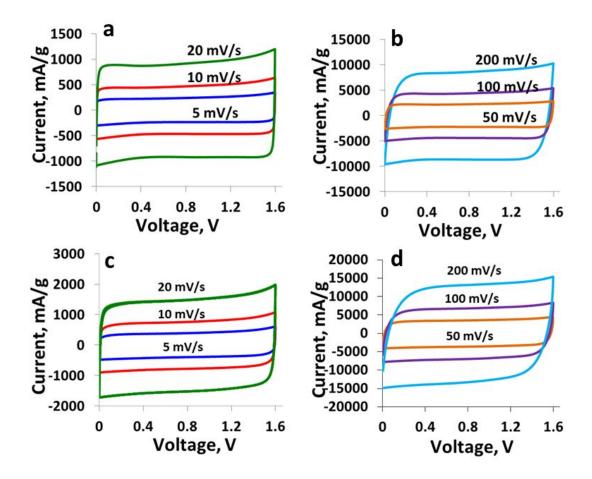


Figure S11. CV comparison of symmetric supercapacitor using PVP+PVA (a-b) and PVP+PVA+graphene (c-d) based electrodes in 5 mol  $L^{-1}$  NaNO<sub>3</sub> at different scan rates up to 200 mV s<sup>-1</sup>.

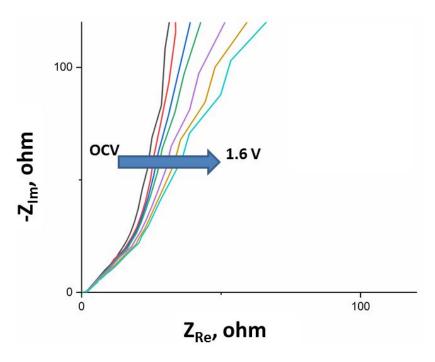


Figure S12. Nyquist plots at different voltage from open circuit voltage up to 1.6 V for symmetric supercapacitor using PVP+PVA based electrodes in 5 mol L<sup>-1</sup> NaNO<sub>3</sub> electrolyte.

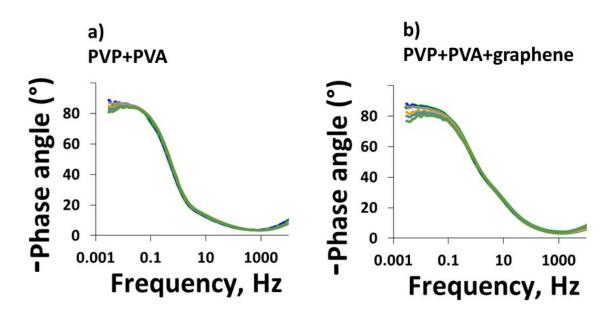


Figure S13. Phase angle versus frequency for carbon/carbon symmetric supercapacitors using (a) PVP+PVA based electrodes and (b) PVP+PVA+graphene based composite electrodes in 5 mol L<sup>-1</sup> NaNO<sub>3</sub>. The data is obtained at various voltage steps from OCV to 1.6 V.

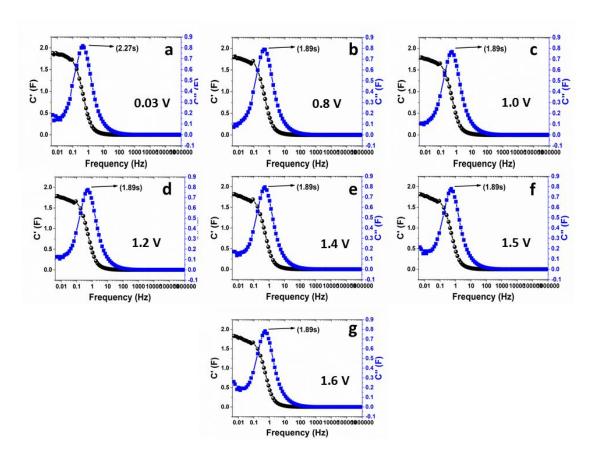


Figure S14. Evolution of real (black circles) and imaginary (blue squares) capacitance versus frequency at various voltage steps from OCV to 1.6 V for the carbon/carbon supercapacitor using PVP+PVA based electrodes in 5 mol L<sup>-1</sup> NaNO<sub>3</sub>.

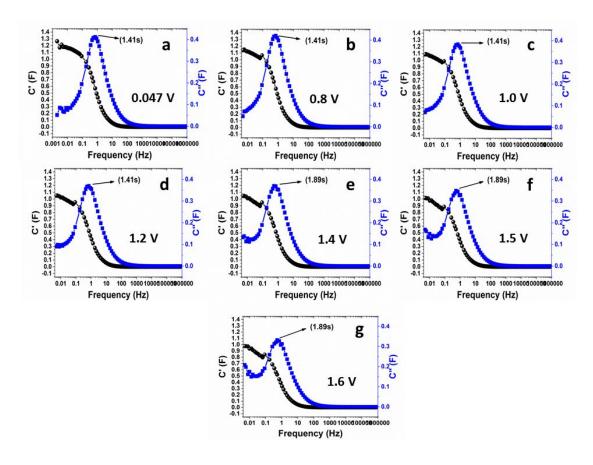


Figure S15. Evolution of real (black circles) and imaginary (blue squares) capacitance versus frequency at various voltage steps from OCV to 1.6 V for the carbon/carbon supercapacitor using PVP+PVA+graphene based electrodes in 5 mol L<sup>-1</sup> NaNO<sub>3</sub>.

Carbon material	Electrolyte	Gravimetric capacitance (F/g)	Energy density Wh/kg	Power density W/kg	Areal capacitance (F/cm <sup>2</sup> )	Areal Energy density Wh/cm <sup>2</sup>	Areal Power density W/cm²
PVP+PVA	5 mol L <sup>-1</sup> NaNO <sub>3</sub>	96	8	2000	191	0.017	0.02
PVP+PVA +graphene	5 mol L <sup>-1</sup> NaNO <sub>3</sub>	163	12	2000	324	0.115	0.02

Table S2. Summary of electrochemical performance of the symmetric supercapacitor devices using PVP+PVA and PVP+PVA+graphene based electordes in 5 mol L-1 NaNO<sub>3</sub>.