

# **Porous Carbon Nano-sheets as Excellent Carbocatalysts for Organic Pollutants Removal via Persulfate Activation: The Role of $sp^2/sp^3$ Carbon Ratio**

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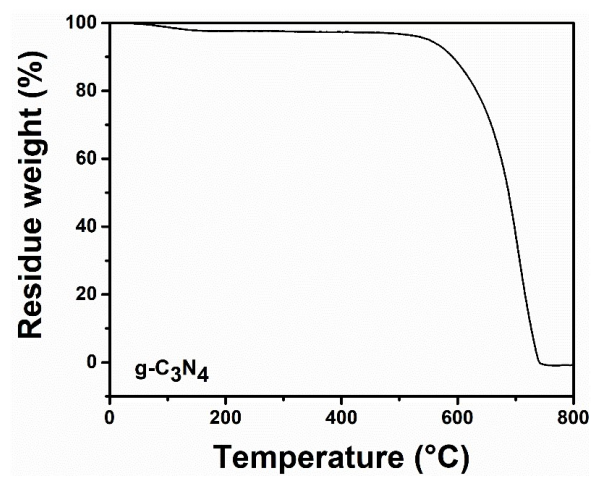
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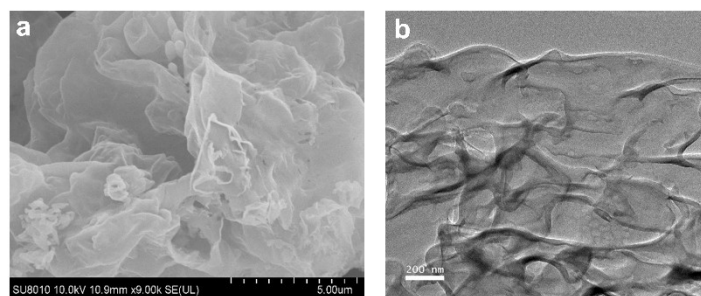
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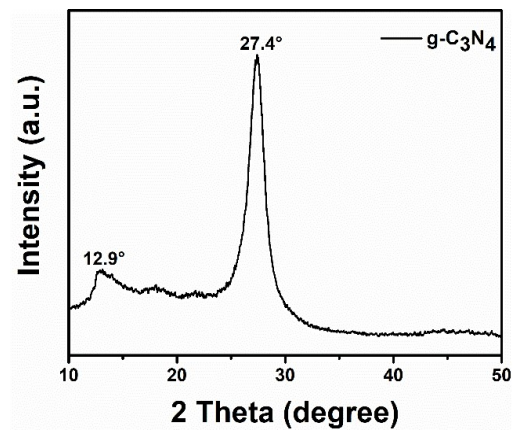
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**Fig. S1** TGA of g-C<sub>3</sub>N<sub>4</sub> in N<sub>2</sub> with a temperature rise rate of 10 °C min<sup>-1</sup>.



↵ **Fig. S2** (a) SEM image of pristine g-C<sub>3</sub>N<sub>4</sub>. (b) TEM image of pristine g-C<sub>3</sub>N<sub>4</sub>.



