

Performance of heterogeneous Fenton-like system for degradation of nitrobenzene-containing wastewater

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Abstract A heterogeneous Fenton-like catalytic system, consisted of 3A-Fe zeolite catalyst that was prepared in the laboratory and characterized by SEM, EDS and XRD, and H_2O_2 was used for degradation of nitrobenzene-containing wastewater. The effects of pH, H_2O_2 and nitrobenzene concentration, and catalyst dose on degradation of nitrobenzene were studied. An attempt to reveal the degradation mechanism was also done. The results showed that nitrobenzene had been significantly degraded in this Fenton-like system provided that pH value of wastewater was in the range of 2 to 10. No dissolved Fe(/) was detected for the Fenton-like system, indicating that nitrobenzene was directly degraded on surface of 3A-Fe zeolite catalyst rather than by the aqueous Fenton system. The 3A-Fe zeolite catalyst exhibited good stability in repeat use and the degradation rate of nitrobenzene could reach 94.1%, and the removal rate of COD_{Cr} and TOC, under the optimal conditions, were 78.6% and 60.5% respectively.

2013-08-05 2013-11-01 1980—

Project of Taizhou (111KY003), the Scientific Environmental Protection Bureau 2013B018 stentation Foundation of Zhejiang Province 6] 2

		1.2.2 3A-Fe	
		S-4800	
		SEM D8 adv	ance
[1]		X (XRD)	
		1.3	
		_	
	Fenton ^[2]	COD TOC TOC	
[3-5]	Fenton pH	ոկ ոկ	
2 4	[6-9]	14	
2 7		1.4 Fenton	
	Fenton	$100 \text{ mg} \text{ J}^{-1}$	uл
[10-13]	i enton		п ₂ О ₂
	Fenton	$Q_{th}($) 0	50 g
nH	I enton	H_2O_2	
Fento	n [14-16]	CO_2 H_2O H_2O_2	2004
3 A_Fe	11	$I \qquad IQ_{th}$	30%
Fenton		H_2O_2 0.61 ml· L·	
1 Chton		$2C_6H_5NO_2 + 29[O] = 12CO_2 + 2NO_2 + 5H_2O$	(1)
		2	
1		_	
		2.1	
1.1		1 3A 3A	SEM
		1 3A	
		Fe	
31	A		
		2(b) 3A-Fe	
		Fe	
UV-7504 A	-	2 3A 3A-Fe	
DELIA-520 pr	TOC	3A-Fe	
S 4800	IUC D ⁰ advance	1	
S-4000 V	Do auvance	3A-Fe 3A Fe Fe	
л 12		11.03%	
1.2 1.2.1 2.4 Eq.		3A	
1.2.1 SA-Fe	$25 \sim 1^{-1} E_{2} SO$	3 XRD 3A 3A-1	Fe
Na_2CO_3	$2.5 \text{ g} \cdot \text{L}$ FeSO ₄	6 21.7° 24.0° 27.1° 2	9.9°
(1.2)	(1.1.1)	32.5° 34.1° 3A 3A-Fe	
(1.2)	1 1.2 h		
Na_2CO_3 FeSO ₄	SA 400	1 3A 3A-Fe	10/
	ЗA	Table 1 Element mass percentage of 3A and 3A-Fe	/%
Na_2CO_3 FeSO ₄	2	Zeolite Fe C O Na Al Si	K
2 0 N	[17]	3A-Fe 11.03 16.52 46.34 4.04 8.60 9.68	3.79
з 3A-Fe			

Key words: Fenton-like system; zeolite; wastewater; hydrogen peroxide; degradation





2.2.2 H₂O₂ 3A-Fe





4 pH



 H_2O_2 Fenton 0.5 g pH 10 H_2O_2 $100 \text{ mg} \cdot \text{L}^{-1}$ 500 ml 5 5 H_2O_2 Q_{th} 1/2 Q_{th} 1/5 Q_{th} $1/10 Q_{th}$ $1/7Q_{th}$ 120min 97.9% 97.6% 90.9% 79.7% 66.7% 0.037 0.034 0.023 0.013 0.010 min^{-1} $1/10 \textbf{Q}_{th}$ H_2O_2 120 min 60% Fenton $H_2O_2 \\$ H_2O_2 1/2 H_2O_2 1/20.3 H_2O_2 Fenton

