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# Oxidation of acetyl-pyrimidine wastewater by Fenton process

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## ABSTRACT

In this study, contaminant degradation, priority performance, intermediates, and kinetics in catalytic oxidation of Acetyl-Pyrimidine wastewater by Fenton reagent were investigated. In batch experiments, the main parameters that govern the complex reactive system, i.e. temperature, pH,  $Fe^{2+}$ , and  $H_2O_2$  initial concentrations have been studied. The results showed total COD and Acetyl-Pyrimidine removal was above 50% and 90% respectively under the following oxidation conditions: T at 25°C, pH at 3.5,  $H_2O_2$  at 300 mmol/l,  $Fe^{2+}$  at 40 mmol/l, with the reaction time 2 h. Control experiment and UV-Infrared spectrums analysis indicated that Acetyl-Pyrimidine was removed prior to the small molecule organic. The reaction fit to a first-order rate equation and the activation energy of Pyrimidine was 2.365 kJ/mol. **Key words** | acetyl-pyrimidine, catalytic oxidation, Fenton reagent, kinetics,

UV-infrared spectrums

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## INTRODUCTION

Acetyl-Pyrimidine (2-methyl-4-amino-5-acetylamidemethyl-Pyrimidine, Pyrimidine for short) is used as an important intermediate for the synthesis of pharmaceutical such as Vitamin  $B_1$ , which is a medicine used for the treatment of beriberi, dropsy and cardiopathy, etc. (Kerr 2007; Silver *et al.* 2007). In north China, at least 1,000 tons of Pyrimidine was produced per year. What is more, vast quantities of wastewaters containing Pyrimidine were generated during the manufacturing processes. Because of its toxic and recalcitrant nature (An 2008; Xie *et al.* 2008), Pyrimidine makes pollution serious. It is, therefore, necessary to treat the Pyrimidine wastewater before discharging.

For this drug is produced mainly in China and some developing countries, so such studies were few to crawl out of other countries' wastewater treatment literatures. In China, An (2008) has reported the removal of Pyrimidine using Potassium Permanganate Pretreatment-Denitration/ Bio-ferric Process. Xie *et al.* (2008) stated that it was effective by dynamic mixing aeration micro-electrolysis process. Wet oxidation technology shown as an effective doi: 10.2166/wst.2010.491 method, Qian *et al.* (2007) observed that Pyrimidine can be easily removed by this process. Catalytic oxidation and resin adsorption process has been used to treat pyrimidine and get a good result (Yan *et al.* 2001; Cheng & Yun 2005). However, conventional biological wastewater treatment processes (e.g. activated sludge) are not effective in treating Pyrimidine water because of its biological toxicity (An 2008; Wang *et al.* 2008). Thus, although some methods are effective, they deal with high costs and easily cause secondary pollution, or just transfer pollutants from the environment. These problems are the limiting factors in industrial wastewater treatment.

Fenton oxidation is the most common advanced oxidation process for the treatment of organic contaminants because of its simplicity and efficacy (Maletzky & Bauer 1998; Zoh & Michael 2002; Alexandra *et al.* 2005). And Fenton's reagent has been applied for the treatment of several industrial pollutants and industrial effluents were quite abundant, and the effect of pH,  $Fe:H_2O_2$  molar ratio, temperature, etc. were investigated in many studies

(Chamarro *et al.* 2007; Ghaly *et al.* 2007; Lu *et al.* 2003; Oh *et al.* 2003; Ipek *et al.* 2006). In this study, determining the potential to use the Fenton reaction to destroy Pyrimidine in aqueous solution; using a modified pseudo-first-order model in order to determine the kinetic constants; various factors that are important to maximize the oxidation were studied, including  $H_2O_2$  concentration,  $Fe^{2+}$  concentration, and temperature. Furthermore, the priority degradation and intermediate of Pyrimidine degradation were also investigated. The objective of this study is to develop an effective oxidation process for the degradation of Pyrimidine between some small organic molecule by Fenton process.