**Fig. S3** XRD pattern of g-C<sub>3</sub>N<sub>4</sub>.

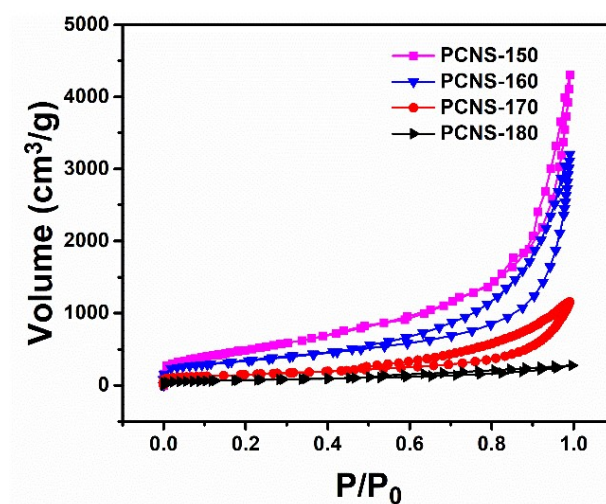
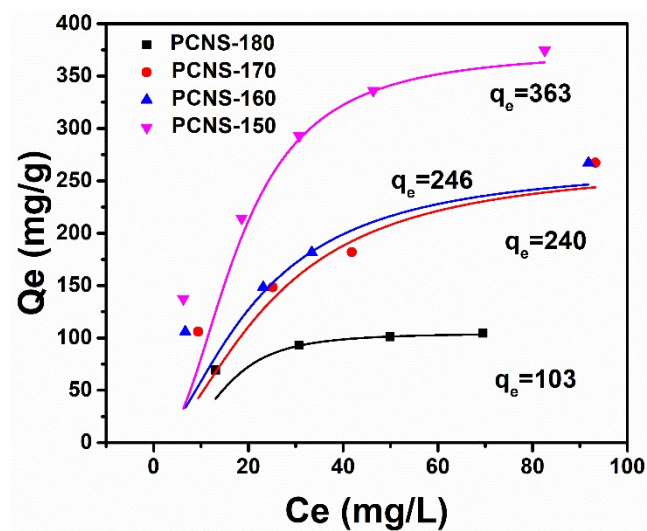
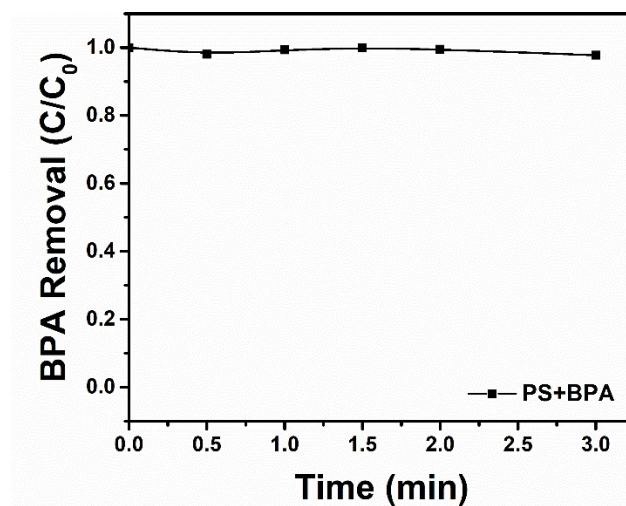


Fig. S4 N<sub>2</sub> adsorption/desorption isotherms.

