

## **Facile synthesis of a highly effective g-C<sub>3</sub>N<sub>4</sub>-based catalyst for advanced oxidative degradation of organic pollutants**

Zhujian Huang<sup>\*,a,b</sup>, Minxian Shen<sup>a</sup>, Junhong Liu<sup>a</sup>, Jiaer Ye<sup>c</sup>, Tewodros Asefa<sup>\*,b</sup>

*a. Guangdong Laboratory for Lingnan Modern Agriculture, College of Natural Resources and Environment, South China Agricultural University, 483 Wushan St, Guangzhou, 510642, P. R. China*

*b. Department of Chemistry and Chemical Biology & Department of Chemical and Biochemical Engineering, Rutgers, 610 Taylor Road, The State University of New Jersey, Piscataway, NJ 08854, USA*

*c. Department of Chemistry, University College London, 20 Gordon St, London WC1H 0AJ, United Kingdom*

\*Corresponding authors' E-mails: zjhuang@scau.edu.cn (Z. Huang); tasefa@chem.rutgers.edu (T. Asefa).

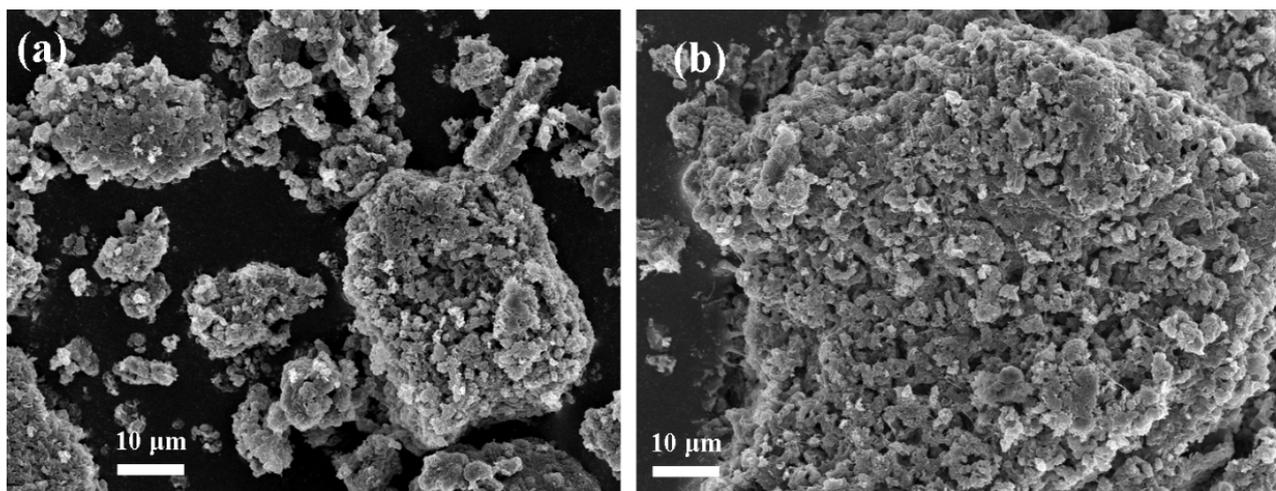


Fig. S1. SEM images of (a) Fe-C<sub>3</sub>N<sub>4</sub> and (b) Mn-C<sub>3</sub>N<sub>4</sub>.