## MATERIALS AND METHODS

 Table 1
 Wastewater qualities and analytical methods

#### Wastewater sources

The raw Pyrimidine wastewater quality and analysis methods (APHA 1992) are shown in Table 1.

The main organic pollutants in this kind of wastewater was Pyrimidine, which structure was shown in Table 1, and we can see the pollutant was heterocyclic nitrogencontaining compound, which is stable and asymmetric, and they can inhibit the biological treatment to some extent (An 2008). In addition,  $SO_4^{2-}$  and Na<sup>+</sup> were also found in this wastewater.

## Chemicals

Ferrous sulphate heptahydrate (FeSO<sub>4</sub>·7H<sub>2</sub>O), AG was used as sources of the ferrous solutions, which were prepared daily; Stock hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) of 30% purity AG was used to prepare the H<sub>2</sub>O<sub>2</sub> solution; Catalase powder (AG) was made into 3.0 g/l catalase solution; Sulfuric acid, ethanol, sodium propionate and sodium hydroxide were all analytical grade. MERCK peroxide paper (range: 0.5-25 mg/L), German. Solutions were prepared using deionized water.

#### Experimental procedure

The experiments were conducted in the Coagulation test mixer (ZR4-6), which has 6 plexiglass cylindrical with 1.01 volume. During each experiment, 100-fold diluted wastewater was prepared by ultrapure deionized water as solvent. The laboratory reactor unit was filled with 1,000 ml of this diluted wastewater solution. In the test runs, the Fenton processes usually lasted 2 h at temperatures 25°C, except in the oxidation priority test and kinetics section. pH of the solution was set at the desired value by the addition of a H<sub>2</sub>SO<sub>4</sub> or a NaOH solution. Then, a given weight of iron salt which would not cause pH change was added. At last, H<sub>2</sub>O<sub>2</sub> at different amounts was injected before the beginning of each experiment. The dose of H<sub>2</sub>O<sub>2</sub> chosen between

Item	Value	Analysis method	
COD (mg/L)	120,000-160,000	GB11914 ~ 89 Potassium dichromate method	
BOD (mg/L)	2,000-2,500	Lovibond BOD <sub>5</sub>	
Pyrimidine (mg/L)	120,000-160,000	HPLC	
Colority (times)	2,000	Dilution method	
pH	9.3-9.8	Electrode (TI2100)	
Source	Tianjin Zhongjin co., China		
Flow rate (m <sup>3</sup> /d)	About 12.5		
Structure of the main pollutant	NH <sub>3</sub> N CH <sub>2</sub> NHCOCH <sub>3</sub>		
	and the first of the second second		
	CH <sub>3</sub>		

10 to 500 mmol/l,  $Fe^{2+}14$  to 128 mmol/l, pH 1 to 10, temperature 25 to 60°C according to our needs (Ghaly *et al.* 2001). 100 ml wastewater sample was taken into a test tube, and one drop of enzyme catalase solution (0.327 mg/L COD) was added to each sample in order to decompose any residual  $H_2O_2$  which can be determined by a peroxide paper (Peyton 1990).

#### Analytical methods

Coagulation test mixer (ZR4-6) was provided by Aishengte Shanghai Environmental Protection Technology Co., Ltd. special; UV–Vis spectrophotometer (2550) purchased from Shimadzu Corporation, Japan.

Pyrimidine concentration was determined by an Agilent 1100 Series HPLC instrument (USA). The HPLC was equipped with a variable wavelength detector and an autosampler. Liquid samples were firstly filtered through sterile 0.45 um syringe-microfilter before the analysis by HPLC. An Adsorbosphere C-18 ( $150 \text{ mm} \times 3.9 \text{ mm}$ , Nova-Pak) was used with an isocratic mobile phase consisting of 75% methanol and 25% water (volume %) at a flow rate of 1.0 ml/min. A UV detector setting to 232 nm was used (An 2008), and Pyrimidine peak at retention time of 4.8 min was observed. The LODs of Pyrimidine is 0.02 mg/L and the recoveries between 94.1–96.3%.

