Supporting Information:

High reaction activity enables carbon dots to construct multicomponent nanocomposites with superior catalytic performance

Shijia Li,[a] Ernan Pang,[a] Ning Li,[a] Qing Chang,[a] Jinlong Yang,[a,b] Shengliang Hu[a]*

[a] *Research Group of New Energy Materials and Devices, North University of China, Taiyuan 030051, P. R. China.*

*Corresponding author, E-mail: hsliang@yeah.net

[b] *State Key Laboratory of New Ceramics and Fine Processing, Tsinghua University, Beijing 100084, P. R. China.*

1. Experimental details

1.1 Materials and reagents

Ferrous sulfate heptahydrate($FeSO_4 \cdot 7H_2O$), formic acid (88 v/v%), hydrogen peroxide (H_2O_2, 30 v/v%), ammonia solution(25 v/v%), sodium hydroxide (NaOH), hydrochloric acid (HCl), sulfuric acid (H_2SO_4), sodium persulfate ($Na_2S_2O_8$, PS), terephthalic acid (TA), Tert butyl alcohol (TBA) and methanol (MeOH) were obtained from Aladdin Reagent Co., Ltd. (Shanghai, China). All the chemicals were directly used without further purification if not elsewise specified in our experiments. All aqueous solutions in the experiment were prepared with ultrapure water.

1.2 Analytical methods

The X-ray diffraction (XRD) patterns of the samples were performed using a Bruker D8 ADVANCE powder diffractometer at Cu Kα radiation with a scanning rate of 8° min^{-1} in the 2θ range of 10–90°. TEM, HRTEM and elemental distribution mapping images were conducted on a FEI Tecnai G2 F20 microscope with a field-emission gun operating at 200kV. XPS data of all samples were collected by a Kratos AXIS 165 mutitechnique electron spectrometer with an Al Ka X-ray source. The specific surface areas of the samples were determined at 77 K by a Brunauer–Emmett–Teller (BET) analysis of nitrogen adsorption measurements (ASAP2020). The Raman spectra of the samples were conducted with a Renishaw Raman system model in Via spectrometer operating with a 20 mW air-cooled argon ion laser (514.5 nm) as the excitation source. Thermogravimetric (TG) analysis was performed on an STA449F3 instrument in N_2 atmosphere at a heating rate of 10 °C min^{-1}. Absorption spectra of

samples were performed on a Shimadzu UV-2550 UV–Vis. spectrometer.

1.3 Synthesis of FeNS@PC and other samples

Typically, 0.1 g CDs and 0.2 g FeSO$_4$·7H$_2$O were dispersed in 20 mL water under ultrasonic for 0.5 h. Then, 20 mL ammonia solution was added into the mixed solution of CDs and FeSO$_4$·7H$_2$O under ultrasonic for another 0.5 h, afterwards the mixed solution dried at 70 °C to obtain solid powders. Subsequently, these powders were further treated at 800 °C for 2 h in a tube furnace under an Ar atmosphere with a heating rate of 5 °C/min. After cooling to room temperature, FeNS@PC was obtained finally. For comparison, contrast samples 1 (CS1) was prepared via the same procedure without adding CDs. Contrast samples 2 (CS2) was obtained by etching the FeNS@PC sample in H$_2$SO$_4$ (0.5 M) for 24 h. The other contrast samples (CS3, CS4 and CS5) were prepared via the same procedure, except for the different calcination temperature (400 °C, 600 °C, 1000 °C), which were shown in the following table S1.

