Supporting Information

Enhanced Antibiotic Removal through a Dual-Reaction-Center Fenton-like Process in 3D Graphene Based Hydrogels

Yuan Zhuang[1], Qiaozhi Liu[1,2], Yan Kong[1], Congcong Shen[3], Haotian Hao[1], Dionysios D. Dionysiou[4], Baoyou Shi[1,2]*

[1] Key Laboratory of Drinking Water Science and Technology, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing, 100085, China. Tel: 86-10- 62922155, Email: byshi@rcees.ac.cn

[2] University of Chinese Academy of Sciences, Beijing 100049, China

[3] State Key Laboratory of Urban and Regional Ecology, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China

[4] Environmental Engineering and Science Program, Department of Chemical and Environmental Engineering (ChEE), 705 Engineering Research Center, University of Cincinnati, Cincinnati, OH, USA

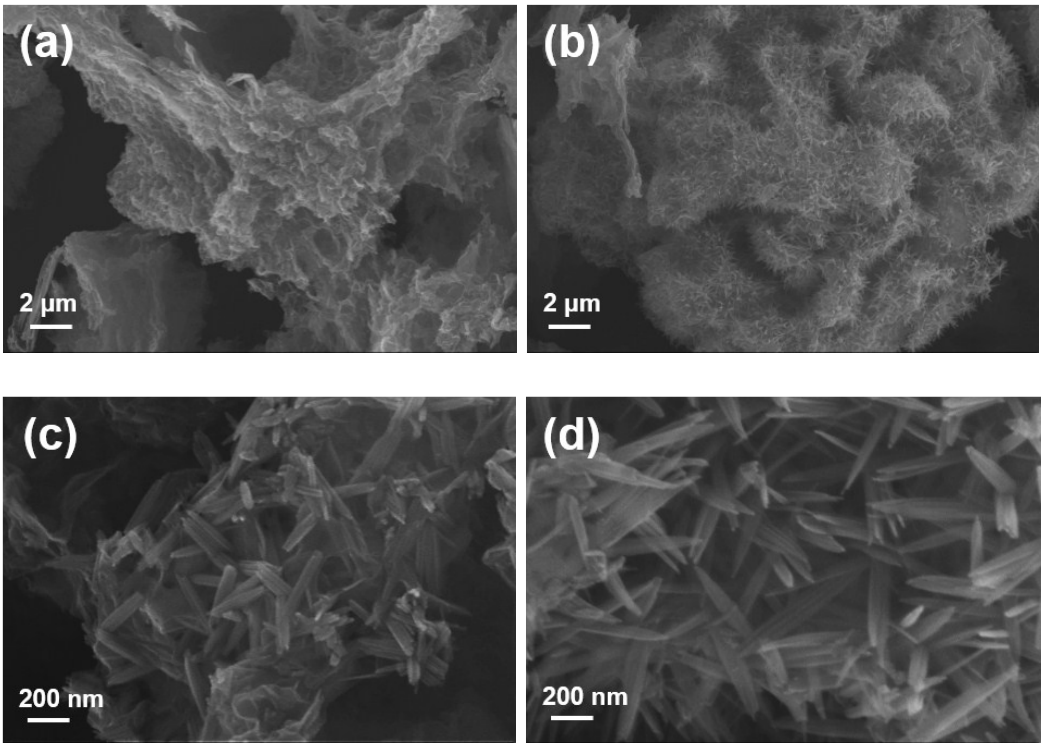


Figure S1 SEM of (a) and (c)FG-0, (b) and (d) FG-3.