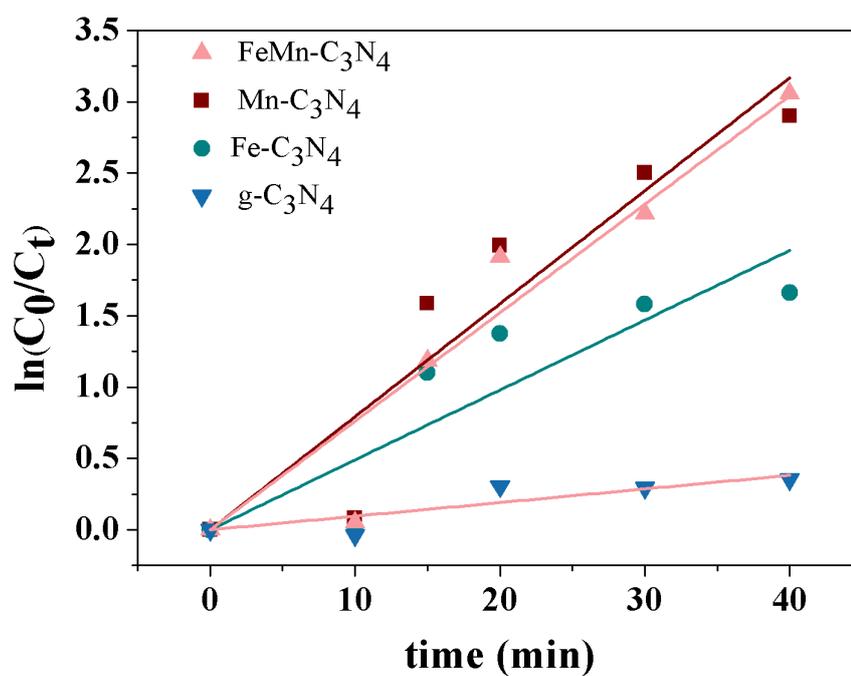
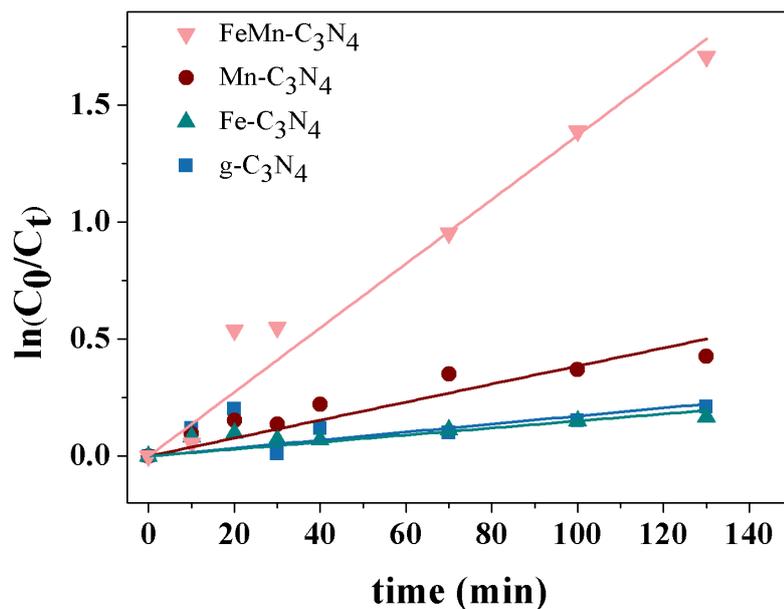
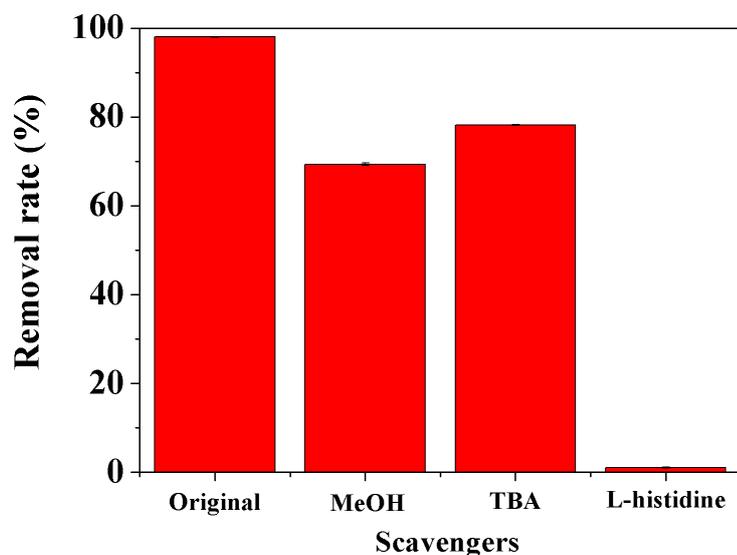