Table S1. The details of reaction conditions for different samples

Samples / Reaction conditions	Pyrolysis Temp. (°C)	Mass of CDs (mg)	Mass of FeSO$_4$·7H$_2$O (mg)
FeNS@PC	800	100	200
Contrast Sample 1 (CS1)	800	0	200
Contrast Sample 2 (CS3)	400	100	200
Contrast Sample 3 (CS4)	600	100	200
Contrast Sample 4 (CS5)	1000	100	200

1.4 Catalytic activity tests

All experiments were conducted in 50 mL beaker containing 20 mL of reaction solution with TC and catalyst at ambient temperature. For degradation experiment by activation H_2O_2, 2 mg of FeNS@PC was added to 20 mL of TC solution (50 mg/L), then the pH of reaction solution was adjusted to 4.0 by HCl (0.1 M) and stirred for 0.5 h to achieve adsorption-desorption equilibrium. Afterwards, H_2O_2 with different concentrations was added into the reaction solution to initiate the degradation process. The reaction solution was withdrawn and centrifuged at specific time intervals to be used for TC degradation estimation. The supernatant absorbance at 359 nm, which is a characteristic absorption peak, was monitored using a UV-Vis. spectrophotometer.

For degradation experiment by activation PS and H_2O_2+PS, the initial pH of reaction solution was unregulated. 2 mg of FeNS@PC was added to 20 mL of TC solution (50 mg/L) under stirring for 0.5 h to achieve adsorption-desorption equilibrium. The degradation process was initiated by PS (0.5 mM) and the mixture of H_2O_2 (1 mM) and PS (0.5 mM). The pH of reaction solution was monitored at each time interval (0, 30, 60, 90 min). Besides, the reaction solution was withdrawn and centrifuged at specific time intervals to be further used for TC degradation estimation. The supernatant absorbance at 359 nm was monitored using a UV-Vis. spectrophotometer.

To investigate the roles of reactive radicals, the HO$^\bullet$ radicals were detected by monitoring fluorescence emission of the product resulted from the reaction of terephthalic acid (TA, 0.2 mM) with OH$^\bullet$ radicals. Moreover, MeOH (0.5 M) and TBA (0.5 M) were used as the quencher for $SO_4^{\bullet-}$ and HO$^\bullet$, respectively.
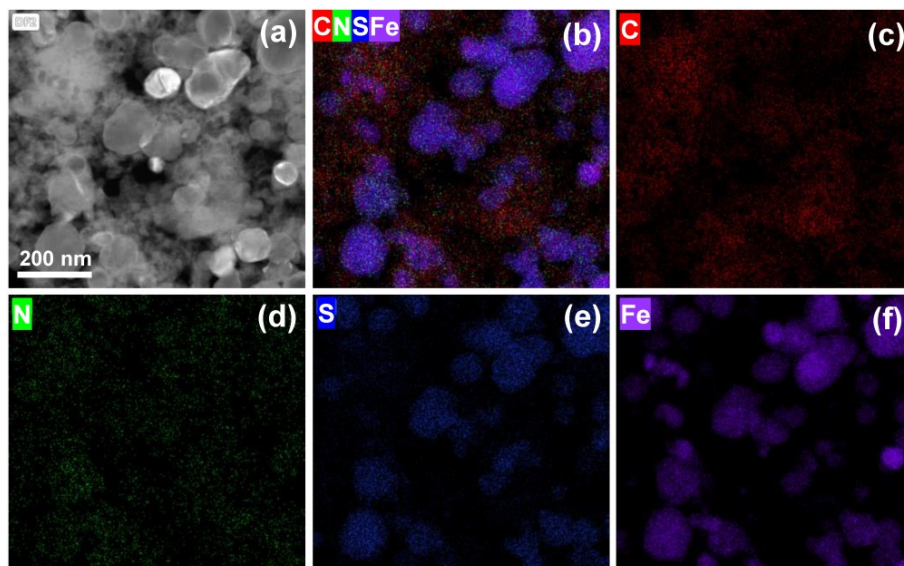
2. Supporting results

