

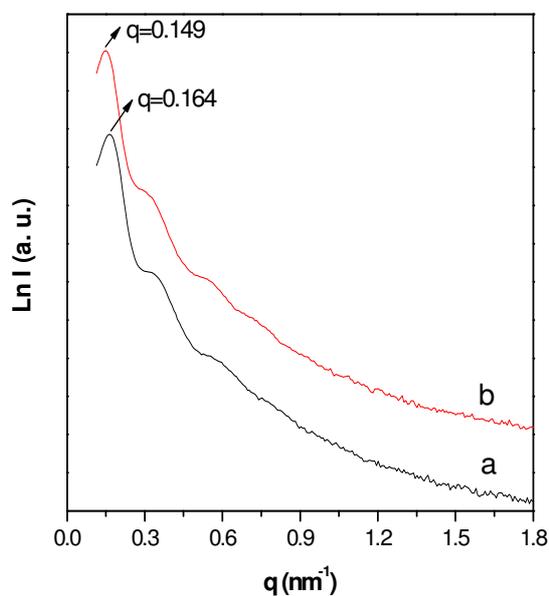
## Supporting Information

### Ultra-Large Pore Mesoporous Carbons Templated from Poly(ethylene oxide)-*b*-Polystyrene Diblock Copolymer by Adding Polystyrene Homopolymer as a Pore Expander