· OH

Fig.5 Effects of H_2O_2 concentration on degradation of nitrobenzene

 $1/2 \mathbf{Q}_{th}$ $H_2O_2 \\$ [23] H_2O_2 $H_2O_2 \cdot OH$ HO_{2} · [(2)] HO_{2} · \cdot OH[(3)] H_2O_2 $H_2O_2 \ \cdot \ OH$ H_2O_2 $1/2Q_{th}$ $H_2O_2 + \cdot OH \longrightarrow HO_2 \cdot + H_2O$ (2) HO_2 + · OH \longrightarrow H_2O + O_2 (3) 2.2.3 3A-Fe H_2O_2 Fenton H_2O_2 $1/5Q_{th}$ pH 10 3A-Fe $100 \text{ mg} \cdot \text{L}^{-1}$ 500 ml 6





66.3% 0.05 0.1 0.5 1 g 86.1% 86.7% 90.9% 95.8% 0.018 0.027 mol· $(L \cdot s)^{-1}$

 H_2O_2

 H_2O_2

 H_2O_2 3A-Fe

 $mg \cdot \ L^{-1}$

2.2.6

[24] [25] Arends H_2O_2

Fenton

3A-Fe

HO- HO-

2.2.4 pН $0.5 g H_2O_2$ 0.12 10 $mL \cdot \ L^{-1}$ 100 600 $mg \cdot L^{-1}$ 7 $100 \quad 600 \text{ mg} \cdot \text{ L}^{-1}$ Fenton 100 $mg \cdot \ L^{-1}$ 91% 68.3%

 H_2O_2 · OH

[14]



Fig.7 Effects of initial concentration on degradation of nitrobenzene

2.2.5				pН	10	H_2O_2
$1/5Q_{th}$			0.5 g			500 ml
	100 mg·	L^{-1}				

120 min



ml 0.5 g $1/5Q_{th}$ H_2O_2





硝基苯 → 降解产物 ·OH + {

3

1 3A-Fe				Fenton	L		
pН	2	10					
Fenton		рН					
2	3A	-Fe					Т

References

- Wei Chaohai(), Chen Chuanhao(), Wang Gang(), et al. Characteristics of nitrobenzene containing wastewater catalytic oxidation degradation by Fenton reagent [J]. Environmental Sciences(), 2001, 22(5): 60-64
- [2] Barbara Bianco, Ida De Michelis, Francesco Vegliò. Fenton treatment of complex industrial wastewater: optimization of process conditions by surface response method[J]. Journal of Hazardous Materials, 2011, 186: 1733-1738
- [3] Laat J D, Le G T, Legube B, et al. A comparative study of the effects of chloride, sulfate and nitrate ions on the rates of decomposition of H₂O₂ and organic compounds by Fe()/H₂O₂ and Fe()/ H₂O₂[J]. Cherrosphere, 2004, 55: 715-723
- [4] Li Chunjuan(). Investigation of removal of contaminants in water by Fenton and Fenton-like oxidation[D]. Harbin: Harbin Institute of Technology, 2009
- [5] Arturo A B, Dionysios D D, Makram T S. Effect of oxidant-tosubstrate ratios on the degradation of MTBE with Fenton reagent[J].
 Water Research, 2008, 42: 3225-3239
- [6] Deng Jingheng, Jiang Jingyuan, Zhang Yuanyuan, et al. FeVO₄ as a highly active heterogeneous Fenton-like catalyst towards the degradation of Orange [J]. Applied Catalysis B: Environmental, 2008, 84:468-473
- [7] He Chun(), Xu Kefeng(),Xi Hongxia(), et al. Catalytic oxidation of wastewater containing phenol by homogeneous and heterogeneous fenton-type catalysts [J]. Journal of South China University of Technology Natural Science Edition(

