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Kinetics of aerobically activated sludge on terylene artificial silk printing and dyeing wastewater treatment

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Abstract: Aerobically activated sludge processing was carried out to treat terylene artificial silk printing and dyeing wastewater (TPD wastewater) in a lab-scale experiment, focusing on the kinetics of the COD removal. The kinetics parameters determined from experiment were applied to evaluate the biological treatability of wastewater. Experiments showed that COD removal could be divided into two stages, in which the ratio BOD/COD (B/C) was the key factor for stage division. At the rapid-removal stage with B/C>0.1, COD removal could be described by a zero order reaction. At the moderate-removal stage with B/C<0.1, COD removal could be described by a first order reaction. Then Monod equation was introduced to indicate COD removal. The reaction rate constant (K) and half saturation constant (K_S) were 0.0208−0.0642 L/(gMLSS)·h and 0.44−0.59 (gCOD)/L respectively at 20 °C−35 °C. Activation energy (E_a) was 6.05×10^4 J/mol. By comparison of kinetic parameters, the biological treatability of TPD wastewater was superior to that of traditional textile wastewater. But COD removal from TPD-wastewater was much more difficult than that from domestic and industrial wastewater, such as papermaking, beer, phenol wastewater, etc. The expected effluent quality strongly related to un-biodegradable COD and kinetics rather than total COD. The results provide useful basis for further scaling up and efficient operation of TPD wastewater treatment.

Key words: Process kinetics, Aerobically activated sludge, COD (chemical oxygen demand), Biological treatability, Printing and dyeing wastewater

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INTRODUCTION

In order to get pliable and elegant terylene fabric just like silk, terylene greige cloth is always pretreated with alkali-decomposition process wherein terylene greige cloth is hydrolyzed to some extent in NaOH solution at certain temperature (T) and pressure. During this process, the superficial terylene fibre is peeled off from the cloth and dissolved into solution, in which terylene acid (TA) and ethylene glycol are discharged as pollutants in wastewater. The obtained terylene fabric with silken wrinkle and soft feeling is called artificial silk fabric. The alkali-decomposition of terylene can be described by the chemical equation below.

$$\begin{aligned}&\mathrm{H[\text{-}OOC\text{-}Ar\text{-}COO\text{-}CH_2CH_2\text{-}]_nOH + NaOH}\\&\rightarrow \mathrm{Na[\text{-}OOC\text{-}Ar\text{-}COO\text{-}CH_2CH_2\text{-}]_mOH}\\&\mathrm{+(Na\text{-}OOC\text{-}Ar\text{-}COO\text{-}Na)}\\&\mathrm{+(HO\text{-}CH_2CH_2\text{-}OH)}\qquad n>m\end{aligned}$$

The flow sheet of terylene fabric treatment is shown in Fig.1.

The wastewater from the alkali-decomposition process mixed with wastewater from printing, dye-

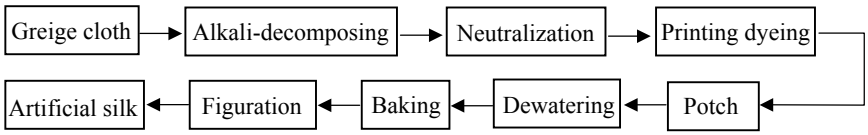

Fig.1 Flow sheet of terylene fabric treatment

ing, potch and other processes is named terylene artificial silk printing and dyeing wastewater (TPD wastewater).

TPD wastewater, which arose in the 1980's and developed in the 1990's, is now the popular industrial wastewater in China. In spite of the active research in this area, wastewater has not yet been well understood. Not enough knowledge and experience on wastewater treatment have been obtained for engineering design. Experimental investigation showed that the aerobically activated sludge process did not work well in the treatment of TPD wastewater (Han, 1986; Cai and Zhou, 1989; Wang *et al*., 1998). Some studies attributed the dissatisfaction to the hard biodegradability of TA in TPD wastewater (Wang *et al*., 1998; Ma *et al*., 1999; Zhou and Zhan, 1999). Nevertheless, TA was proved to be readily bio-degradable in aqueous solution or wastewater under aerobic conditions (Tong and Bai, 1990; Wan *et al*., 1990; Zhao, 1994; Cheng *et al*., 1997; Guan *et al*., 2003). It was also reported that sludge activity was inhibited and substrate removal rate decreased remarkably at mixed liquor pH>9.4 as the TPD wastewater was treated by aerobic biological process (Guan *et al*., 2002).