In addition, COD values of wastewater samples were also investigated in order to estimate the effect of Fenton reagent on Pyrimidine wastewater degradation.

## **RESULTS AND DISCUSSION**

## The effect of pH

pH affect HO radicals production for Fenton reagent systems. Figure 1 shows the effect of the pH with  $H_2O_2$ 100 mmol/l, Fe<sup>2+</sup>100 mmol/l. As expected, Pyrimidine and COD removal rates varied with the pH value, and the maximum Pyrimidine degradation and COD removal rates were 74.89 and 42.16% respectively at pH = 3.5. In addition, the irritating smell reduced significantly at this point. Ghaly *et al.* (2001) reported that at pH = 3.0, the *p*-chlorophenol got a maximum degradation by Fenton process. For pH above 4.0 the degradation strongly decreases because at higher pH, iron precipitates as



Figure 1 | Pyrimidine degradation as a function of the pH by using Fenton processes:  $[Fe^{2+}]_0 = 100 \text{ mmol/l}, [H_2O_2]_0 = 100 \text{ mmol/l}, [Pyrimidine]_0 = 1,000 \text{ mg/L}.$ 

hydroxide and that reduces the transmission of the radiation (Faust & Hoigne 1990). More over, this little loss of COD was noted during this process, which indicating that Pyrimidine converted to intermediate species, but not be oxidized completely. Using further oxidation or biological treatment is still under study.

## The influence of initial H<sub>2</sub>O<sub>2</sub> concentration

Figure 2 shows the effect of the initial  $H_2O_2$  concentration on the degradation of Pyrimidine by Fenton reagent. The degradation of Pyrimidine was increased by increasing the concentration of  $H_2O_2$ , but after a certain volume (300 mmol/l), the removal was no longer increased obviously. This may be due to the auto-decomposition of  $H_2O_2$  to oxygen, water and the recombination of HO radicals. Since HO radicals react with  $H_2O_2$ ,  $H_2O_2$  itself contributes to the HO scavenging capacity (Lin & Chang 2000; Montserrat *et al.* 2002). Therefore,  $H_2O_2$  should be added at an optimal concentration to achieve the best degradation.



Figure 2 Pyrimidine degradation as a function of initial hydrogen peroxide concentration by using Fenton processes: pH = 3.5,  $[Fe^{2+}]_0 = 100 \text{ mmol}/J$ ,  $[Pyrimidine]_0 = 1,000 \text{ mg/L}$ .





#### The effect of the amount of Fe<sup>2+</sup>

Iron in its ferrous form acts as catalyst and requires a working pH below 4.0 (Ghaly *et al.* 2007). To obtain the optimal Fe<sup>2+</sup> amounts, the investigation was carried out with various amounts of the iron salt. Figure 3 shows that the degradation of Pyrimidine as a function of the added Fe<sup>2+</sup>. The degradation rate of Pyrimidine distinctly increased with increasing amounts of iron salt, however, when the addition of Fe<sup>2+</sup> above 40 mmol/l it did not affect the degradation obviously, even if the concentration threefold increased. Moreover, a higher addition of Fe<sup>2+</sup> resulted in high turbidity, brown color, and caused the recombination of HO radicals as well. In this case, Fe<sup>2+</sup> reacted with HO radicals as a scavenger (Wailling 1975). Apart from this, more iron salt improved the cost greatly and made the follow-up treatment more difficult as well.

## Oxidation priority of pyrimidine by Fenton reagent

To assess whether there was oxidation priority of Pyrimidine by Fenton reagent, samples for the studies were



## Intermediates analysis

A series of experiments was performed to determine the liquid phase intermediate in experiments.