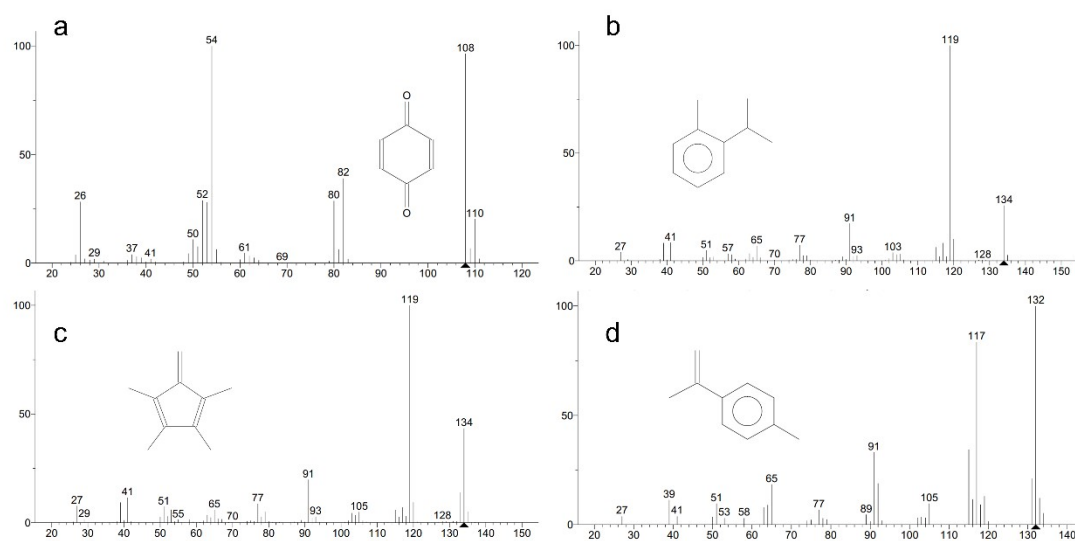


**Fig. S5** The fitting curves of Langmuir model for the adsorption isotherms of BPA on the PCNSs.

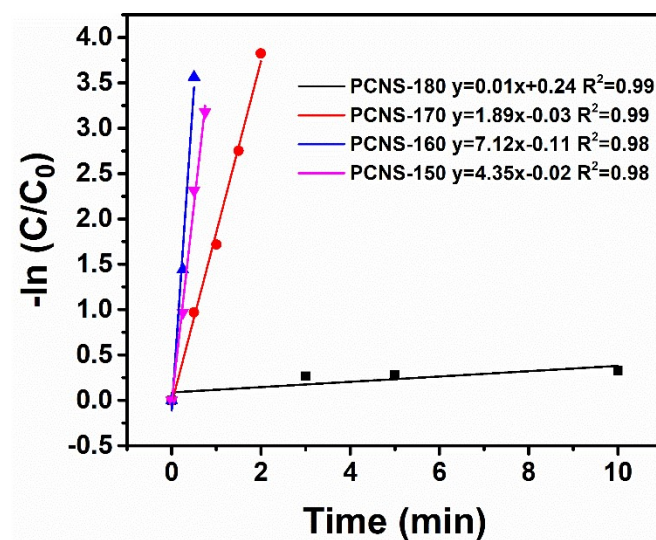
$C_e$  ( $\text{mg L}^{-1}$ ) is the concentration of equilibrium adsorption in the solution, and  $Q_e$  ( $\text{mg g}^{-1}$ ) is the equilibrium adsorption capacity.



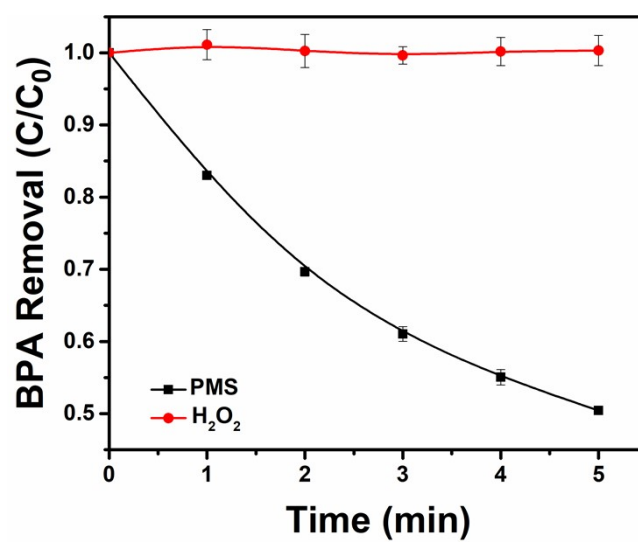
**Fig. S6** Degradation curve of BPA in the presence of PS. [PS] = 1.5 mM; [BPA] = 80 mg/L.