It can be seen that the factors related with the biological treatability of TPD wastewater may be complicated. There is still lack of accurate information on the factors affecting substrate removal from the wastewater. Few reports of work on the kinetics are available. So it is necessary to conduct investigation on the kinetics to obtain biological kinetic parameters which are essential for TPD wastewater treatment. In this presentation, chemical oxygen demand (COD) removal kinetics is introduced to evaluate the wastewater biological treatability with aerobically activated sludge process. Emphasis was placed on comparison between the kinetics of TPD wastewater and the other types of wastewater so that the COD removal from TPD wastewater could be evaluated.

MATERIAL AND METHOD

Material

The wastewater in the experiment was taken from the central pump station for TPD wastewater in Saoxing County, Zhejiang Province, P. R. China. Results of an about one year survey of main pollutants in the wastewater are given in Table 1. As a new kind of textile industry wastewater, TPD wastewater characterized by high pH, COD and color (COL) is different from traditional printing and dyeing wastewater. COD varies from 780 mg/L to 3116 mg/L; and biological oxygen demand for 5 days (BOD) 325 mg/L to 1436 mg/L. TA ranging from 386 mg/L to 1279 mg/L is the characteristic pollutant controlling 40%–78% of the total COD in TPD wastewater. In this study, COD was an overall measure of substrate concentration in the wastewater.

Activated sludge for the experiment was obtained from the treatment facility for pesticide wastewater. Sludge was acclimated in a laboratory reactor running in a fill and drawn mode for the particular reaction conditions (concentration, temperature, dissolved oxygen, etc.). After acclimation,

Table 1 Concentration values of TPD wastewater

Indexes	pH	COD (mg/L)	BOD (mg/L)	TA (mg/L)	COL (unit)	PO_4^{-3}-P (mg/L)	NH_4-N (mg/L)
Range	8.2–12.8	780–3116	325–1436	286–1279	250–600	1.8–10.2	1.8–42.2
Average	–	1780	703	710	–	4.3	10.2

the sludge showed good performance characterized by high metabolic activity and setting ability.

Setup and procedure

TPD wastewater was treated with a laboratory scale aerobically activated sludge process. A schematic drawing of the experimental setup is shown in Fig.2.

In order to eliminate the inhibition from high pH and shortage of N and P, the feeding wastewater was first adjusted to pH≤10.0 and COD:N:P= 200:5:1. A peristaltic pump was used to supply wastewater continuously to a 9.0 L plexiglass reactor charged with mixed activated sludge capable of carbon oxidation. Reaction temperature was controlled with a heater and a temperature controller. The wastewater and sludge in the reactor were uniformly and thoroughly mixed by compressed air supplied through diffusers at the bottom of the reactor. As mixed liquor passed through the sludge precipitator, the sludge was separated and refluxed to the reactor at the bottom, while effluent flowed into the storage vessel from the weir.

The reactor was operated under conditions as follows: Temperature (T) of 15 °C to 35 °C, mixed liquor suspended solid (MLSS) 2.5–3.5 g/L, dissolved oxygen (DO) of 3.0–5.5 mg/L with bubbling at air to wastewater ratio (A/W) of 15–20 m^3/m^3. Sludge volume index (SVI) was controlled between 60 ml/g and 100 ml/g to achieve good flocculability. The optimum operational factors are given in Table 2.

Significant parameters for wastewater treatment

Batch treatment of wastewater was carried out with the same system as the continuous one except that the wastewater and sludge were poured into the reactor almost simultaneously. In this system, the oxidation was maintained until a stable COD plateau and no appreciable biomass activities were observed.

Analytical methods

Changes of pH in the wastewater tank were monitored with a pH meter. The DO concentration in the reactor was determined with an oxygen probe connected to a dissolved oxygen meter. The analysis of the other items was conducted according to standard methods (Editorial Board of Environment Protection Bureau of China, 1997).

