Supporting Information

Bulk Phosphorus-Doped Graphitic Carbon

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Contents:

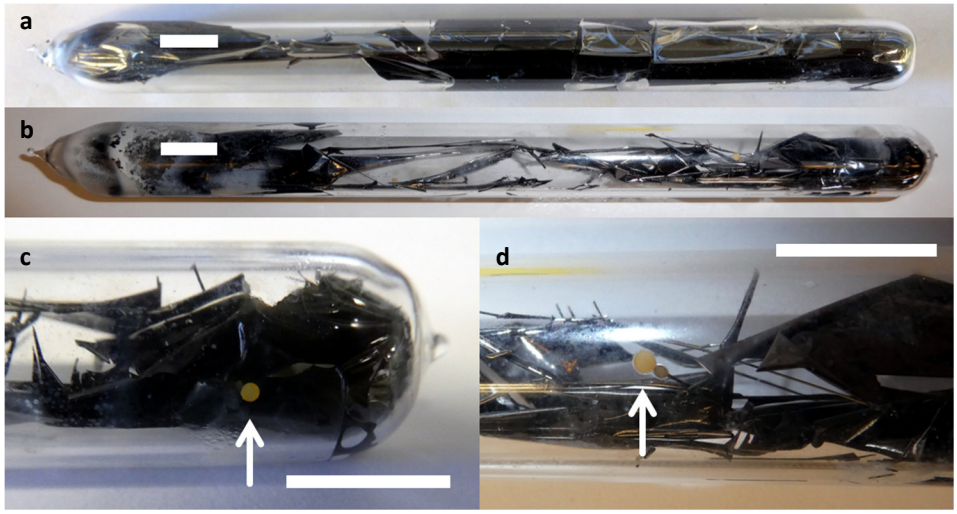

Figure S1. Photographs of (a) graphitic PC_3 synthesized at 800 °C, and (b-d) graphitic PC synthesized at 1050 °C showing large yellow liquid droplets of white phosphorus (P_4). All scale bars correspond to 1 cm.