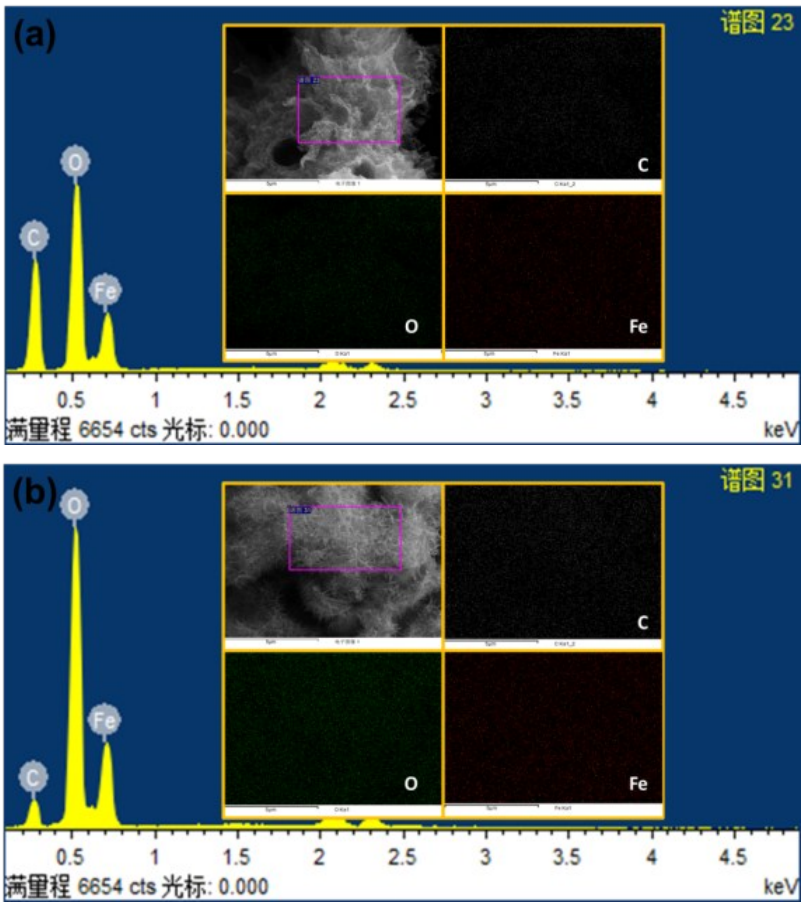


Figure S2 EDS of (a) FG-0, (b) FG-3.

Table S1 Thickness of graphene sheets and surface roughness parameters of FG-0 and FG-3.

Parameters	FG-0	FG-3
Thickness (nm)	1.3	1.6
R_a	0.18	0.21
R_q	0.28	4.30
R_z	2.63	1.93

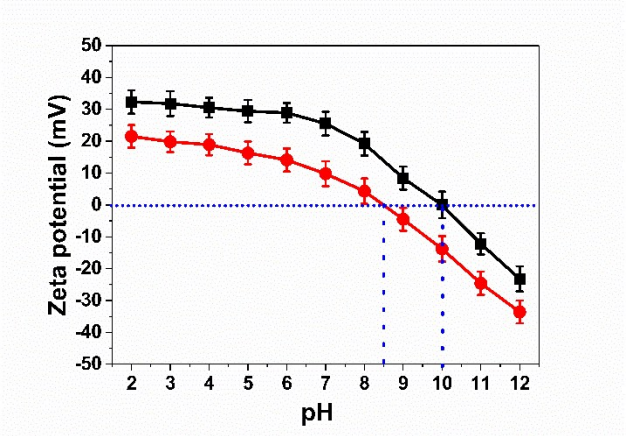
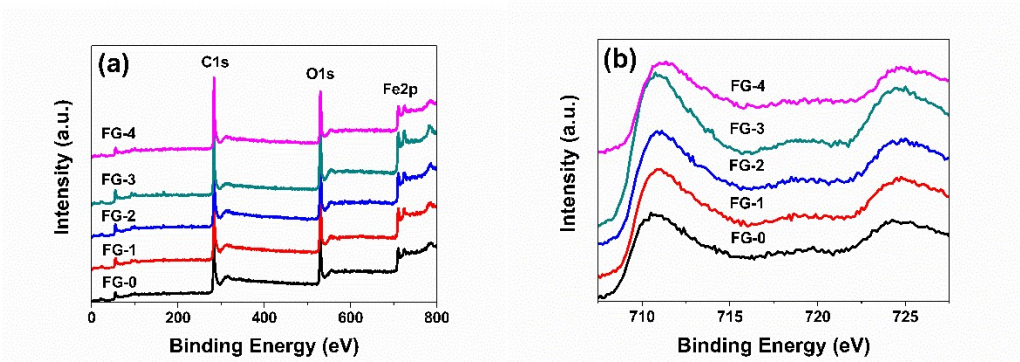


Figure S3 Zeta potential of FG-0 and FG-3



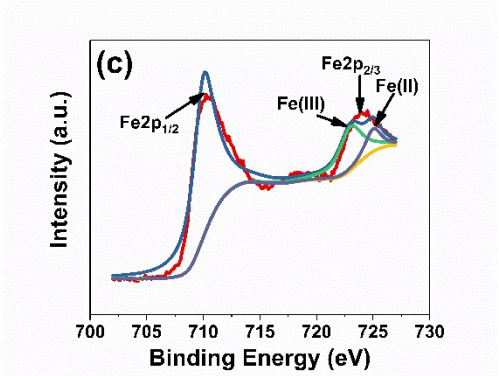


Figure S4 XPS spectra. (a) Full survey XPS spectra of FG-0 and FG-3, (b) Fe2p XPS spectra of FG-0 and FG-3, (c) Fe2p XPS spectra FG-3.