RESULT AND DISCUSSION

Stages of substrate removal

A series of batch treatments were done at 15 °C–35°C. The COD and MLSS in the mixed liquor at the beginning was 1751–2020 mg/L and 3.30–3.50

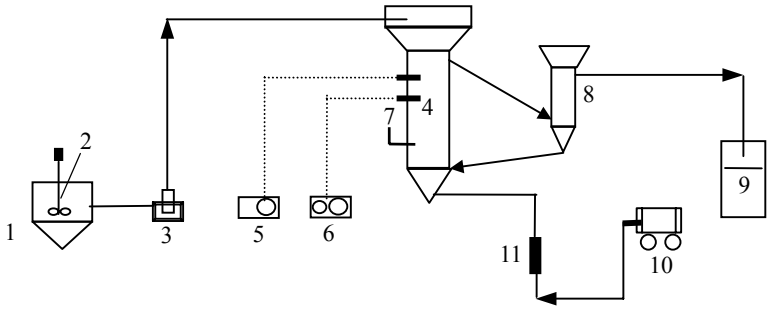

Fig.2 Schematic diagram of the experimental setup

1. Wastewater tank; 2. Stirrer; 3. Peristaltic pump; 4. Reactor; 5. DO monitor; 6. Temperature controller; 7. Thermometer; 8. Sludge precipitator; 9. Storage vessel; 10. Air compressor; 11. Gas flow meter

Table 2 Significant parameters for wastewater treatment

Parameter	A/W (m^3/m^3)	SVI (ml/g)	DO (mg/L)	MLSS (g/L)	Influent pH
Value	15–20	60–100	3.0–5.5	2.5–3.5	≤10.0

g/L, respectively. Substrate (*S*) was measured by COD and BOD in the experiment. The profiles of COD or BOD vs time are shown in Fig.3. The COD was removed rapidly until the ratio BOD/COD (B/C) approached 0.1. Then, COD was removed moderately. The substrate removal could be distinctly divided into a rapid-removal stage and a moderate-removal stage.

At *T*=25 °C, the rapid-removal stage lasted 12 h and the COD of mixed liquor decreased from 1758 mg/L to 404 mg/L; and B/C from 0.45 to 0.12. About 77.25% COD removal was achieved. Subsequently, only 218 mg/L COD was removed within 12 h at the moderate-removal stage. At *T*=30 °C, the rapid-removal stage lasted about 11 h and the COD of the mixed liquor decreased from 1890 mg/L to 410 mg/L; and B/C from 0.42 to 0.10. About 78.31% COD removal was obtained. At the 13 h moderate-removal stage, only 220 mg/L COD was removed.

Though the COD removal could be also divided into two stages at *T*=15 °C, the cut-off point of rapid-removal and moderate-removal prolonged to 20 h after operation. At *T*=35 °C, the rapid-removal stage lasted only 8 h and the COD removal efficiency reached 83.6%. The cut-off point of the two stages was at B/C = 0.12.

The COD removal was essentially a two-stage process. It was noticed that two stages occupied different retention time at different temperature. This is likely due to the temperature effect of the biological reaction. Consequently, COD removal stages division should be on the basis of B/C rather than reaction time. B/C of 0.1 was a turning point of COD removal rate.

Fig.3 also shows that COD concentration decreased linearly with time when COD≥300–400 mg/L. So that COD removal could be described by zero-order reaction. As COD<300–400 mg/L, it was the logarithm of *S* rather than *S* that showed linearity with reaction time. One of the typical profiles of *S* versus time at 30 °C is detailedly plotted in Fig.4. Therefore, COD removal was first-order reaction at low concentration. In other words, the distinct behavior of COD removal in relation to reaction time revealed that rapid-removal stage was dominated by zero-order reaction; moderate-removal stage by first order reaction. Rapid-removal stage removed 75%–90% COD and moderate-stage only accounted for 5%–15% COD removal.