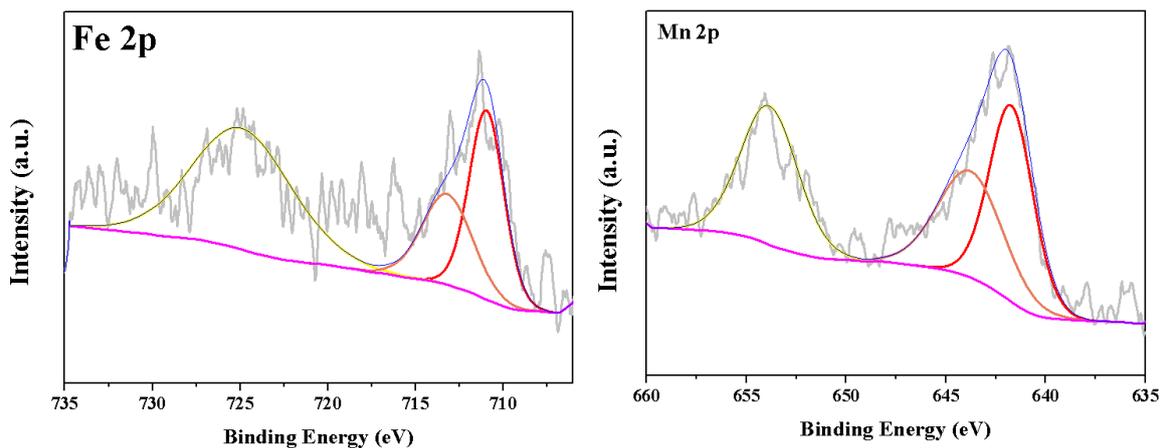
Fig. S2. The Pseudo-first order kinetic curves of degradation of RhB by persulfate activation.



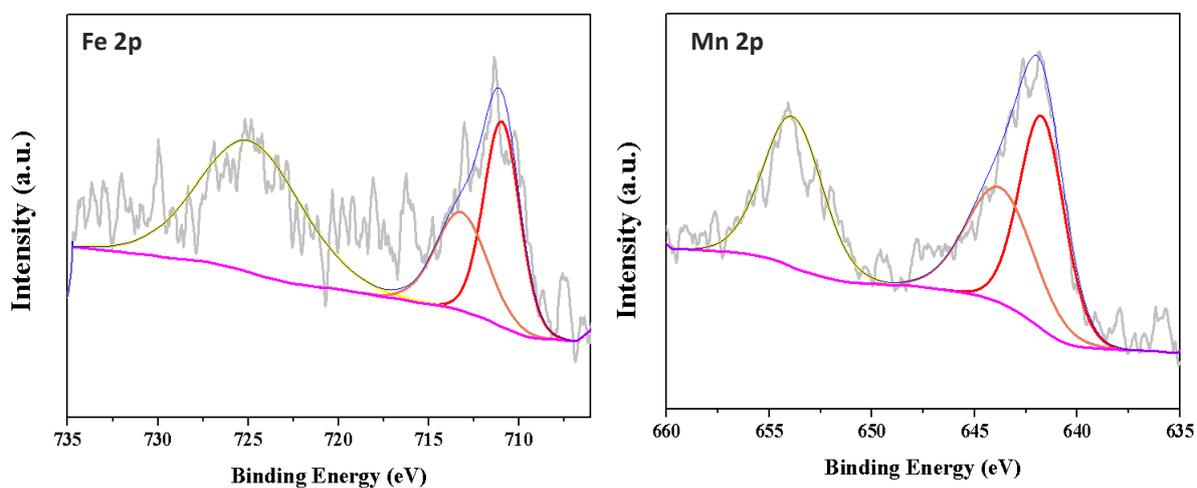
**Fig. S3.** The Pseudo-first order kinetic curves of degradation of RhB via Fenton reaction catalyzed by FeMn-C<sub>3</sub>N<sub>4</sub> and the corresponding control materials (Mn-C<sub>3</sub>N<sub>4</sub>, Fe-C<sub>3</sub>N<sub>4</sub> and g-C<sub>3</sub>N<sub>4</sub>).