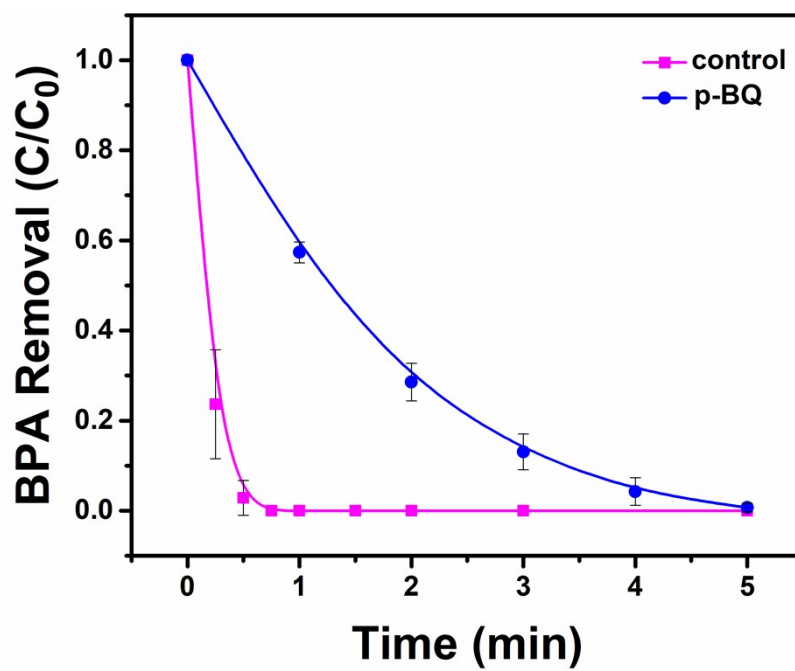
**Fig. S7** GC-MS spectra of the BPA degradation intermediates.



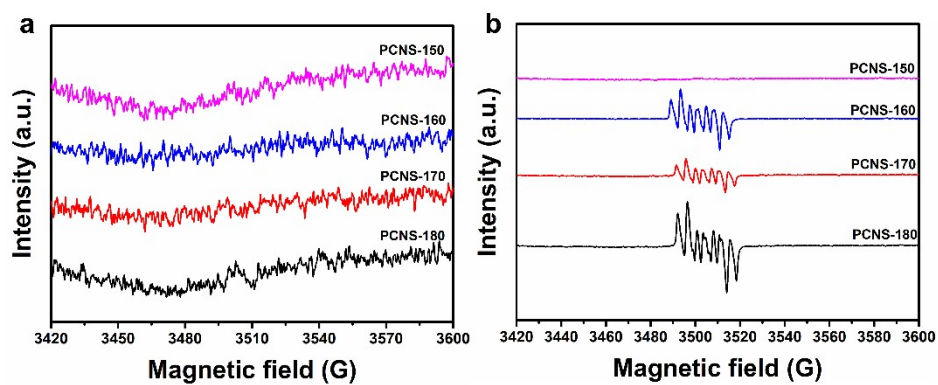
**Fig. S8** The kinetic analyses of BPA degradation to pseudo-first-order model.



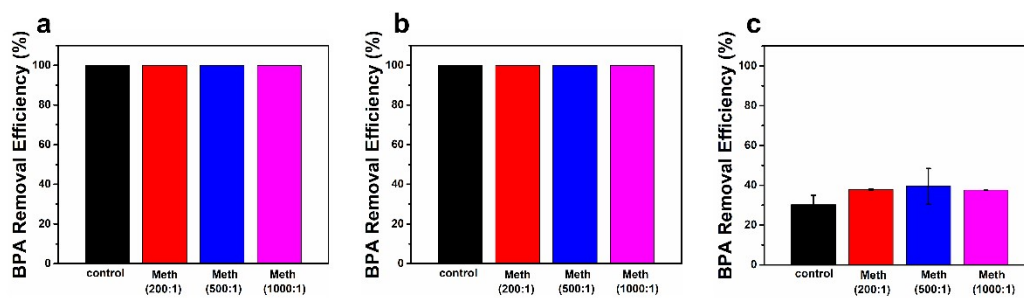
**Fig. S9** The effects of different oxidants on BPA degradation [PCNS] = 0.1 g/L; [PMS] = [H<sub>2</sub>O<sub>2</sub>] = 1.5 mM; [BPA] = 80 mg/L.