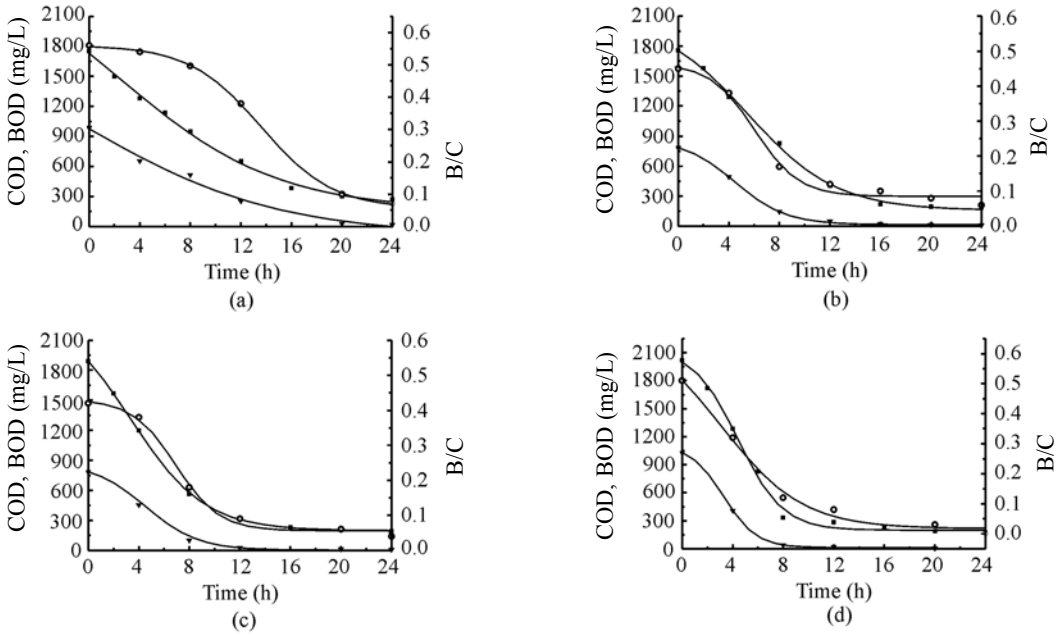

Fig.3 COD (▪), BOD (▾) and the ratio B/C (○) versus reaction time in batch treatment
(a) *T*=15 °C; (b) *T*=25 °C; (c) *T*=30 °C; (d) *T*=35 °C

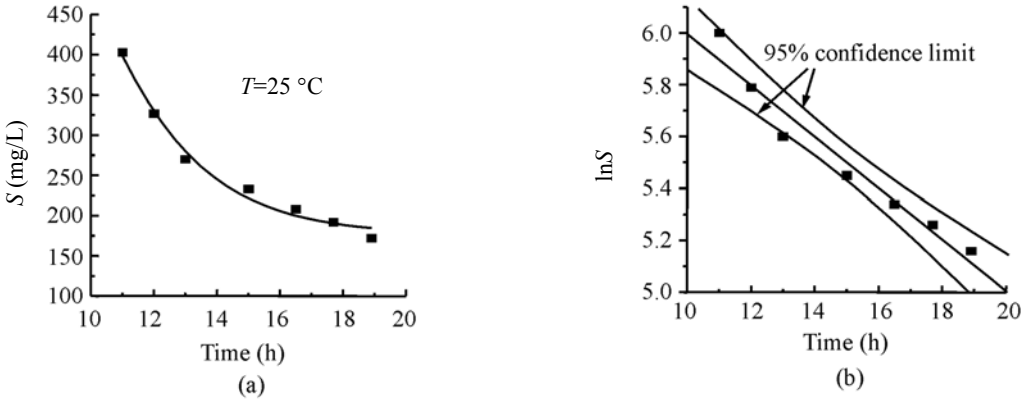

Fig. 4 The substrate (COD) concentration (a) and logarithm of substrate concentration (b) versus reaction time in a batch treatment at a low concentration

Un-biodegradable COD

Under the conditions of the experiment in a batch treatment system mentioned above, fractional COD in TPD wastewater could not be biodegraded and mineralized by activated sludge even aerobically oxidized for more than two weeks. As shown in Fig.5, about 82.4% COD was removed on the first day and only 6.6% COD removal was achieved the following 13 days. During the subsequent two weeks aeration, the COD concentration remained stable at 220 mg/L which we considered to be un-biodegradable COD (COD_N) or inert COD under the the presented conditions. By this means, we tested the COD_N of the TPD wastewater. COD_N stayed within 170–262 mg/L as COD in the range of measurements around a year (Table 3). The COD_N was the low limit COD of the effluent in the wastewater treatment under experimental conditions. In other words, the COD of the wastewater has a predominantly biodegradable characteristic, being only 5%–10% that of inert fractions.

