Supplementary Data

Facilitating Fe(III)/Fe(II) redox cycle by organic pollutant for enhancement of heterogeneous Fenton-like catalytic performance of iron-containing catalyst

Cong Gao[a*], Houfen Li[b], Jize Liu[a], Wenchao Yang[a], Hong Chen[b], Jianbo Han[b]

[a] Key Laboratory of Coastal Ecology and Environment of State Oceanic Administration, National Marine Environmental Monitoring Center, Dalian 116023, China

[b] College of Environment and Ecology, Taiyuan University of Technology, Taiyuan 030024, China

* Corresponding authors.

E-mail: cgao@nmemc.org.cn, gaocong.200746033@163.com

pH_{PZC} measurement

MIL-100-Fe powder (0.015 g) was added into 10 mL KNO_3 solution (0.01 M), followed by 24 h of equilibration to ensure complete dispersion of the catalyst particles. After equilibration, the pH value of the resulting suspension was adjusted to a series of target values using HCl (0.4 M) and NaOH (0.4 M). Subsequently, the zeta potential of MIL-100-Fe suspension at each adjusted pH was performed on a Zetasizer nano ZSP (Malvern nano-ZS90, UK). The pH_{PZC} of MIL-100-Fe is defined as the pH value at which the zeta potential of MIL-100-Fe was zero, which can be directly obtained from the intersection point of the zeta potential-pH curve with the vertical axis at 0 mV zeta potential.

Linear sweep voltammetry (LSV) measurement

LSV measurements were performed using a three-electrode glass cell. The working electrode was a glassy carbon electrode (GCE), the reference electrode was Ag/AgCl$_3$/3 M KCl. Pt electrode was used as auxiliary electrode.

Experiment conditions for testing the redox potential of MIL-100-Fe(III)/Fe(II): the sweep range was from 0.9 V to -0.4 V at a scan rate of 0.01 V/s. The experiment was carried out at room temperature in N_2-saturated 0.1 M Na_2SO_4 electrolyte at pH 4.0.

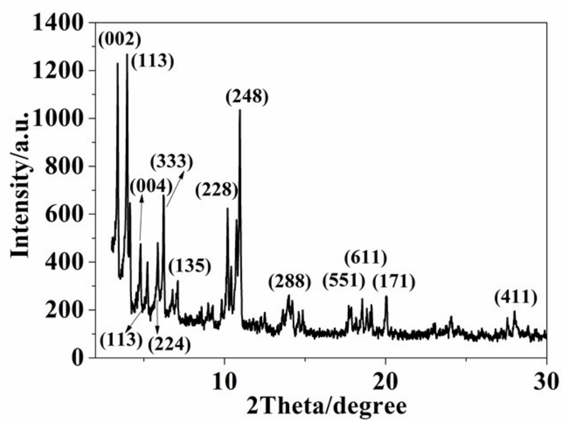

Figure S1. XRD pattern of MIL-100-Fe. (A)

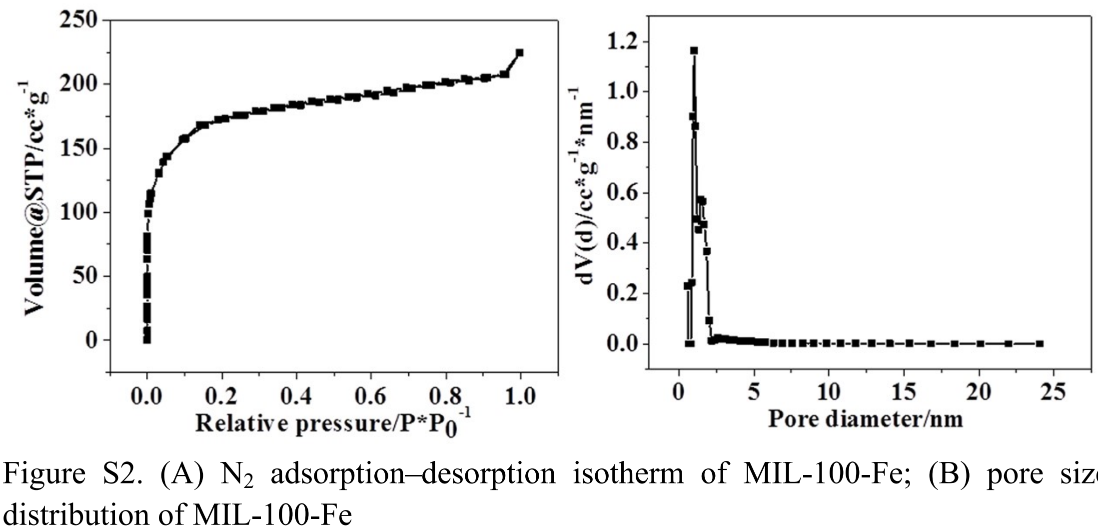

Figure S2. (A) N_2 adsorption–desorption isotherm of MIL-100-Fe; (B) pore size distribution of MIL-100-Fe