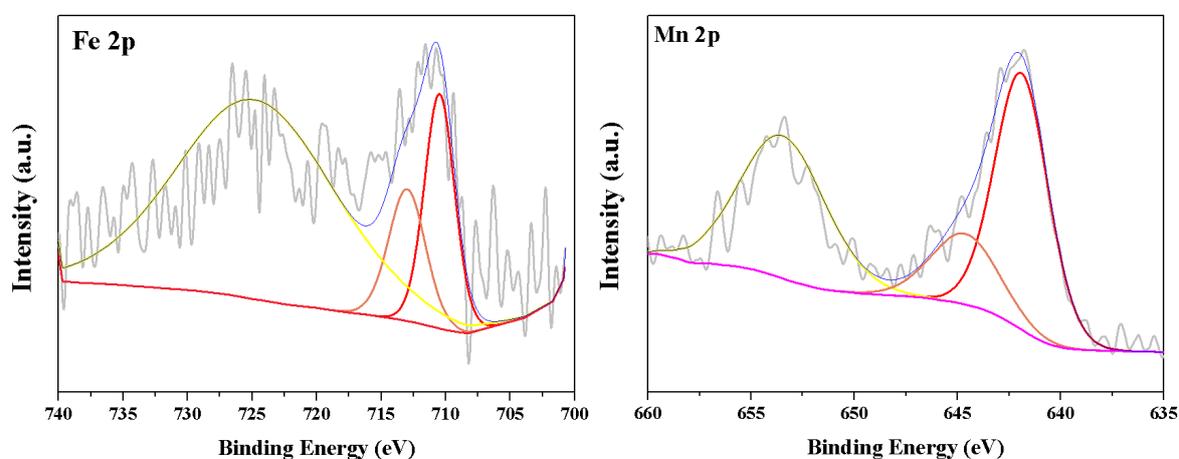
**Fig. S4.** The catalytic activity of FeMn-C<sub>3</sub>N<sub>4</sub>/PMS system for removing RhB (in %) in the presence of different scavengers (MeOH or methanol, which is used to quench SO<sub>4</sub><sup>•-</sup> and ·OH; TBA or *tert*-butyl alcohol, which is used to quench ·OH; and *L*-histidine, which is used to quench <sup>1</sup>O<sub>2</sub>). The one in absence of quenching agents (denoted Original) is also included for comparison.



**Fig. S5.** XPS spectra of peaks associated with (a) Fe 2p and (b) Mn 2p in FeMn-C<sub>3</sub>N<sub>4</sub>/PMS after it is used persulfate activation reaction.



**Fig. S6.** XPS spectra of peaks associated with (a) Fe 2p and (b) Mn 2p in FeMn-C<sub>3</sub>N<sub>4</sub>/H<sub>2</sub>O<sub>2</sub> after it is used in Fenton reaction.



**Fig. S7.** XPS spectra of peaks associated with (a) Fe 2p and (b) Mn 2p in FeMn-C<sub>3</sub>N<sub>4</sub> after it is used visible-light photocatalytic reaction.

**Table S1.** The kinetic parameters ( $K_1$ ,  $10^{-3} \text{ min}^{-1}$ ) of persulfate (PMS) activation, heterogeneous Fenton reaction and photocatalysis as catalyzed by the different  $\text{C}_3\text{N}_4$ -based materials studied.