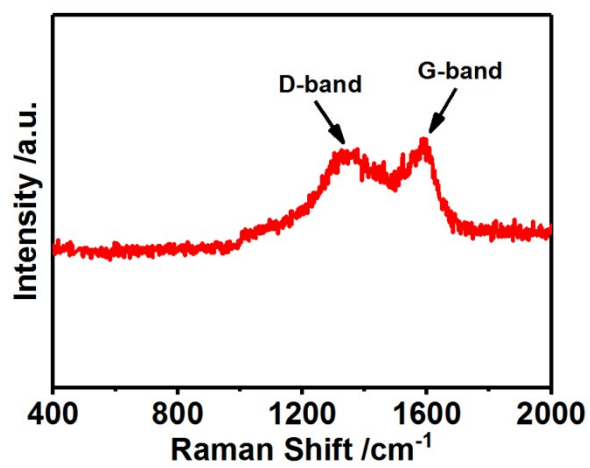Figure S1. EDS mapping images of FeNS@PC
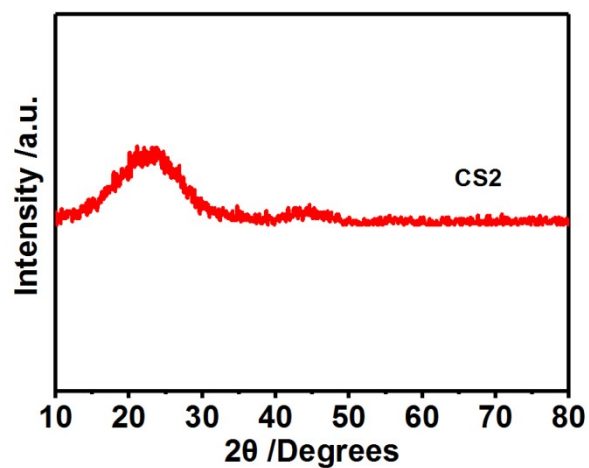