Figure 5 shows the UV spectrums of raw Pyrimidine wastewater and the treated Pyrimidine wastewater by Fenton reagent. Pyrimidine has only one peak at 245 nm in UV spectrogram at pH = 3.5, and this peak was caused by N=C-N=C from aromatic. After oxidation by Fenton



Figure 4 | Prior character in catalytic degradation of COD and Pyrimidine by Fenton process:  $[H_2O_2]_0 = 300 \text{ mmol/l}, [Fe^{2+}]_0 = 40 \text{ mmol/l}, pH = 3.5, [Pyrimidine]_0 = 1,000 \text{ mg/L}$ 



Figure 5 | UV spectrums of raw Pyrimidine wastewater and the treated Pyrimidine wastewater:  $[H_2O_2]_0 = 300 \text{ mmol/l}$ ,  $[Fe^{2+}]_0 = 40 \text{ mmol/l}$ , pH = 3.5,  $[Pyrimidine]_0 = 1,000 \text{ mg/L}$ .

reagent this peak disappeared but with not any other peaks formed, which indicated that the aqueous solution has been a non-conjugated system of functional groups, namely, decomposition of Pyrimidine and formed other substances.

In order to further examine the productions of this process system, infrared detection has been carried out, Figure 6 shows the IR of raw Pyrimidine and disposed Pyrimidine by Fenton reagent respectively.

From Figure 6 we can see that the structure of Pyrimidine has greatly changed after Fenton process. Some representative characteristic peaks disappeared and appeared, which are mainly:  $3,382.30 \text{ cm}^{-1}$ , peaking width and strong, -OH stretching vibration;  $2,176.78 \text{ cm}^{-1}$ , cumulative double bond, initially judged as -C=C=N;  $1,578.59 \text{ cm}^{-1}$  and  $1,415.62 \text{ cm}^{-1}$ , showing the appearance of anhydride (maybe acetic acid or imide);  $1,352.30 \text{ cm}^{-1}$ , likely being nitrate  $-NO_2$  (or  $NO_3^-$ ) stretching vibration;  $1,129.76 \text{ cm}^{-1}$ , for -C=O- or -C-O- stretching vibrations. The reason is that Pyrimidine contains four carbons in its ring and several carbon atoms and nitrogen atoms in



Figure 6 | Infrared spectrums of raw Pyrimidine wastewater and disposed water.

its side chains. Once the structure of Pyrimidine broken, these small structures appeared, then we know there still some other molecules. Moreover, Pyrimidine water is solution not colloid, thus, coagulation/sedimentation has little effect on it, thus, it can not remain in sludge. Therefore, it can be seen that Pyrimidine was decomposed into simpler structures and smaller molecules, which were easily bio-degradable materials (Kao & Wu 2000). Both Marco et al. (1997) and Chamarro et al. (2001) reported that using a Fenton process can improve the bio-degradability of pollutants obviously. Then, activated sludge treatment system could be used for its economics. An Y. reported B/C of Pyrimidine wastewater increased to 0.325 from 0.09 after Fenton process (An et al. 2008), Therefore, a semi-oxidation of Pyrimidine could be an attractive way, for it made suitable conditions for biological treatment as the follow-up step.

## Kinetics of pyrimidine degradation

A series of batch experiments was conducted to determine the temperature dependence of Pyrimidine oxidation by Fenton reagents (Zoh & Michael 2002). Initial conditions were fixed at Pyrimidine 1,000 mg/L, H<sub>2</sub>O<sub>2</sub> 300 mmol/l; Fe<sup>2+</sup> 40 mmol/l; the pH was fixed at 3.5 and experiments were performed at temperatures 25°C, 40°C, 50°C, 60°C. Figure 7 shows the results of the oxidation of Pyrimidine by Fenton reagent at different temperature. At 25°C and pH = 3.5, the oxidation of Pyrimidine is very fast, and after 20 min, only 30% Pyrimidine remains. In this period of time, these reactions follow pseudo-first-order kinetics with respect to Pyrimidine,

$$-d(Pyrimidine)/dt = -k(Pyrimidine)$$
(1)

which can be integrated between t = 0 and t = t, yielding:

 $\ln\left[(\text{Pyrimidine}_0)/\text{Pyrimidine}\right] = kt \tag{2}$ 

In amore accurate way it should be necessary subtract, in this equation, to the numerator and denominator, the fraction of the initial Pyrimidine and/or initial products generated after the oxidation that are difficult to be further removed by the Fenton system. This fraction corresponds to organic matter not oxidizable under the test conditions after an "infinite" period (Pyrimidine<sub>∞</sub>), in this case





after a period where the oxidation system in almost ineffective: 2 h. Therefore, this expression will be modified to (Lucas & Peres 2009):

### $\ln [(Pyrimidine_0 - Pyrimidine_\infty)/(Pyrimidine)]$

$$-\operatorname{Pyrimidine}_{\infty})] = kt \tag{3}$$

Table 2 shows the rates of Pyrimidine oxidation. These show an excellent agreement between pseudo first-order reaction mechanism and observations over the temperature range (25–60°C);  $R^2$  ranges from 0.980 to 0.996. And this result was similar with Kyung-Duk Zoh's report (Zoh & Michael 2002).

The temperature dependence of the observed rate constants can be represented by the Arrhenius equation, as follows:

$$K = A \exp\left(E/RT\right) \tag{4}$$

which is equivalent to

 $\lg K = \ln A_0 - E/(RT)$ 

 
 Table 2
 Pseudo first-order rate constants (min<sup>-1</sup>) of Fenton Pyrimidine oxidation at different temperatures

T (°C)	Pyrimidine $_{\infty}$ (mg/L)	R <sup>2</sup>	K (min <sup>-1</sup> )
25	41.7	0.9796	0.0662
40	70.1	0.9957	0.1127
50	95.9	0.9915	0.1247
60	68.9	0.9932	0.1916

Pyrimidine<sub>0</sub> = 1,000 mg/L.



Figure 8 Arrhenius plot of Pyrimidine oxidation rate constants by Fenton process.

where,  $A_0$  the pre-exponential factor (min<sup>-1</sup>), E is the activation energy (J mol<sup>-1</sup>), T is the temperature (K), R is the gas constant (8.314 J mol<sup>-1</sup> K<sup>-1</sup>). Figure 8 shows the Arrhenius plot of the reaction rate constants of Pyrimidine. As shown in this figure, the E/R value is 2.8451 × 10<sup>3</sup> for Pyrimidine, and therefore activation energy of Fenton oxidation for Pyrimidine is 2.365 kJ/mol. This activation energy value is match well with other results (Lin & Lo 1997), and it is important to minimize the H<sub>2</sub>O<sub>2</sub> required, especially for treating the full-scale Pyrimidine wastewater. Moreover, using too high concentration of Fenton reagent may result in excessive iron sludge (Sedlak & Andren 1991), though part of it can be made into coated-sand which can be used for filtration technology (Chang *et al.* 2008), we still want to reduce the use of it for economic reasons.

## CONCLUSIONS

(5)

The Fenton reaction is effective in the degradation of Pyrimidine, the total COD removal was over 50%, and the Pyrimidine degradation rate could be more than 90% under the following oxidation conditions: reaction time 1 h, T 25°C, Fe<sup>2+</sup> 40 mmol/l, and H<sub>2</sub>O<sub>2</sub> 300 mmol/l. The aromatic compounds may be removed prior to the small organic molecules by Fenton reagents. After infrared and ultra-violet spectrograph analysis, we knew that the Pyrimidine has been decomposed into materials which were easily degraded.

The reaction rate coefficient was strongly dependent on temperature, higher the temperature higher the coefficient, K increased from 0.0662 min<sup>-1</sup> to 0.1916 min<sup>-1</sup> with temperature from 25°C to 60°C, this Fenton catalytic degradation of Pyrimidine followed first-order kinetic model and obtained activation energy for 2.365 kJ/mol.

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