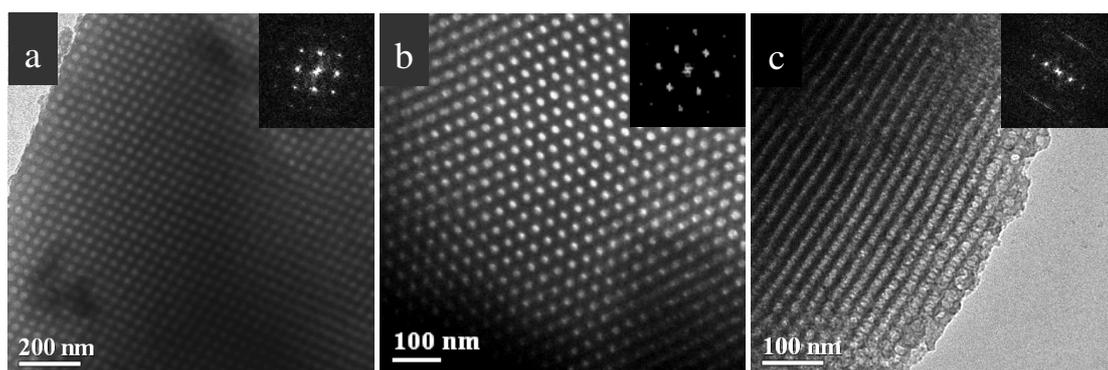
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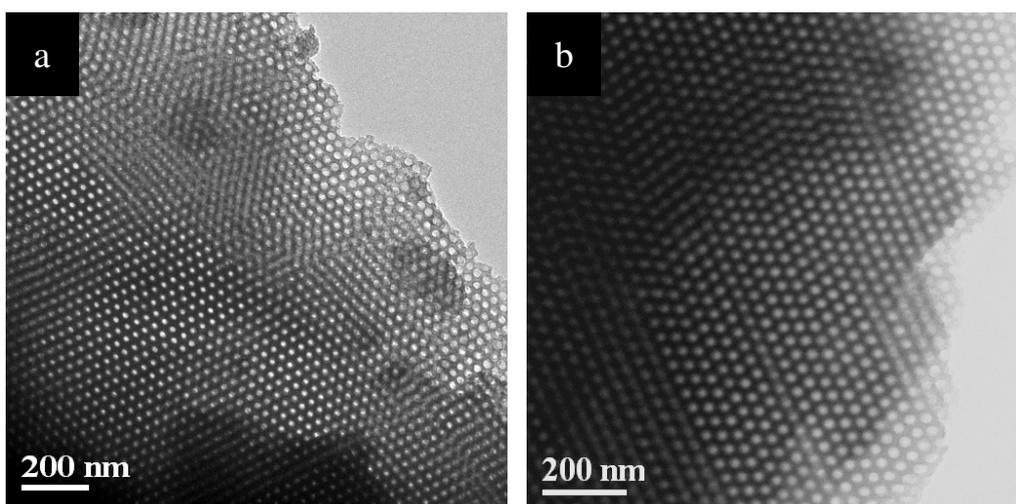
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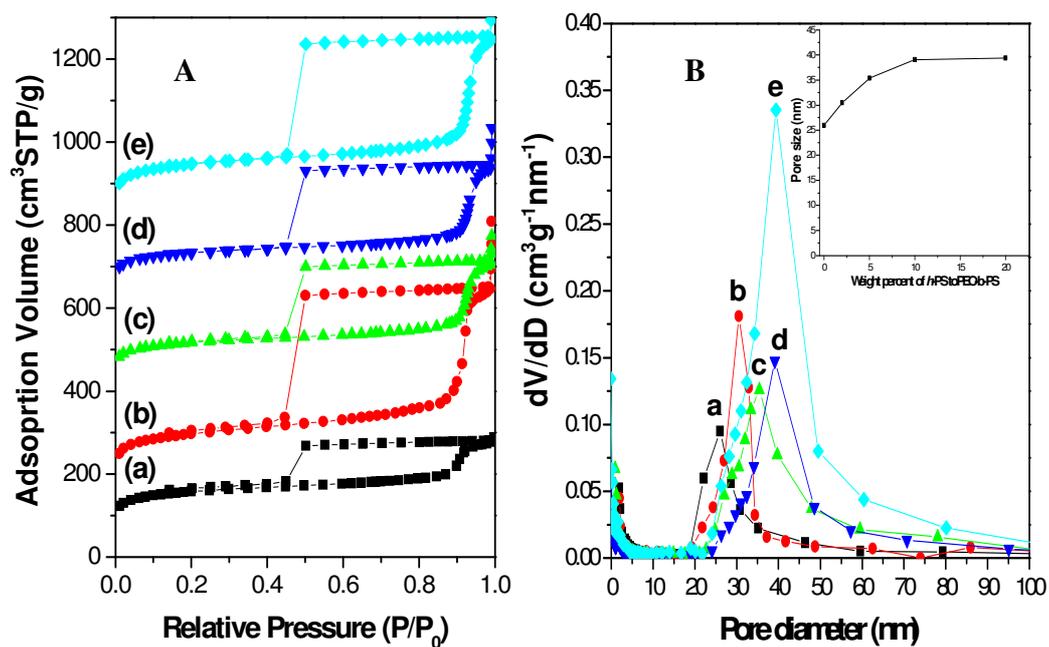
**Figure S1.** SAXS patterns of (a) LP-FDU-18-25-450 and (b) LP-FDU-18-35-450 prepared by adding excessive *h*-PS<sub>49</sub> (25 and 35 wt%, respectively) after pyrolysis at 450 °C in N<sub>2</sub>.