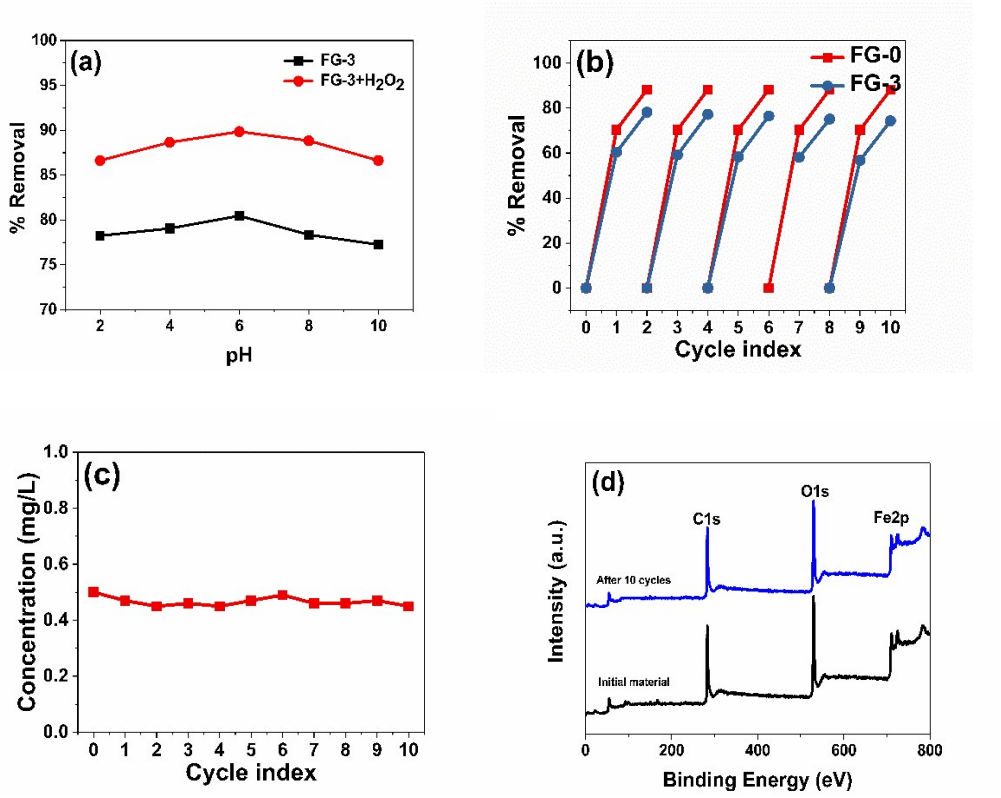


Figure S5 (a) Removal (%) of tetracycline by FG-3 at different initial pH values, (b) cycling runs in the degradation of tetracycline, (c) iron leaching in regeneration cycle, (d) XPS of FG-3 after 10 cycle runs.

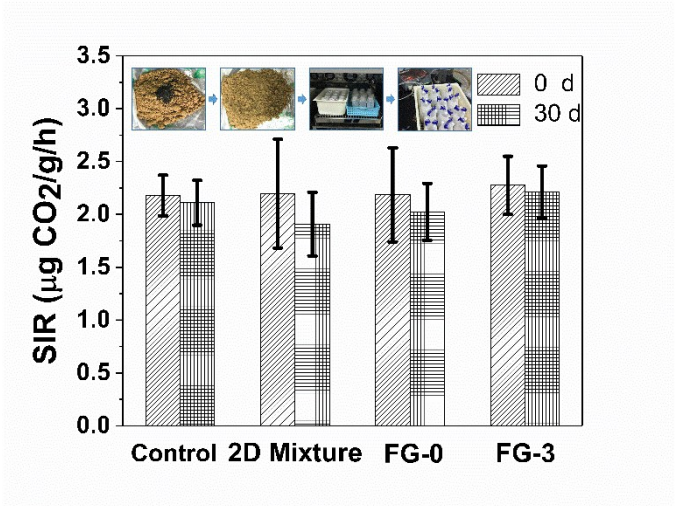


Figure S6 Treatment effects on SIR after 30 days of incubation. Error bars indicate the standard error of the mean (n = 3).