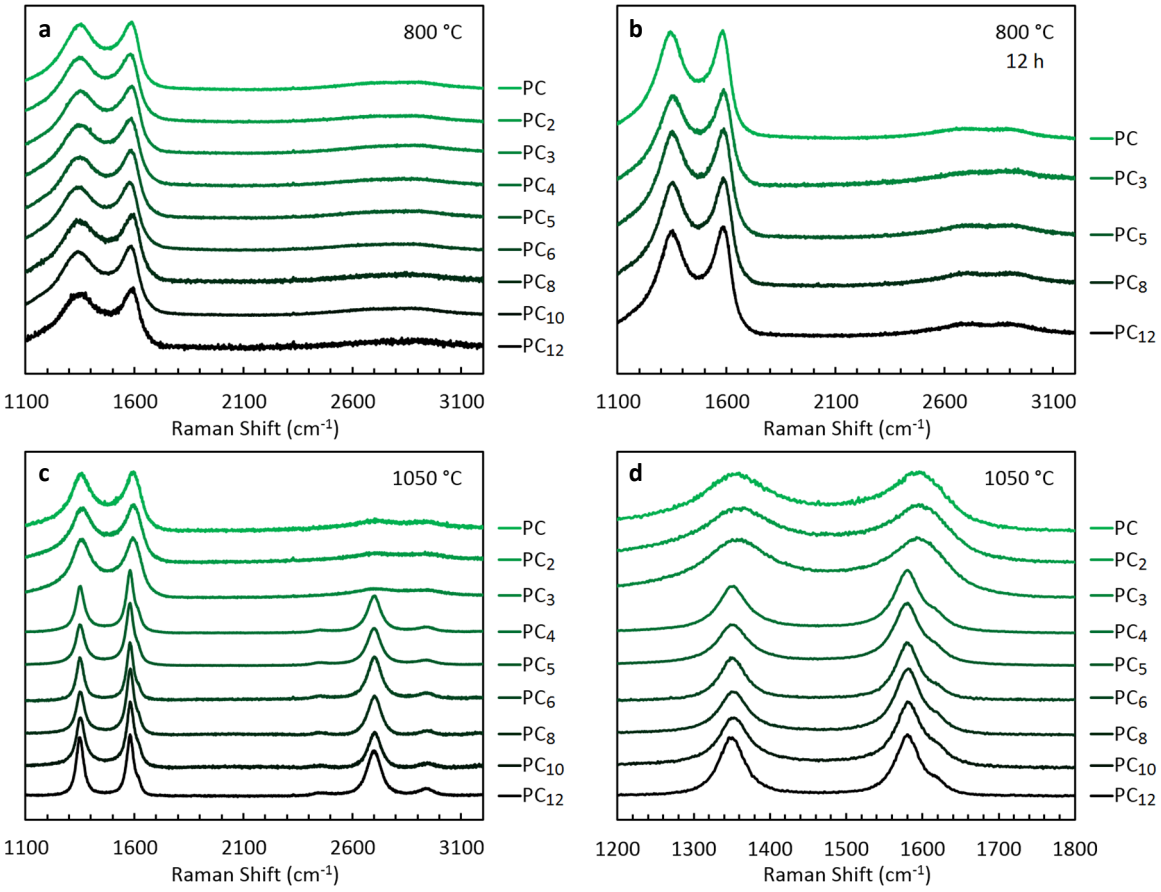

Figure S2. Raman spectra of graphitic PC_x synthesized at (a) 800 °C (for 1 h at setpoint), (b) 800 °C (for 12 h at setpoint), and (c-d) 1050 °C (for 1 h at setpoint).

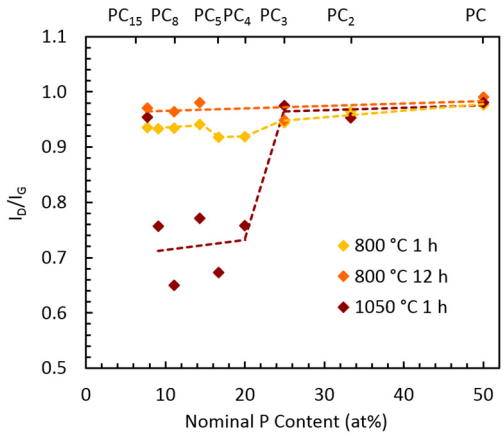

Figure S3. Variation of I_D/I_G measured by Raman spectroscopy of graphitic PC_x synthesized at 800 and 1050 °C (for 1 h or 12 h at setpoint).

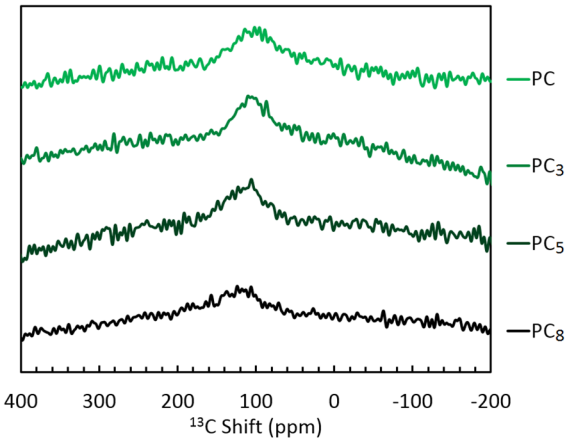

Figure S4. ^{13}C MAS NMR spectra of graphitic PC_x synthesized at 1050 °C, acquired with a 30° single pulse sequence.