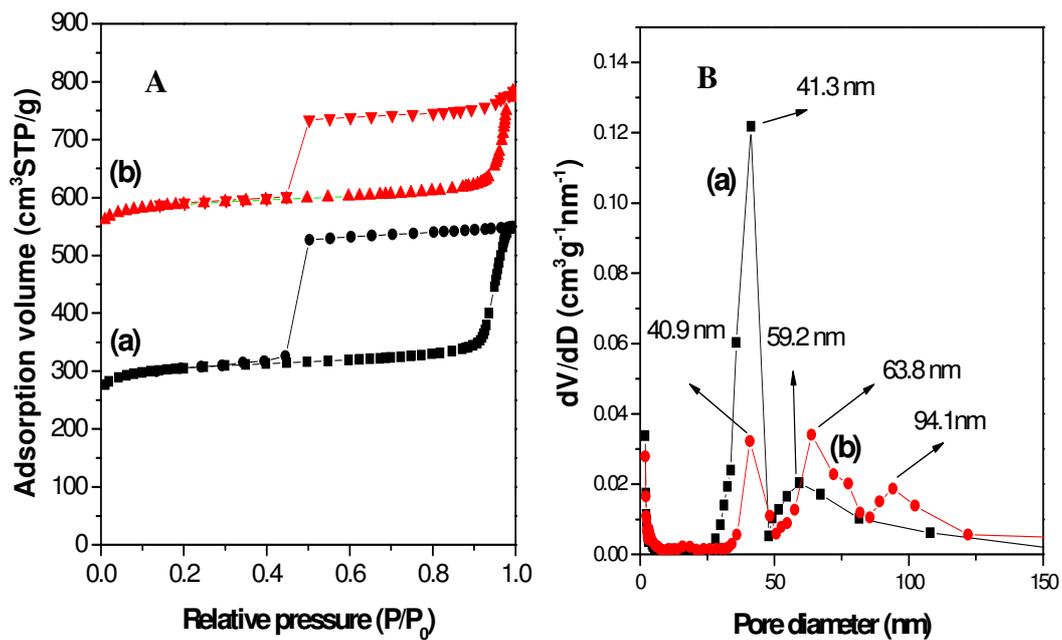
**Figure S2.** TEM images of LP-FDU-18-0-800 sample prepared without adding  $h$ -PS<sub>49</sub> viewed from (a) 100, (b) 110 and (c) 211 directions. The insets show the corresponding FFT diffractometers.



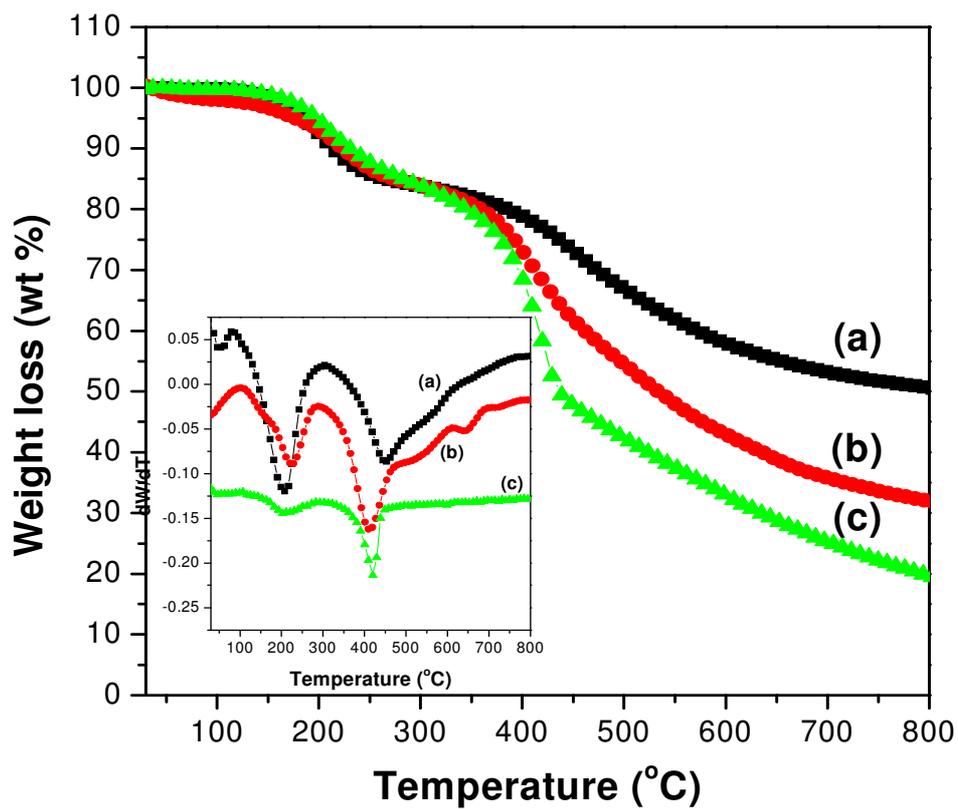
**Figure S3.** TEM images of FDU-18-20-800 sample prepared by adding 20 wt% *h*-PS<sub>49</sub> as pore expander after carbonization at 800 °C under N<sub>2</sub>, indicating the existence of structure defects.