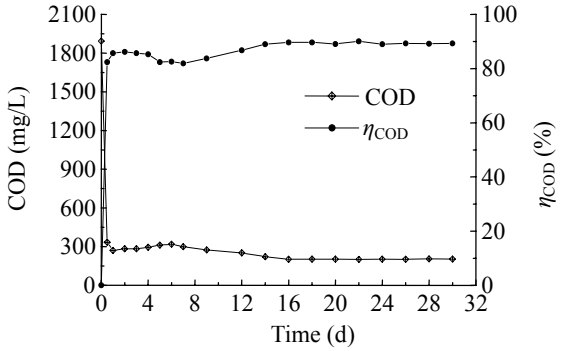

Fig.5 The COD concentration and removal efficiency versus reaction time in a batch treatment
(η_{COD}: efficiency of COD removal)

The hydraulic retention time (HRT) of aerobically activated sludge process was always less than one day in a wastewater treatment facility and, therefore the effluent sometimes contains organic material above the specified COD limits because non-biodegradable substrate requires long HRT to be mineralized. In order to get satisfactory effluent quality, for example with COD less than 150 mg/L, other processes should be integrated with the aerobic process.

Reaction constant of COD removal

Though traditional mathematical models formulated for overall substrate and biomass are limited to indicating the endogenous decay, they are still widely used in practice. The Monod equation is one of the simplest models for describing the biode-

Table 3 COD_N of TPD wastewater around a year

Month	April	June	August	October	November
COD_N (mg/L)	205±20	190±20	220±20	252±10	242±10

gradation of a substrate by an acclimated bacterial population:

$$\tau = -\frac{\mathrm{d}S}{X\mathrm{d}t} = \tau_{\max}\frac{S}{K_S + S} \tag{1}$$

$$\tau_{\max} = -\left(\frac{\mathrm{d}S}{X\mathrm{d}t}\right)_{\max} \tag{2}$$

Where, τ is specific rate of substrate removal (h^{-1}), $\tau_{\max}$ is maximum specific rate of substrate removal (h^{-1}), S is substrate concentration in reactor (g/L), X is concentration of biomass (g/L). S and X are measured with COD and MLSS, respectively. K_S is half saturation constant for substrate (gCOD/L).

For COD_N denoted by S_N is:

$$\tau = -\frac{\mathrm{d}S}{X\mathrm{d}t} = \tau_{\max}\frac{S - S_N}{K_S + S - S_N} \tag{3}$$

It is always assumed that in a complete mixing reactor of continuous flow system, COD concentration of the mixed liquor is equal to that of the effluent. The results above showed that COD removal could be described by first-order reaction when COD was at concentration of less than 400 mg/L. So Eq.(3) can be changed into:

$$\tau = -\frac{\mathrm{d}S}{X\mathrm{d}t} = \tau_{\max}\frac{S - S_N}{K_S} \tag{4}$$

As X is kept constant in a reactor, the rate of substrate removal can be expressed by:

$$V = -\frac{\mathrm{d}S}{\mathrm{d}t} = KX\left(S - S_N\right) \tag{5}$$

With:

$$K = \tau_{\max} / K_S \tag{6}$$

Where, V is rate of substrate removal (gCOD/L·h), K is reaction rate constant (L/(gMLSS)·h).

After integration, Eq.(5) is rewritten as:

$$\ln\frac{S_0 - S_N}{S - S_N} = K\ X\ t \tag{7}$$

Where, S_0 is substrate concentration of effluent (g/L).

Eq.(7) can be rearranged into the final form:

$$-\frac{\ln\left(S - S_N\right)}{X} = K\ t - \frac{\ln\left(S_0 - S_N\right)}{X} \tag{8}$$

The left hand side in Eq.(8) is represented by Y in the following discussion.

Essential data from wastewater treatment are listed in Table 4. Fig.6 shows a graphical representation of the linearized form of Y versus time (HRT). From the slope and the intercept of the straight line, it is easy to calculate the reaction rate constant K of the kinetic equation at different temperature.

Table 4 Experimental results related to COD removal

HRT, h		4.5	6.7	8	9	10	12	16	20	24
20 °C	S_0	0.917			1.113		1.181		1.316	1.254
	S	0.394			0.335		0.286		0.219	0.189
	X	3.730								
	Y	0.362			0.432		0.502		0.652	0.772
25 °C	S_0	1.253		1.401			1.486	1.455	1.571	1.400
	S	0.594		0.307			0.298	0.300	0.242	0.190
	X	3.128								
	Y	0.248		0.558			0.578	0.568	0.708	0.908
30 °C	S_0	0.971	0.787		0.642		0.992		0.626	0.820
	S	0.383	0.288		0.213		0.217		0.221	0.242
	X	2.781					2.846		2.700	
	Y	0.264	0.664		0.824		0.867		1.018	
35 °C	S_0		1.247	1.498		2.094	1.914	1.932	1.753	
	S		0.306	0.299		0.264	0.210	0.181	0.179	
	X		2.178	2.340			2.734		3.120	
	Y		0.757	0.762		0.872	0.937	0.984	1.107	