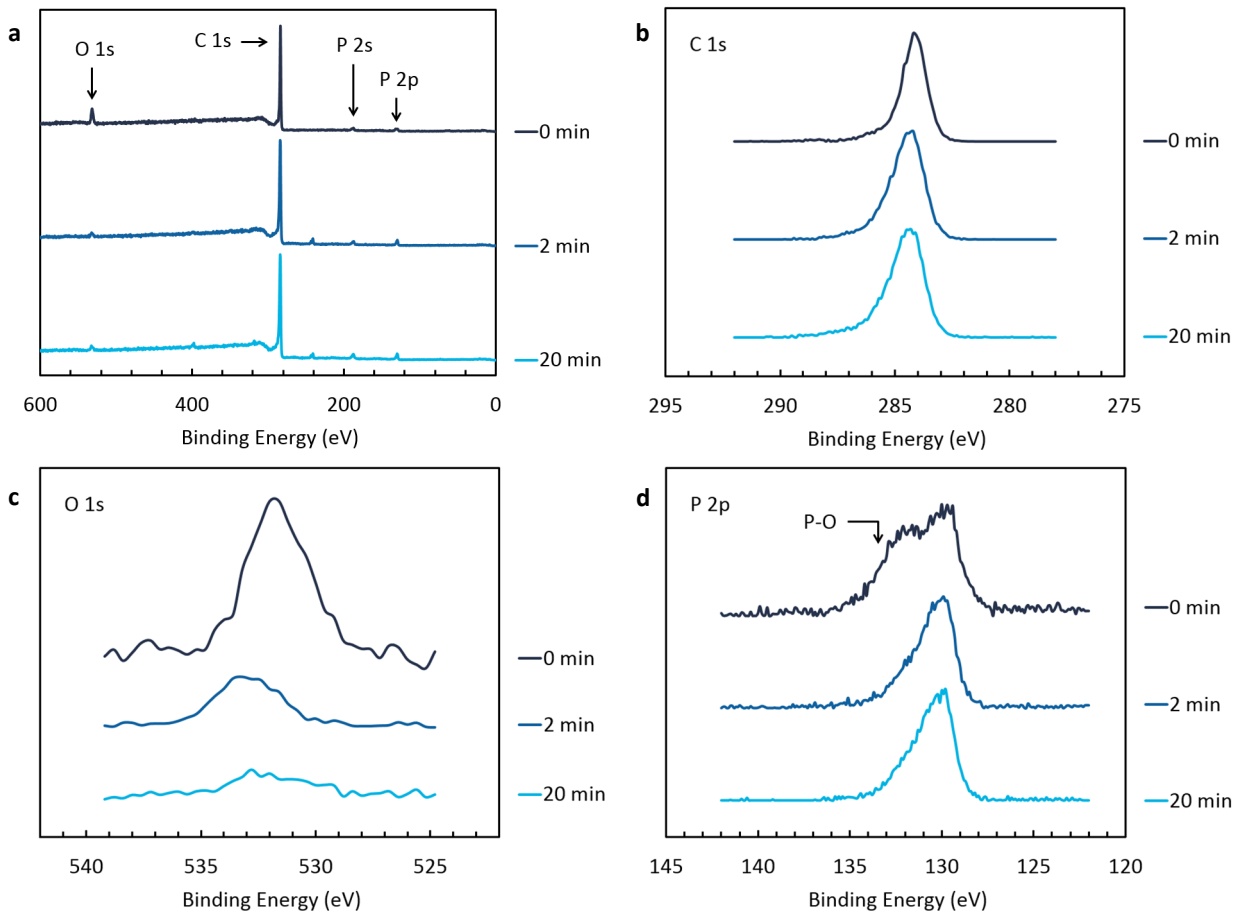

Figure S5. XPS depth-profile analysis of graphitic PC_5 synthesized at 800 °C. (a) Survey spectra and (b-d) detail regions were measured at three depths: after 0 min sputtering (dark blue), 2 min sputtering (blue), and 20 min sputtering (light blue).

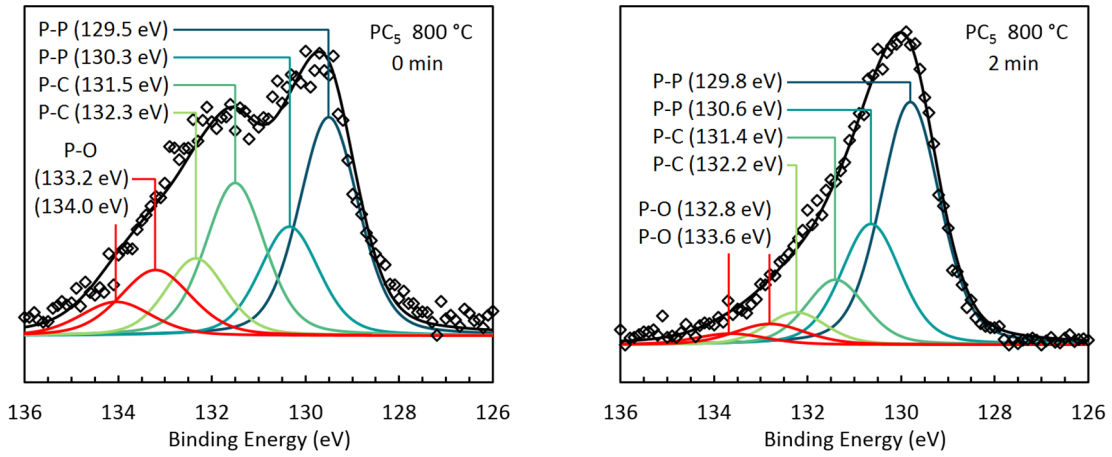

Figure S6. XPS depth-profile analysis of graphitic PC_5 synthesized at 800 °C (as in Figures 7 and S5), showing detailed fitting results after 0 min sputtering (left) and 2 min sputtering (right).