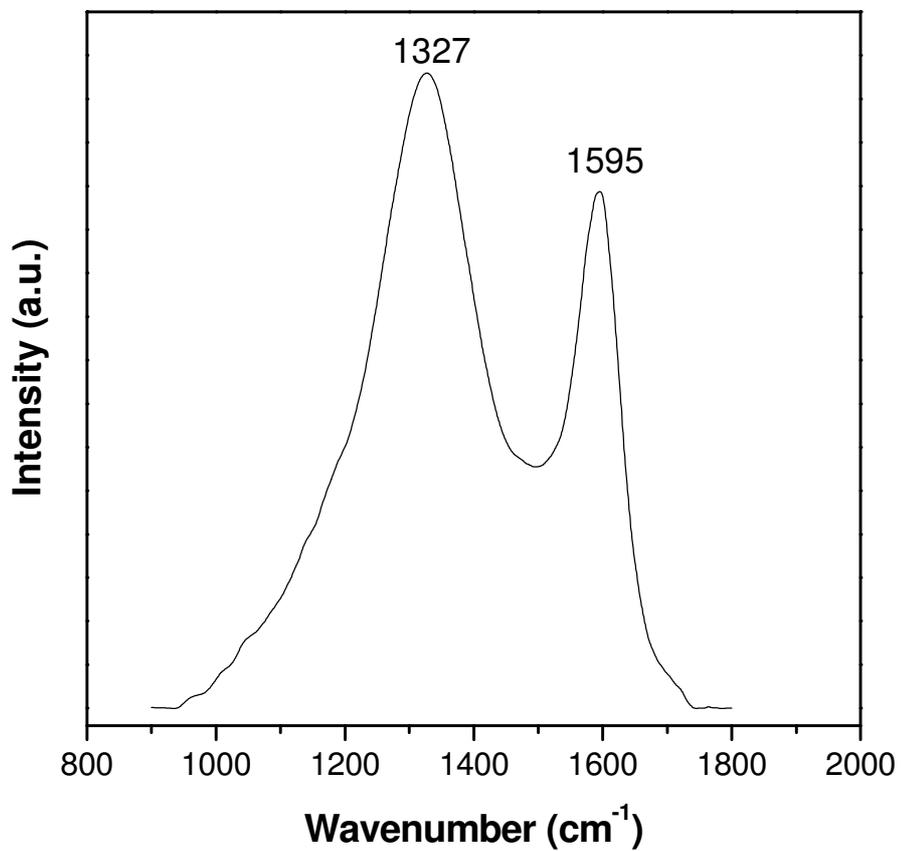
**Figure S4.** (A) N<sub>2</sub> isotherms and (B) pore size distribution of (a) LP-FDU-18-0-450, (b) LP-FDU-18-2-450, (c) LP-FDU-18-5-450, (d) LP-FDU-18-10-450 and (e) LP-FDU-18-20-450. The N<sub>2</sub> isotherms of b, c, d and e are offset vertically by 100, 300, 500 and 700 cm<sup>3</sup>/g, respectively. The inset shows the pore size changes of the LP-FDU-18-450 samples upon the increase of *h*-PS<sub>49</sub> addition amount.