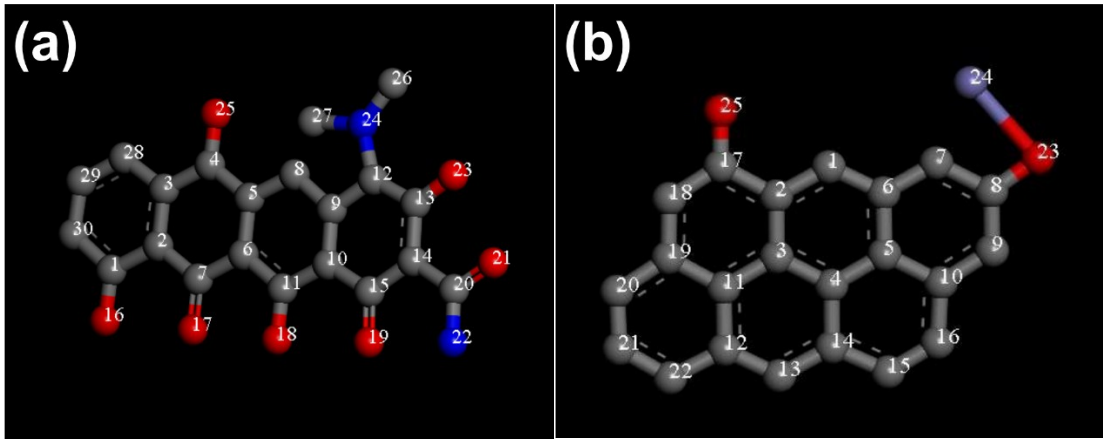


Figure S7 DFT optimized structure of (a) tetracycline, (b) rGO/FeOOH

$$\mu = (E_{HOMO+} E_{LUMO})/2 \quad \text{(S1)}$$

$$\eta = (E_{HOMO-} E_{LUMO})/2 \quad \text{(S2)}$$

$$\omega = \mu 2/2\eta \quad \text{(S3)}$$

$$\chi = -\mu \quad \text{(S4)}$$

Table S2 Parameter of chemical activity analysis.

Molecule	Tetracycline	FG composite
E_{HOMO}	-0.20404	-0.19158
E_{LUMO}	-0.20039	-0.18883
$\Delta E_{LUMO-HOMO}$	0.00365	0.00275
μ	-0.20221	-0.19021
χ	0.20221	0.19021

η	0.00183	0.00138
ω	11.17183	13.10864

Table S3 Fukui function of tetracycline.

Atom	$\mathbf{f_k^+}$	$\mathbf{f_k^-}$
C(1)	0.018	0.018
C(2)	0.02	0.02
C(3)	0.014	0.014
C(4)	0.022	0.022
C(5)	0.019	0.019
C(6)	0.019	0.02
C(7)	0.022	0.022
C(8)	0.016	0.016
C(9)	0.015	0.015
C(10)	0.019	0.019
C(11)	0.022	0.023
C(12)	0.025	0.025
C(13)	0.022	0.021
C(14)	0.021	0.021
C(15)	0.022	0.022
O(16)	0.048	0.048

O(17)	0.04	0.04
O(18)	0.042	0.042
O(19)	0.039	0.039
C(20)	0.022	0.022
O(21)	0.049	0.049
N(22)	0.066	0.066
O(23)	0.042	0.042
N(24)	-0.003	-0.003
O(25)	0.043	0.043
C(26)	0.088	0.088
C(27)	0.063	0.063
C(28)	0.056	0.056
C(29)	0.05	0.05
C(30)	0.059	0.059

Table S4 Fukui function of rGO/FeOOH.

Atom	$\mathbf{f_k^+}$	$\mathbf{f_k^-}$
C(1)	0.055	0.056
C(2)	0.016	0.017
C(3)	0.02	0.02
C(4)	0.02	0.02
C(5)	0.02	0.021

C(6)	0.013	0.014
C(7)	0.028	0.028
C(8)	0.021	0.021
C(9)	0.065	0.065
C(10)	0.012	0.012
C(11)	0.021	0.021
C(12)	0.011	0.012
C(13)	0.064	0.064
C(14)	0.011	0.012
C(15)	0.059	0.06
C(16)	0.059	0.06
C(17)	0.023	0.023
C(18)	0.064	0.064
C(19)	0.011	0.012
C(20)	0.061	0.062
C(21)	0.056	0.056
C(22)	0.061	0.061
O(23)	0.041	0.041
Fe(24)	0.138	0.131
O(25)	0.049	0.05