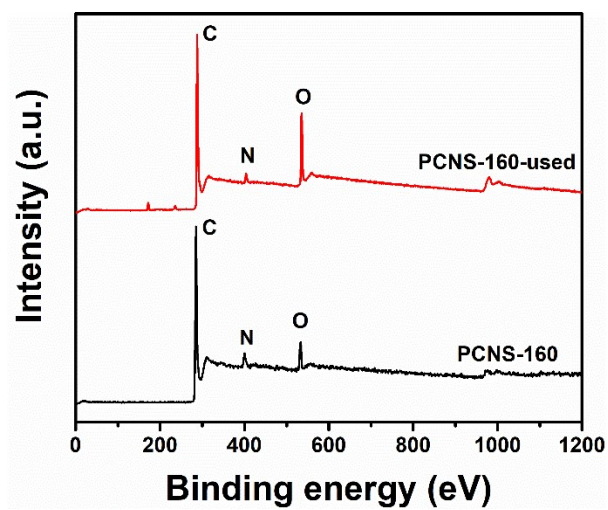
**Fig. S10** The effects of p-BQ on BPA degradation [PCNS-160] = 0.1 g/L; [PS] = 1.5 mM; [p-BQ] = 7.5 mM [BPA] = 80 mg/L.



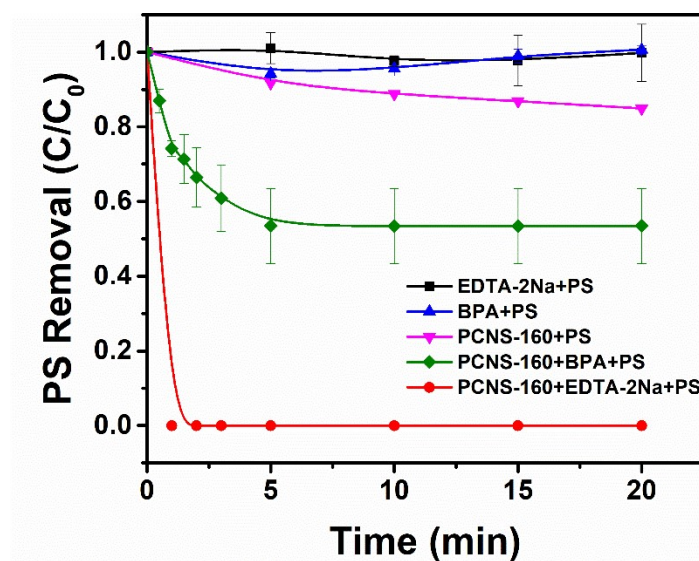
**Fig. S11** EPR spectra obtained by spin trapping with TEMP and DMPO in the presence of PS and PCNSs. [PCNS] = 0.1 g/L; [PS] = 1.5 mM; [BPA] = 80 mg/L; [DMPO/TEMP] = 10 mM.



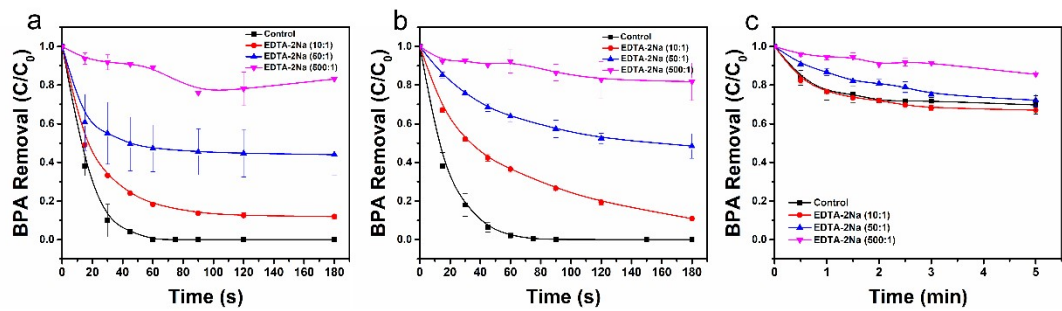
**Fig. S12** Effects of methanol on BPA removal in (a)PCNS-150/PS system, (b) PCNS-170/PS system and (c) PCNS-180/PS system. [PCNS] = 0.1 g/L; [PS] = 1.5 mM; [BPA] = 80 mg/L.