Note: (1) $-\frac{\ln(S-S_N)}{X}$ is denoted by Y; (2) the values of S_0, S and X are in g/L

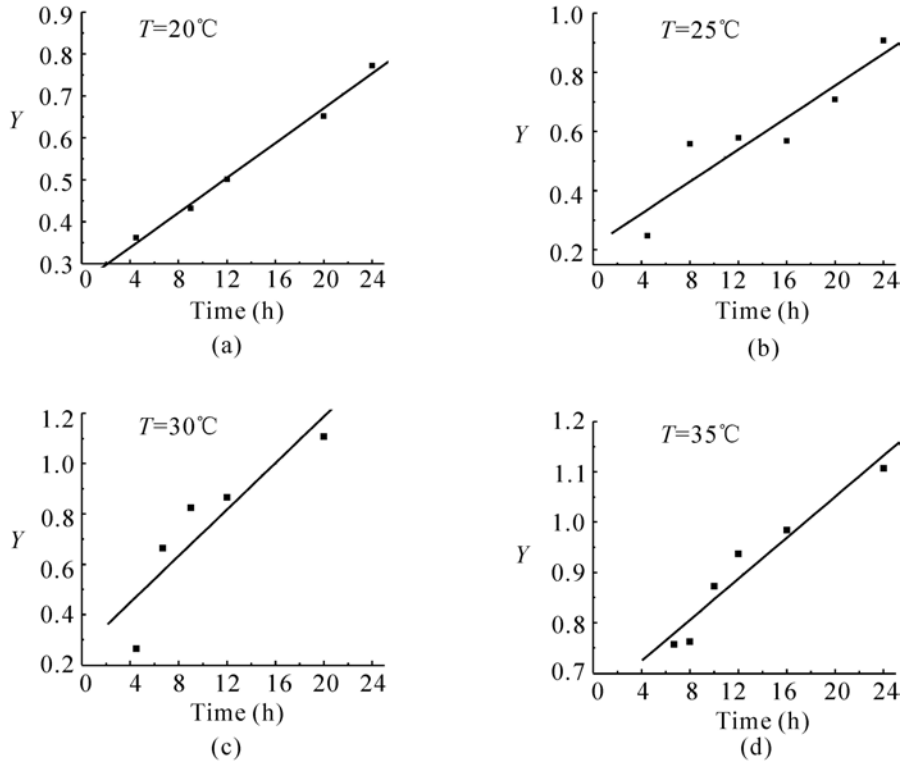

Fig. 6 Linear regression for kinetic equation of COD removal from TPD wastewater
(a) T=20 °C; (b) T=25 °C; (c)T=30 °C; (d)T=35 °C

Table 5 shows the K and kinetics equations described by Eq.(8). Under the experiment conditions, K increased from 0.0208 L/(gMLSS)·h to 0.0642 L/(gMLSS)·h as T rose from 20 °C to 35 °C. As T increased from 20 °C to 25 °C, then to 30 °C, ending at 35 °C, the value of K increased 29.8%, 71.1% and 60.6% correspondingly. The value of K at 35 °C was found to be more than 3 times that at 20 °C. It implicated that temperature had a remarkable influence on the substrate removal.

Reaction rate constants of some kinds of wastewater treated by aerobically activated sludge are given in Table 6. The value of K of TPD wastewater was only 2%–5% of that of domestic wastewater, about 10%–30% of that of beer, pharmaceutical or papermaking wastewater, 20%–70% of that of phenolic wastewater, but approximately 2 times that of traditional textile wastewater at 25 °C. This indi-

Table 5 Kinetic equations and reaction rate constants

T (°C)	20	25	30	35
Kinetic equation	Y=0.0208t + 0.2553	Y=0.0270t + 0.2138	Y=0.0462t + 0.2631	Y=0.0642t + 0.2041
Standard deviation	0.0179	0.0948	0.1642	0.0400
K (L/g·h)	0.0208	0.0270	0.0462	0.0642

Table 6 Reaction rate constants for different wastewater treated by aerobic biological process

Wastewater	Domestic	Papermaking	Pharmaceutical	Phenolic	Beer	Textile	TPD
K (L/(gMLSS)·h)	1.08–1.37	0.417	0.21–0.57	0.092	0.22	0.015	0.0208–0.0642
Literature cited	(Leslie *et al*., 1989)						This paper

cated the treatability of TPD wastewater was much worse than that of domestic or other industrial wastewater mentioned above, but was better than that of traditional textile wastewater.