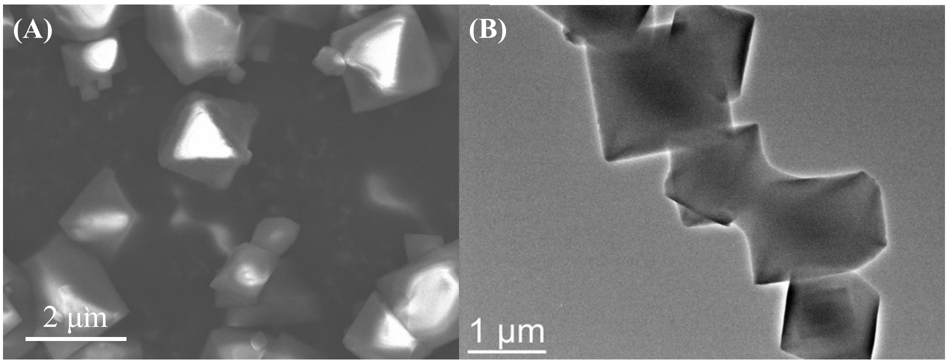

Figure S3. (A) SEM and (B) TEM pictures of MIL-100-Fe.

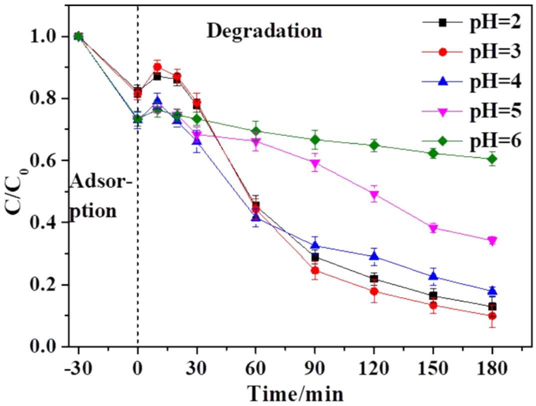

Figure S4. Influence of initial solution pH on the catalytic performance of MB in MIL-100-Fe catalyzed Fenton-like reaction. Reaction condition: [catalyst] = 0.1 g/L, [H_2O_2] = 16 mM, [organic pollutants] = 100 mg/L, at room temperature.

Table R1. Comparison of the performance of different Fenton-like catalysts for dye removal

Catalyst	Reaction condition	TOC removal rate	TOC removal rate per unit mass of catalyst	ref
Prussian-blue (PB)-modified - Fe_2O_3	pH=5.5, T=298 K, [catalyst]=0.3 g/L, [H_2O_2]=0.4 mol/L, [MB]=20 μg/mL	45%	150%/g L^{-1}	[1]
H-Fe-S	[RhB]=2.0*10^{-4} M; [H-Fe-S]=2 g/L; [H_2O_2] = 12 mM, visible light	38%	19%/g L^{-1}	[2]
Fe $_0$/Fe_3O_4	[MB]=100 mg/L, [H_2O_2] = 0.3 M, pH 6.0±0.2,[catalyst]=3 g/L	75%	25%/g L^{-1}	[3]
Graphite tailing	[RhB]=100 mg/L, [catalyst]=50 g/L, [H_2O_2]=10 g/L, pH = 3.92	49.02%	0.98%/g L^{-1}	[4]

Iron impregnated biochars	[AR1]=50 mg L^{-1}, pH 3, $[H_2O_2]$=16 mM, [catalyst]=4 g L^{-1}	86.7%	21.68%/g L^{-1}	[5]
	[MB]=100 mg/L, $[H_2O_2]$=0.133 mL/L, [catalyst]=1.3 g L^{-1}, 125 W Hg lamp	21.1%-89.1%	16.2-68.5	[6]
MIL-100-Fe	[MB]=100 mg/L, [catalyst]=0.1 g/L, $[H_2O_2]$=16 mM, pH= 4	49%	490%/g L^{-1}	this study