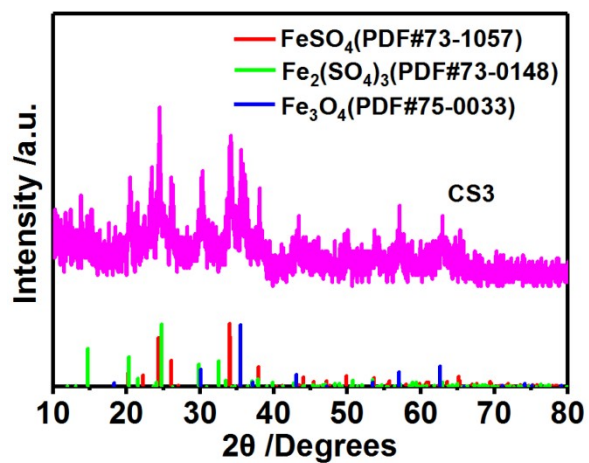Figure S2. Raman spectrum of FeNS@PC

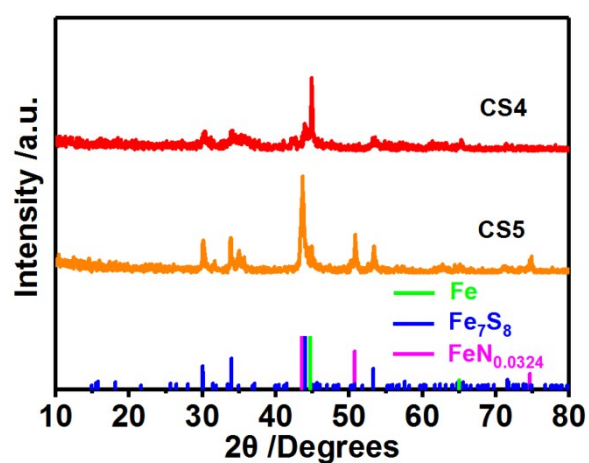Figure S3. XRD pattern of CS2

Figure S4. XRD pattern of CS3

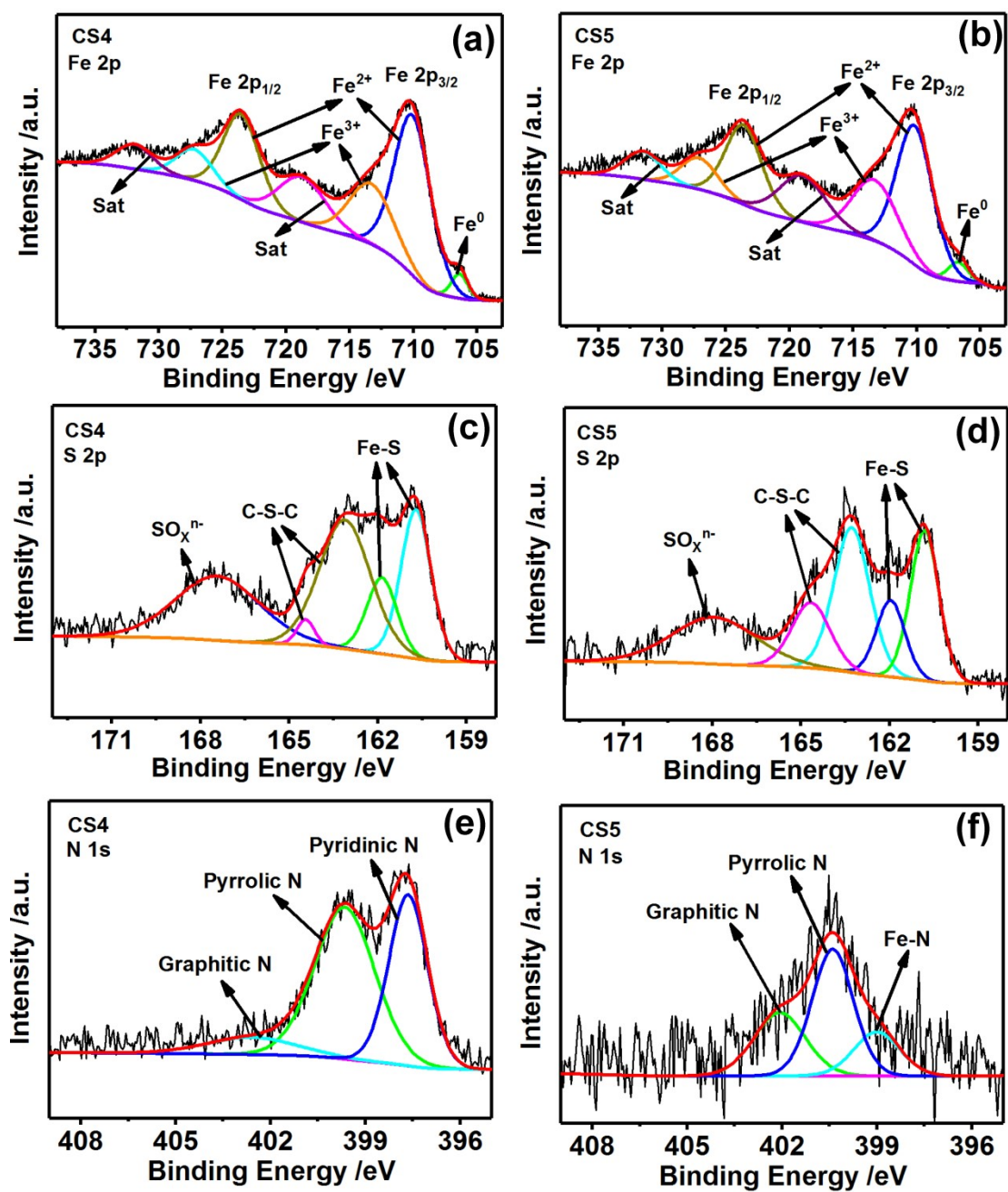Figure S5. XRD pattern of CS4 and CS5

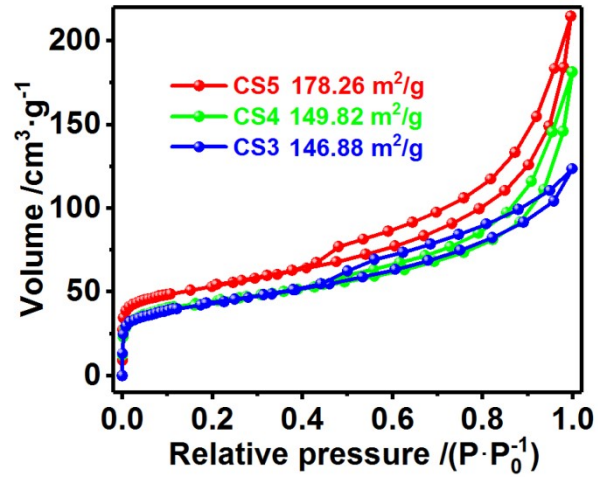Figure S6. XPS spectra of CS4 and CS5: Fe 2p (a, b); S 2p (c, d) and N 1s (e, f)