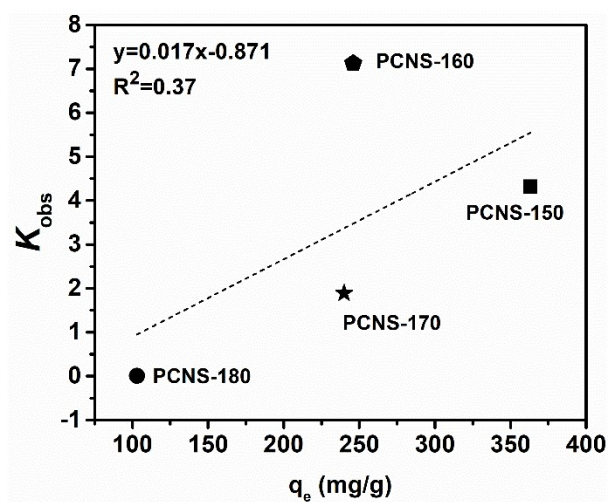
**Fig. S13** XPS survey of PCNS-160 and PCNS-160-used.



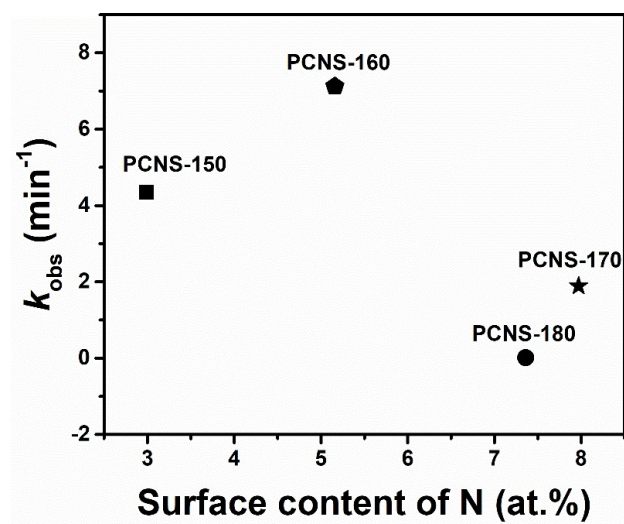
**Fig. S14** Consumption curves of PS.  $[PS] = 1.5 \text{ mM}$ ;  $[PCNS-160] = 0.1 \text{ g/L}$ ;  $[BPA/EDTA-2Na] = 0.36 \text{ mM}$ .



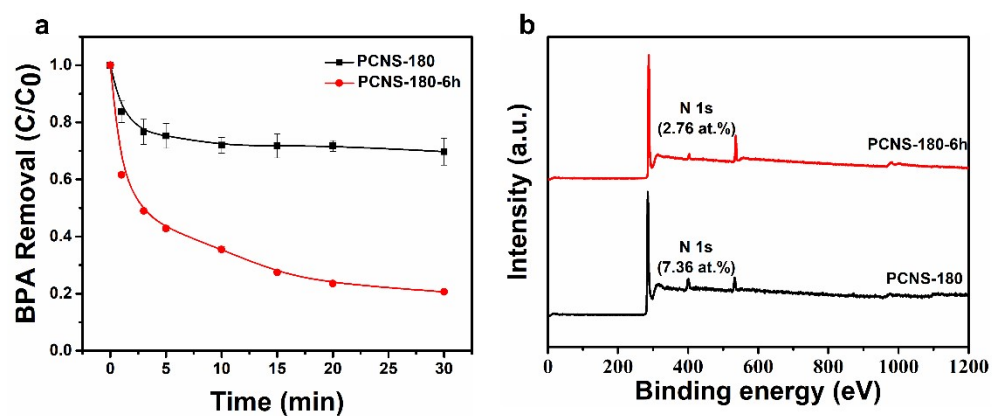
**Fig. S15** Effects of EDTA-2Na on BPA removal in (a)PCNS-150/PS system, (b) PCNS-170/PS system, (c) PCNS-180/PS system. [PCNS] = 0.1 g/L; [PS] = 1.5 mM; [BPA] = 80 mg/L.