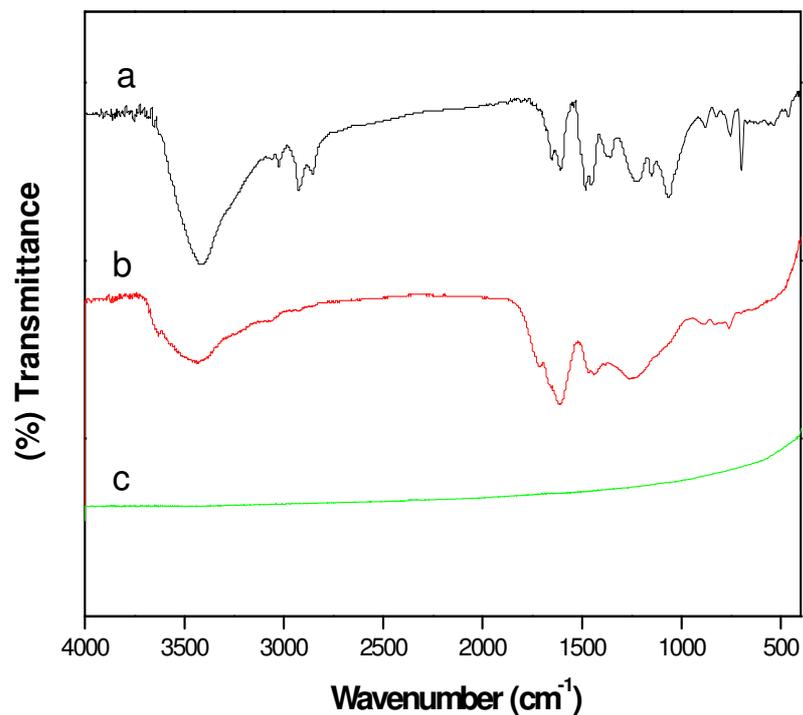
**Figure S5.** (A)  $N_2$  isotherms and (B) pore size distribution of (a) LP-FDU-18-25-800, (b) LP-FDU-18-35-800 prepared by adding excessive  $h$ -PS<sub>49</sub> (25 and 35 wt%, respectively) after carbonization at 800 °C under  $N_2$ . The  $N_2$  isotherms of b are offset vertically by 300  $cm^3/g$ , respectively.



**Figure S6.** TGA and DTG curves of (a) phenolic formaldehyde (PF), (b) as-made LP-FDU-18 without addition of *h*-PS<sub>49</sub>, and (c) as-made LP-FDU-18-20 with 20 wt% addition of *h*-PS<sub>49</sub>. The pyrolysis processes were conducted in N<sub>2</sub> with a heating rate of 5 °C /min.



**Figure S7.** Raman spectra of LP-FDU-18-20-800 prepared by adding 20 wt% *h*-PS<sub>49</sub> after carbonization at 800 °C under N<sub>2</sub>, showing G band at 1595 cm<sup>-1</sup> and D band at 1327 cm<sup>-1</sup>. It suggests that the obtained ordered mesoporous carbons have a low degree of graphitization.



**Figure S8.** FT-IR spectra of as-made LP-FDU-18-20 (a), LP-FDU-18-20-450 (b) and LP-FDU-18-20-800 (c). In the curve (a), the strong and broad band at  $3400\text{ cm}^{-1}$  is associated with the phenolic  $-\text{OH}$  groups, and the bands at  $1100$  and  $2800\text{ cm}^{-1}$  are assigned to PEO-*b*-PS and *h*-PS. The disappearance of characteristic bands at  $1109\text{ cm}^{-1}$  associated with PEO block and at  $2850\text{-}2930\text{ cm}^{-1}$  assigned to PS block in curve (b) suggests the decomposition of PEO-*b*-PS template and *h*-PS by pyrolysis in  $\text{N}_2$ . For curve (c), no absorption was observed, suggesting a carbon framework by pyrolysis at higher temperature.