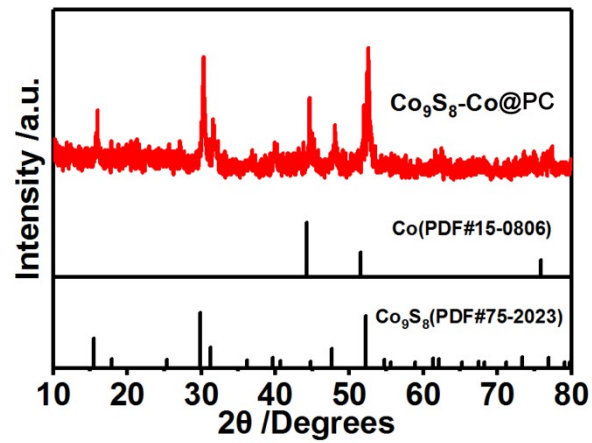Figure S7. Nitrogen adsorption-desorption isotherms of CS3, CS4 and CS5.

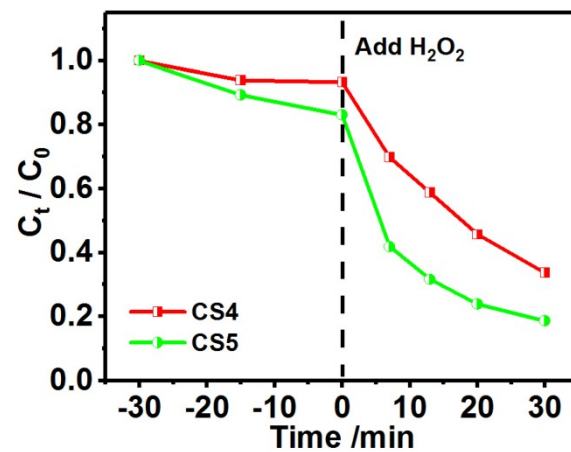

Figure S8. XRD pattern of as-prepared sample

Figure S9. TC degradation performances of CS4 and CS5 in the presence H_2O_2
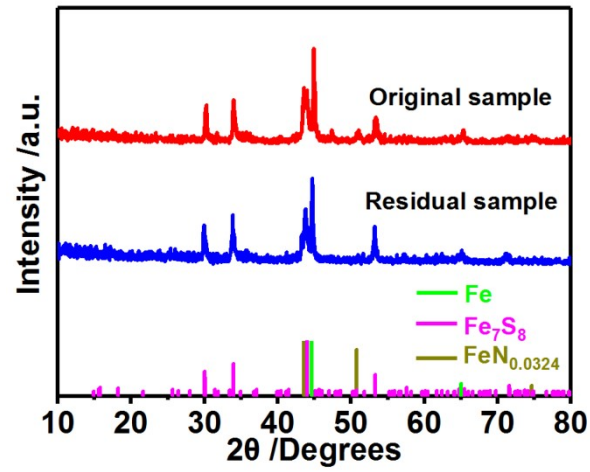
Figure S10. XRD patterns of as-prepared FeNS@PC and its residuals after accomplishing catalytic reaction