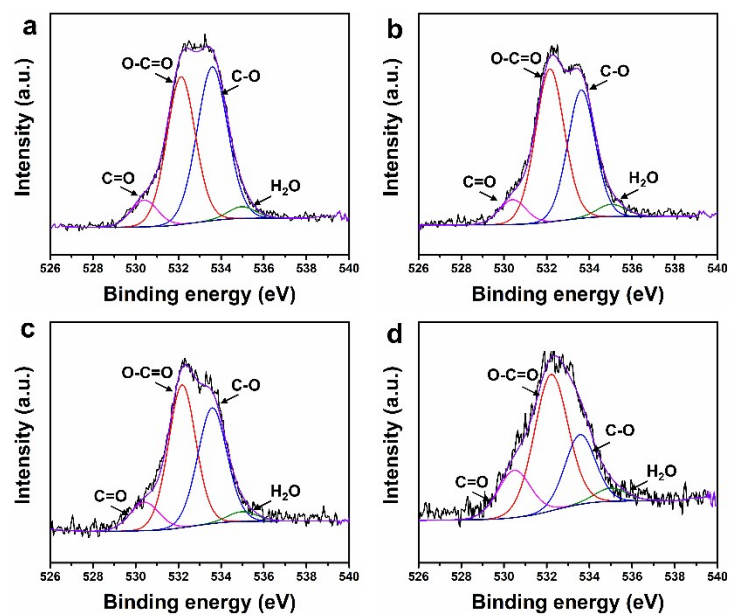
**Fig. S16.** Fitted curve of  $k_{obs}$  and the equilibrium adsorption capacities of BPA by PCNSs.



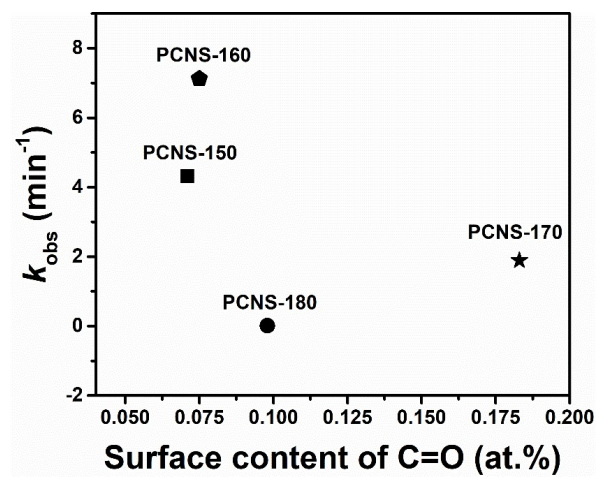
**Fig. S17.** Variation of  $k_{\text{obs}}$  and the surface content of N.



**Fig. S18.** (a) BPA degradation curve, (b) XPS survey of PCNS-180 and PCNS-180-6h. [PCNS] = 0.1 g/L; [PS] = 1.5 mM; [BPA] = 80 mg/L.



