

Treatment of Landfill Leachate with Fenton's Reaction

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In this study, Fenton's reaction was used in landfill leachate treatment, aiming to find a convenient and flexible way for the treatment of such kind of high-concentration organic wastewater. COD and absorbance of UV_{254} were selected as the main water quality indicators for landfill leachate. pH value (6) and Fe²⁺/H₂O₂ (10) ratio were tested to obtained the optimum condition, based on which, a series of comprehensive experiments were carried out for proof-checking. It was found that the Fenton's process had effect in real landfill leachate treatment. By controlling reaction condition, the removal of COD and UV₂₅₄ reduction can be above 40 %, which indicated Fenton's process can be used as an effective pretreatment or post-treatment method for high-concentration organic wastewater.

Key Words: Fenton's reaction, Landfill leachate, COD removal, UV₂₅₄ reduction.

INTRODUCTION

Municipal solid waste landfills (MSWLFs) have long been the important locations all over the world to receive household waste, industrial solid waste, construction and demolition debris, *etc*. Though advanced technical control and strict regulating requirements are available, especially in Europe and in USA (Federal MSWLF standards), considering the existence of old closed landfills (which reminds us time and time of the movement Not-in-My-Backyard), together with numerous landfills in operation without sufficient risk control, many concerns generated, among which, landfill leachate is the most serious one¹⁻³.

Composition and volume of landfill leachate are variable, depending on such conditions as rainfall, groundwater and components of solid waste. In some cases, landfill leachate was transferred into waste water treatment plants (WWTP) in specific locations and high-concentration of heavy metallic ions and biological toxic organics may lead to negative effects on the normal operation of WWTP. Fortunately, there were also some evidences to show that some landfills were combined with sufficient leachate collecting and treatment facilities, especially in developing regions.

In this study, one typical kind of advanced oxidation processes (AOPs) named Fenton's reaction was used in landfill leachate treatment, aiming to find a convenient and flexible way for the treatment of such kind of high-concentration organic wastewater^{4,5}.

EXPERIMENTAL

Sample of landfill leachate: Landfill leachate was collected from Heishizi landfill in Chongqing on April, 2010 and there was no raining for 5 days before the sampling date. Totally 20 kg landfill leachate was collected from 2 different landfill leachate storage ponds.

 H_2SO_4 , FeSO₄·7 H_2O and all related chemicals are all analytical reagents. pH meter(pH 510), electronic balance (AL104), magnetic Stirrer (101S), double-beam UV-VIS spectrophotometer (TU-1901), COD Digestion equipment (MH-2800D) and COD instrument (LPG422.99.00012) were used in water quality test before and after treatment.

Fenton's experiments: All Fenton's experiments were beaker experiment. By experiment design, three series of system were tested, based on which, optimum pH and Fe^{2+}/H_2O_2 ratio were determined and the COD removal and absorbance of UV₂₅₄ reduction effect were obtained under optimum condition. All experiments were designed with 2 parallel experiments^{6,7}.

Water quality indicators: Considering the complex components of landfill leachate, COD and absorbance of UV_{254} were selected as the main indicators for leachate water quality. COD is a typical indicator for wastewater, which is commonly used to indirectly measure the amount of organic compounds in water. There were many studies to ascertain UV absorbance at 254 nm was a direct indicator for the concentration of total

organic carbon or dissolved organic carbon, which was comparatively easy to detect and record^{6,7}.

RESULTS AND DISCUSSION

Effect of pH condition on COD and UV₂₅₄ reduction: Based on the understanding about Fenton's reaction, six different pH values were tested *i.e.*, 2.04, 2.99, 3.99, 5.00, 6.12 and 7.03. The reaction system was as follows: 100 mL landfill leachate, H_2SO_4 was used in pH adjustment, 10 mL FeSO₄·7H₂O solution (Fe²⁺ concentration is 0.018 M) and 10 mL H₂O₂ (H₂O₂ concentration is to 0.98 M) were added. Stirring rate was 200 rpm and reaction time was 0.5 h. For every pH value, 2 parallel experiments were designed. For COD detection of every system, 3 samples were taken for test to avoid subjective error. Before detection, all samples were diluted for 20 times and filtered.



Fig. 1. Effect of pH value on COD and UV_{254} reduction

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TABLE-1 EFFECT OF pH VALUE ON COD AND UV ₂₅₄ REDUCTION (RESULTS OF 20 TIMES DILUTION)					
Number of reaction system	UV ₂₅₄	COD (mg/L)			
#1, #2 (pH = 2.04)	0.408/0.408	286, 286, 287, 291, 303, 308			
#3, #4 (pH = 2.99)	0.398/0.396	215, 221, 218, 189, 196, 200			
#5, #6 (pH = 3.99)	0.434/0.433	232, 230, 230, 230, 232, 229			
#7, #8 (pH = 5.00)	0.498/0.498	271, 284, 283, 268, 269, 269			
#9, #10 (pH = 6.12)	0.492/0.490	290, 310, 311, 296, 309, 313			
#11, #12 (pH = 7.03)	0.545/0.545	303, 315, 318, 302, 320, 315			
#13, #14 Original	0.565/0.563	360, 374, 382,379, 380, 380			
leachate (without					
Fenton's reagent)					

From the results, it was found that the pH condition had significant effect on both COD and UV_{254} reduction. The optimum pH condition was pH = 2.99, under which condition, COD removal was 45.21 % and UV_{254} reduction was 29.60 %, both were the peak compared with other 5 different pH values. Fenton's reagent can hardly play its role in neutral condition, which is a challenge for its practical application.