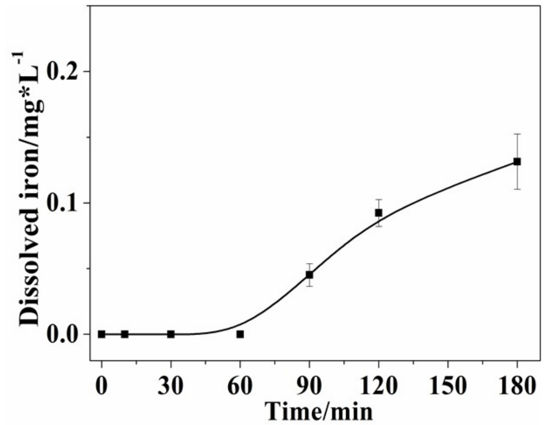

Figure S5. Variation of leached iron ions from MIL-100-Fe during Fenton-like system. Reaction conditions: [catalyst] = 0.1 g/L, $[H_2O_2]$ = 16 mM, [MB] = 100 mg/L, pH 4.0 at room temperature.

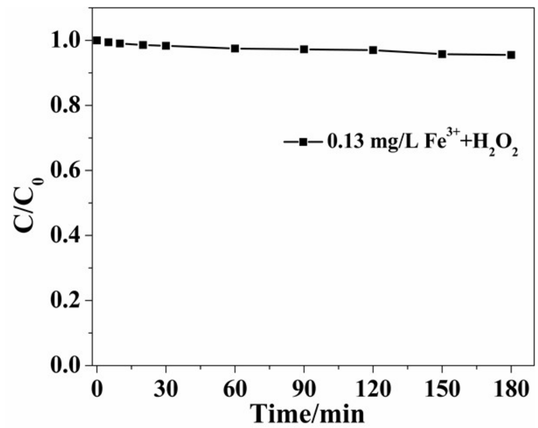

Figure S6. Catalytic degradation of MB in homogeneous Fenton-like reaction. Reaction conditions: $[H_2O_2]$ = 16 mM, [MB] = 100 mg/L, pH 4.0 at room temperature.

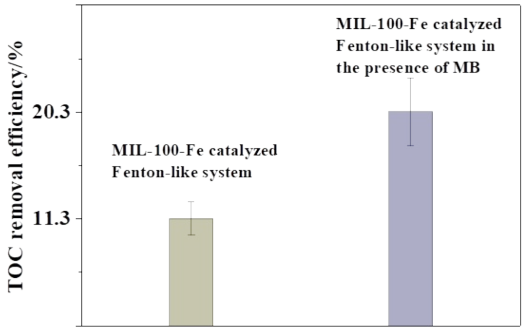

Figure S7. The TOC removal efficiency of the dyeing wastewater by MIL-100-Fe catalyzed Fenton-like reaction. Reaction condition: [catalyst] = 0.1 g/L, [H_2O_2] = 16 mM, [MB] = 100 mg/L, pH 4.0 at room temperature.

Reference

[1] H. Wang, Y. Huang. Prussian-blue-modified iron oxide magnetic nanoparticles as effective peroxidase-like catalysts to degrade methylene blue with H2O2. J. Hazard. Mater. 2011, 191, 163-169.

[2] Y. Gao, H. Gan, G. Zhang, Y. Guo. Visible light assisted Fenton-like degradation of rhodamine B and 4-nitrophenol solutions with a stable poly-hydroxyl-iron/sepiolite Catalyst. Chem. Eng. J. 2013, 217, 221-230.

[3] R.C.C. Costa, F.C.C. Moura, J.D. Ardisson, J.D. Fabris, R.M. Lago. Highly active heterogeneous Fenton-like systems based on Fe0/Fe3O4 composites prepared by controlled reduction of iron oxides. Appl. Catal. B: Environ. 2008, 83, 131–139.

[4] C. Bai , W. Gong, D. Feng, M. Xian, Q. Zhou, S. Chen, Z. Ge, Y. Zhou. Natural graphite tailings as heterogeneous Fenton catalyst for the decolorizationof rhodamine B. Chem. Eng. J. 2012, 197, 306–313.

[5] K.K. Rubeena, P.H.P Reddy, A.R. Laiju, P.V. Nidheesh. Iron impregnated biochars as heterogeneous Fenton catalyst for the degradation of acid red 1 dye. J. Environ. Manage. 2018, 226, 320-328.

[6] F.E. Bimbi Junior, C.G. Neves, M.D.L. do Nascimento, E.A. Falcão, W.R.P. Barros. Photo-Fenton-Based Degradation of Methylene Blue Dye Using Hydroxyapatite Nanoparticles Doped with Fe3O4/γ-Fe2O3. J. Braz. Chem. Soc. 2022, 33, 1441-1455.