**Fig. S19** High resolution XPS spectra of O 1s for (a) PCNS-150, (b) PCNS-160, (c) PCNS-170, and (d) PCNS-180.



**Fig. S20** Variation of  $k_{\text{obs}}$  and the surface content of C=O.

**Table S1.** The BET surface area, total pore volume and XPS results of all samples

| Sample   | $S_{\text{BET}}$<br>m <sup>2</sup> /g | total pore volume<br>cm <sup>3</sup> /g | XPS survey |       |       |
|----------|---------------------------------------|-----------------------------------------|------------|-------|-------|
|          |                                       |                                         | C          | N     | O     |
|          |                                       |                                         | at. %      | at. % | at. % |
| PCNS-150 | 1597                                  | 6.67                                    | 88.45      | 2.99  | 8.55  |
| PCNS-160 | 1297                                  | 5.16                                    | 88.68      | 5.16  | 6.16  |
| PCNS-170 | 469                                   | 1.80                                    | 89.76      | 7.97  | 2.27  |
| PCNS-180 | 250                                   | 0.43                                    | 88.59      | 7.36  | 4.06  |

**Table S2.** The pseudo-first order kinetic constants ( $k_{\text{obs}}$ ) of PCNS/PS system

| Sample   | $k_{\text{obs}}$       | $R^2$ |
|----------|------------------------|-------|
| PCNS-150 | 4.35 min <sup>-1</sup> | 0.98  |
| PCNS-160 | 7.12 min <sup>-1</sup> | 0.98  |
| PCNS-170 | 1.89 min <sup>-1</sup> | 0.99  |
| PCNS-180 | 0.01 min <sup>-1</sup> | 0.99  |

**Table S3.** Comparison of catalytic performances of PCNS-160 and state-of-the-art catalysts for PS or PMS activation

| Study     | Catalyst         | Oxidant     | Substrate  | $k_{\text{obs}}$ (min <sup>-1</sup> ) |
|-----------|------------------|-------------|------------|---------------------------------------|
| This work | PCNS-160 0.1 g/L | PS 1.5 mM   | BPA 80 ppm | 7.12                                  |
| [1]       | HCCNs 0.1 g/L    | PS 1.5 mM   | BPA 20 ppm | 0.32                                  |
| [2]       | NCN-900 0.1 g/L  | PS 2 mM     | BPA 20 ppm | 3.10                                  |
| [3]       | NS-2 0.1 g/L     | PS 1.5 mM   | BPA 20 ppm | 0.18                                  |
| [4]       | HOPC 0.2 g/L     | PS 2 mM     | BPA 10 ppm | 0.25                                  |
| [5]       | CNT 0.1 g/L      | PS 2 mM     | BPA 20 ppm | 0.04                                  |
| [6]       | G-ND 0.1 g/L     | PS 1.5 mM   | BPA 20 ppm | 0.85                                  |
| [7]       | SiM-1 0.4 g/L    | PMS 0.27 mM | BPA 10 ppm | 0.19                                  |
| [8]       | ZVI 0.45 g/L     | PS 0.5 mM   | BPA 5 ppm  | 0.04                                  |
| [9]       | Fe-Chitoal 1 g/L | PS 1 mM     | BPA 10 ppm | 2.11                                  |

**Table S4.** XPS analyses of PCNS-160 and PCNS-160-used

| Sample        | C     | N     | O     | S     |
|---------------|-------|-------|-------|-------|
|               | at. % | at. % | at. % | at. % |
| PCNS-160      | 88.68 | 5.16  | 6.16  | 0     |
| PCNS-160-used | 79.92 | 3.60  | 14.65 | 1.84  |

**Table S5.** The relative atomic percentage of various oxygen functional groups

| Sample   | O<br>at. % | Fitting of the O 1s peak binding energy |       |       |                                 |
|----------|------------|-----------------------------------------|-------|-------|---------------------------------|
|          |            | C=O                                     | O-C=O | C-O   | H <sub>2</sub> O <sub>ads</sub> |
|          |            | at. %                                   | at. % | at. % | at. %                           |
| PCNS-150 | 8.55       | 0.071                                   | 0.424 | 0.472 | 0.033                           |
| PCNS-160 | 6.16       | 0.075                                   | 0.500 | 0.400 | 0.025                           |
| PCNS-170 | 2.27       | 0.183                                   | 0.538 | 0.247 | 0.032                           |
| PCNS-180 | 4.06       | 0.098                                   | 0.467 | 0.402 | 0.033                           |

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