Samples and Reactions	$\text{C}_3\text{N}_4$	$\text{Fe-C}_3\text{N}_4$	$\text{Mn-C}_3\text{N}_4$	$\text{FeMn-C}_3\text{N}_4$
Activation of PMS	9.55	48.97	79.21	76.05
Fenton-like reaction	1.72	1.50	3.85	13.70
Visible-light photocatalytic test	4.92	12.30	4.42	10.80

**Table S2.** Comparison of the catalytic performance of  $\text{FeMn-C}_3\text{N}_4$  in the Fenton-like reaction to degrade different compounds with respect to those of different heterogeneous and solid catalysts reported in the literature for the same reaction.

Heterogeneous Catalysts	Reaction Type	Removal Efficiency	Experimental conditions	References
$\gamma\text{-Fe}_2\text{O}_3/\text{Mn}_3\text{O}_4$	Persulfate activation	Removal rate of RhB = 95.6%	Persulfate concentration = 50 mg/L; catalyst dosage = 50 mg/L; initial pH = 5.1	(Ma, et al. 2019)
Porous Mn/ $\text{Fe}_3\text{O}_4$	PMS activation	Removal rate of bisphenol A = 87%	PMS concentration = 2 mmol/L; catalyst dosage = 0.2 g/L; initial pH = 3	(Du, et al. 2019)
g- $\text{C}_3\text{N}_4/\text{MnFe}_2\text{O}_4$	PMS activation	Removal rate of triclosan (TCS) = 95.9%	PMS concentration = 0.5 mmol/L; catalyst dosage = 0.5 g/L; TCS = 9 mg/L; initial pH = 11	(Wang, et al. 2019)
Mn doped g- $\text{C}_3\text{N}_4$	PMS activation	Removal rate of acetaminophen (ACT) = 100%	PMS concentration = 0.8 g/L; catalyst dosage = 0.2 g/L; ACT = 20 mg/L; initial pH = 6.5	(Fan, et al. 2019)

<b>FeMn-C<sub>3</sub>N<sub>4</sub></b>	<b>PMS activation</b>	<b>Removal rate of RhB = 98.20%</b>	<b>PMS concentration = 50 mg/L; catalyst dosage = 0.5 g/L; pH = 6.80</b>	<b>This paper</b>
Fe doped g-C <sub>3</sub> N <sub>4</sub>	Fenton-like reaction	Oxidation of RhB = 80%	H <sub>2</sub> O <sub>2</sub> concentration = 200 mmol/L; catalyst dosage = 0.5 g/L; initial pH = 6.5	(Bicalho, et al. 2017)
<b>FeMn-C<sub>3</sub>N<sub>4</sub></b>	<b>Fenton-like reaction</b>	<b>Removal rate of RhB = 81.87%</b>	<b>H<sub>2</sub>O<sub>2</sub> concentration = 21.76 mmol/L; catalyst dosage = 0.5 g/L; initial pH = 3;</b>	<b>This paper</b>
Fe/C <sub>3</sub> N <sub>4</sub>	Photocatalysis	Removal rate of RhB = 79%	catalyst dosage = 0.4 g/L; pH=3.5; light source: 350 W Metal halide-Xenon lamp ; with addition of 3 mmol/L persulfate; reaction time: 40 min	(Heidarpour, et al. 2020)
g-C <sub>3</sub> N <sub>4</sub> -Mn-H	Photocatalysis	Removal rate of RhB = 51.4%	catalyst dosage = 0.5 g/L; light source: 300 W Xe arc lamp; reaction time: 60 min	(Wang, et al. 2017)
<b>FeMn-C<sub>3</sub>N<sub>4</sub></b>	<b>Photocatalysis</b>	<b>Removal rate of RhB = 94%</b>	<b>pH = 6.80; catalyst dosage = 0.5 g/L; light source: 30 W LED light; reaction time: 210 min</b>	<b>This paper</b>

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