Table S1. XPS peak fitting analysis of *directly-synthesized* PC_5 (as shown in Figures 7, S5-S6) as a function of depth below the material surface (as indicated by sputtering time) by percentage peak area (peak identity and position as indicated)

Peak (Position)	0 min Sputtering	2 min Sputtering	20 min Sputtering
P–P $p_{3/2}$ (~129.8 eV)	32.3%	48.5%	47.7%
P–P $p_{1/2}$ (~130.6 eV)	16.1%	24.2%	23.8%
P–C $p_{3/2}$ (~131.4 eV)	22.5%	13.0%	16.3%
P–C $p_{1/2}$ (~132.2 eV)	11.4%	6.6%	8.1%
P–O $p_{3/2}$ (~133 eV)	11.7%	5.0%	2.7%
P–O $p_{1/2}$ (~134 eV)	6.0%	2.6%	1.4%

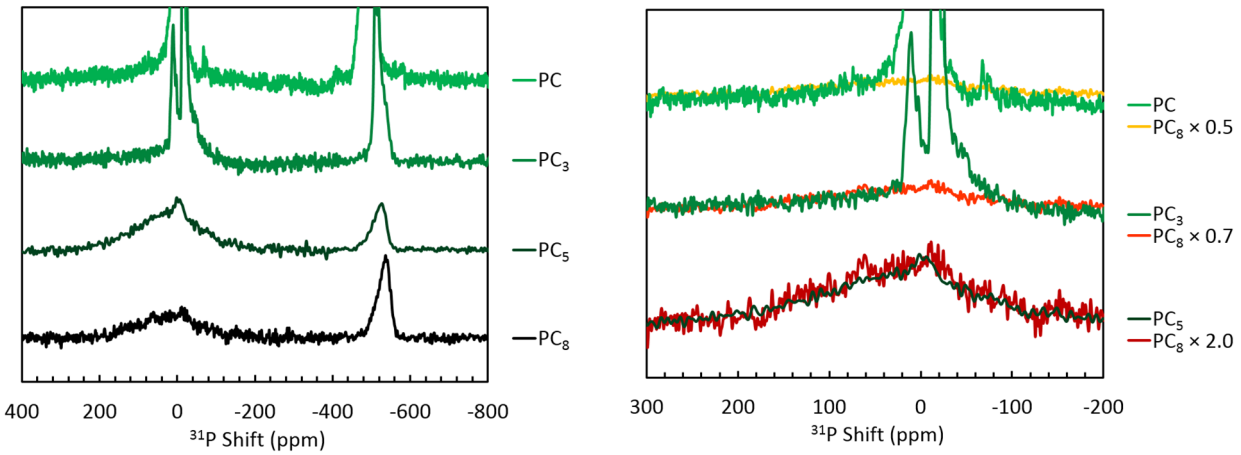

Figure S7. Detail of ^{31}P MAS NMR spectra of PC, PC_3, and PC_5 compared to PC_8 (which shows the minimum contribution from phosphates), all synthesized at 1050 °C, as shown in Figure 5.

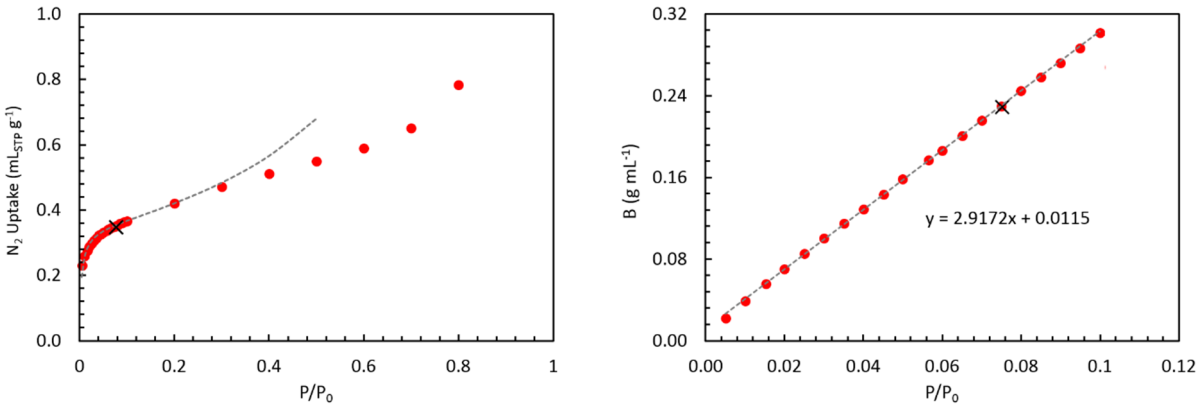

Figure S8. Equilibrium adsorption uptake of N_2 at 77 K on graphitic PC_5 synthesized at 800 °C (left) and corresponding BET plot (right) showing the best fit results (gray dashed line, indicating a BET surface area of 1.49 $m^2\ g^{-1}$).