Effect of Fe^{2+}/H_2O_2 ratio on COD and UV_{254} reduction: Fe²⁺/H₂O₂ ratio was another important parameter for determined the Fenton's reaction. Firstly, with pH value being adjusted into 3.0, ten different Fe²⁺/H₂O₂ ratio were tested, namely 1/10, 1/20, 1/30,1/40, 1/50, 1/60, 1/70, 1/80, 1/90 and 1/100 (with the dosage of Fe²⁺ fixed as 10 mL 0.018 M solution). From the preliminary results (Table-2), it was found 1/10, 1/20 and 1/30 were obviously better than other Fe²⁺/ H₂O₂ ratios, based on which, further experiments were carried out to compare the ratios of 1/10, 1/20, 1/30 and 1/40.

Based on the results of the further experiment, it was found the best Fe^{2+}/H_2O_2 ratio among the ten ratios was 1/30, under which condition, COD removal was 53.06 %. Meanwhile, when the Fe^{2+}/H_2O_2 ratio was 1/30, UV₂₅₄ reduction was 40.58 %.

Comprehensive experiment results: To check the optimum pH condition and Fe^{2+}/H_2O_2 ratio, a series of comprehensive experiments were carried out. Experiment design was listed in Table-3. Stirring rate was 200 rpm and reaction time was 0.5 h. Results of comprehensive experiment were listed in Table-4.

Based on the results of the experiments, it was found among these nine systems, system #5 presented the best landfill degradation effect, with the pH value being 3.0 and the Fe²⁺/ H₂O₂ ratio being 1/30, which provides the further evidence for the optimized pH condition and Fe²⁺/H₂O₂ ratio obtained from former experiments.

Conclusion

Based on experimental study of Fenton's process in the treatment of landfill leachate, the conclusions were listed as

TABLE-2					
EFFECT OF Fe ²⁺ /H ₂ O ₂ RATIO ON COD AND UV ₂₅₄ REDUCTION (RESULTS OF 20 TIMES DILUTION)					
Fe^{2+}/H_2O_2 ratio	UV ₂₅₄	UV ₂₅₄ Reduction (%)	Average COD (mg/L)	COD Reduction (%)	
V_{Fe2}^{+} = 10 mL, $V_{H_2O_2}$ = 1.8 mL Fe ²⁺ /H ₂ O ₂ = 1/10	0.556	38.36	190	47.22	
V_{Fe2}^{+} = 10 mL, $V_{H_2O_2}$ = 3.6 mL Fe ²⁺ /H ₂ O ₂ = 1/20	0.554	38.58	188	47.77	
V_{Fe2}^{+} = 10 mL, $V_{H_2O_2}$ = 5.4 mL Fe ²⁺ /H ₂ O ₂ =1/30	0.536	40.58	169	53.06	
V_{Fe2}^{+} =10 mL, $V_{H_2O_2}$ = 7.2 mL Fe ²⁺ /H ₂ O ₂ = 1/40	0.580	35.70	199	41.94	
Original leachate (without Fenton's reagent)	0.902	-	360	-	

TABLE-3									
			COMPREHENS	SIVE EXPERIM	IENT DESIGN	N			
pH =	= 2.5 (Fe ²⁺ /H ₂ O ₂ ra	tio)	pH =	$pH = 3.0 (Fe^{2+}/H_2O_2 ratio)$			$pH = 4.5 (Fe^{2+}/H_2O_2 ratio)$		
#1 (1/20)	#2 (1/30)	#3 (1/40)	#4 (1/20)	#5 (1/30)	#6 (1/40)	#7 (1/20)	#8 (1/30)	#9 (1/40)	
				TABLE-4					
COMPREHENSIVE EXPERIMENT RESULT									
Number (Fe^{2+}/H_2O_2 ratio)		UV ₂₅₄	UV ₂₅₄ Reduction (%)		Average COD (mg/L) COD	Reduction (%)		
	#1(1/20) pH = 2.5	5	0.556	45.	76	414		33.65	
	#2(1/30) pH = 2.5	5	0.543	47.	02	406		34.94	
	#3(1/40) pH = 2.5	5	0.598	41.	56	438		29.81	
	#4(1/20) pH = 3.0)	0.554	45.	95	417		33.17	
	#5(1/30) pH = 3.0)	0.495	51.	71	353		43.43	
	#6(1/40) pH = 3.0)	0.572	44.	20	458		26.60	
	#7(1/20) pH = 3.5	5	0.557	45.	56	421		32.53	
	#8(1/30) pH = 3.5	5	0.501	51.	12	415		33.49	
	#9(1/40) pH = 3.5	5	0.569	44.4	49	461		26.12	
Original lead	chate (without Fen	ton's reagent)	1.025	-		624		-	



Fig. 2. Effect of Fe2+/H2O2 ratio on COD and UV254 reduction

follows: (i) Fenton's reaction can only take place in acid condition. To make full use of Fenton's reagent, pH = 3.0 is the optimum value for landfill leachate treatment. (ii) $Fe^{2+}/$ H₂O₂ ratio was another important parameter for Fenton's reaction used in organic wastewater treatment. With too high

 Fe^{2+} concentration, waste of H_2O_2 may be too much; with too low Fe²⁺ concentration, generation of radical ·OH can not be realized effectively. 1/30 is the best Fe²⁺/H₂O₂ ratio for landfill leachate treatment in this study. (iii) Through numerous experiments, it was found, Fenton's process had effect in real landfill leachate treatment. By controlling reaction condition, the removal of COD and UV_{254} reduction can both be above 40 %. (iv) The highest COD removal in all experiments was only 53.06 %, which indicates Fenton's process should be integrated with other procedures to give the acceptable water quality. We also carried out a series of internal electrolysis experiment with iron scraps and activated carbon granular in landfill leachate treatment. Preliminary results show integration of Fenton's process with internal electrolysis process might be a possible solution to provide a flexible and convenient method for landfill leachate treatment.

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