, 2003, **31**(5): 51-55

- [8] Xu Huanyan, Murari Prasad, Yu Liu Schorl. A novel catalyst in mineral-catalyzed Fenton-like system for dyeing wastewater discoloration[J]. Journal of Hazardous Materials, 2009, 165: 1186-1192
- [9] Jae-Min Leea, Ji-Hun Kima, Yoon-Young Chang, et al. Steel dust catalysis for Fenton-like oxidation of polychlorinated dibenzo-pdioxins [J]. Journal of Hazardous Materials, 2009,163: 222-230
- [10] Niranjan Panda, Hrushikesh Sahoo, Sasmita Mohapatra. Decolourization of methyl orange using Fenton-like mesoporous Fe₂O₃-SiO₂ composite [J]. Journal of Hazardous Materials, 2011, 185:359-365
- [11] Gao Zhen(), Lei Guoyuan(), Jiang Chengchun().
 Degradation of reactive red MX-5B by heterogeneous Fenton reaction over Si-FeOOH[J]. Acta Scientiae Circumstantiae().
 2011, 31(4):767
- He Liping(), Yang Yingchun(), Xu Chenghua(),
 et al. Degradation of Rhodamine-B wastewater by heterogeneous
 Fenton-like reaction using active carbon-Fe [J]. Chinese Journal of
 Environmental Engineering(), 2009, 3(8): 1433-1437
- [13] Fan Xiangqun, Hao Hongyuan, Shen Xingxing, et al. Removal and degradation pathway study of sulfasalazine with Fenton-like reaction[J]. Journal of Hazardous Materials, 2011, 190: 493-500
- Kasiri M B,Aleboyeh H,Aleboyeh A. Degradation of acid blue 74 using Fe-ZSM5 zeolite as a heterogeneous photo-Fenton catalyst[J].
 Applied Catalysis B-Environmental, 2008, 84(1/2):9-15

- [16] Luo W, Zhu L H, Wang N, et al. Efficient removal of organic pollutants with magnetic nanoscaled BiFeO₃ as a reusable heterogeneous Fenton-like catalyst[J].Environmental Science & Technology, 2010, 44(5): 1786-1791
- [17] Chen Aiyin(). Study on preparation and catalytic oxidation of molecular sieve catalysts loaded with iorn species [D]. Tianjin: Nankai University, 2008: 70-71
- [18] Nalinrut Masomboona, Chavalit Ratanatamskulb, Ming-Chun Lu. Kinetics of 2,6-dimethylaniline oxidation by various Fenton processes[J]. Journal of Hazardous Materials,2011, 192: 347-353
- [19] Shaobin Wang.A comparative study of Fenton and Fenton-like reaction kinetics in decolourisation of wastewater[J]. Dyes and Pigments 2008 76 714-720
- [20] Cai Shaoqing (), Dai Qizhou(), Wang Jiayu(), et al. Heterogeneous catalytic ozone oxidation for pharmaceutical wastewater treatment [J]. Acta Scientiae Circumstantiae(), 2011, 31(7): 1440-1449
- [21] Liu Miao(), Leng Su(), Chen Songyue(), et al. Degradation of nitrobenzene wastewater with modified Ti/SnO₂-Sb electrode [J]. Chemical Journal of Chinese Universities(), 2013, 34(8): 1899-1906
- [22] Zhang Jinbao(), Xi Beidou(), Jiang Yonghai(),
 et al. Degradation effect and mechanism of 2 4-DNT by reduction-ZPF catalytic oxidation [J]. Environmental Science(), 2011, 32(10): 2937-2942
- [23] Lin Zhirong(), Zhao Ling(), Dong Yuanhua(),
 et al. Degradation of PCB28 by goethite-catalyzed hydrogen peroxide
 [J]. Acta Scientiae Circumstantiae(), 2011, 31(11):
 2403-2408
- [24] Arends IWCE, Sheldon R A. Activities and stabilities of heterogeneous catalysts in selective liquid phase oxidations: recent developments [J]. Applied Catalysis A: General,2001, 212: 175-187
- [25] Shen Jimin(), Chen Zhonglin(), Li Xueyan(), et al. Effect and mechanism of degradation of nitrobenzene in aqueous solution by O₃/H₂O₂[J]. Environmental Science(), 2006, 27(9): 1791-1797
- [26] Liou M J, Lu M C. Catalytic degradation of explosives with goethite and hydrogen peroxide[J]. Journal of Hazardous Materials, 2008, 151(2/3): 540-546
- [27] Wu Deli(), Duan Dong(), Ma Ruming(). Fenton-like oxidation of refractory organic contaminants in wastewater using pyrite cinder at neutral pH [J]. CIESC Journal(), 2010, 61(4): 1001-1008
- [28] Chou S Haung C Huang Y H Heterogeneous and homogeneous catalytic oxidation by supported γ-FeOOH in a fluidized-bed reactor kinetic approach[J]. Environ. Sci. Technol., 2001,35(6) 1247-1251
- [29] Zhang Di(), Wang Yixuan(), Niu Hongyun(), et al. Degradation of norfloxacin by nano-Fe₃O₄/H₂O₂ [J]. Environmental Science(), 2011, 32(10): 2944-2948