The maximum specific rate of substrate removal, τ_{max}, was defined with Eq.(2) and determined in batch treatment. The value of K_S was obtained according to Eq.(6) with the help of the K described above. Consequently, K_S of TPD wastewater varied within the range 0.44 gCOD/L to 0.59 gCOD/L. Some values of K_S for industrial wastewater are given in Table 7. The value of K_S of TPD wastewater was higher than those of skim milk, glucose and soybean wastewater, closed to that of poultry wastewater, but lower than that of textile wastewater. It revealed that the treatability of TPD wastewater was inferior to that of textile wastewater but superior to those of the other industrial wastewater mentioned herein. The fact is in accordance with the results from the illustration of K.

Activation energy for COD removal

Arrhenius equation was engaged to describe the effect of temperature on reaction rate for COD removal from TPD wastewater. The logarithm form of the equation:

$$\ln K = -\frac{E_a}{R}\frac{1}{T} + \ln K_0 \qquad (9)$$

where, E_a is activation energy (J/mol), R is ideal gas constant (J/mol·k), K_0 is frequency constant (L/(gMLSS)·h).

Using the K with the corresponding temperature, a plot of $(-\ln K)^{-1}$ versus $1/T$ was established (Fig.7). From the slope and intercept, the filling values were found for the constants:

$$\ln K = -7279 \times \frac{1}{T} + 20.91 \qquad (10)$$

Then, E_a for COD removal was 6.05×10^4 J/mol.

Usually, E_a of wastewater treatment varied from 8.4×10^3 J/mol to 84×10^3 J/mol as the biological process was applied under aerobic conditions (Qing *et al*., 1986). So E_a of TPD wastewater treatment by aerobically activated sludge was on the high side, or 20%–50% more than that of domestic wastewater, but slightly less than that of starchy wastewater (Table 8).

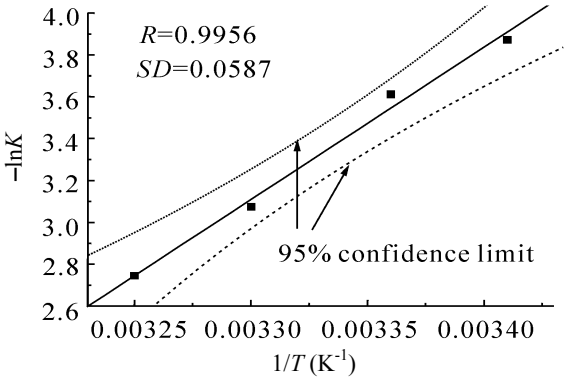

Fig.7 Negative logarithm of reaction rate constant (–ln*K*) versus reciprocal temperature (1/*T*) for TPD wastewater treatment by aerobic activated sludge

Table 7 Comparison of half saturation constant of wastewater bio-treatment

Wastewater	Skim milk	Glucose	Soybean	Poultry	Textile	Textile	TPD
K_S (gCOD/L)	0.110	0.11–0.181	0.355	0.500	0.86	0.29	0.44–0.59
Substrate	COD	COD	BOD	BOD	BOD	COD	COD
Literature cited	(Yu and Quan, 1988)					(Babuna *et al*., 1999)	This paper

The value of K_S is converted based on HRT presented in hour

Table 8 Activation energy of substrate removal from wastewater by aerobic biological process

Wastewater	Substrate	E_a (J/mol)	Literature cited
Domestic	BOD	$4.14–5.02\times10^4$	Yu and Quan,1988
Starchy	COD	6.90×10^4	
TPD	COD	6.05×10^4	This paper

CONCLUSIONS

For TPD wastewater treated by aerobically activated sludge, the COD removal could be divided into rapid-removal stage and moderate-removal stage described with zero-order and first order reaction, respectively. The turning point was at about B/C of 0.1. COD_N ranging from 170 mg/L to 262 mg/L were the low limits for TPD wastewater treatment by aerobic activated sludge.

The kinetics values such as reaction rate constant, half saturation constant for substrate and activation energy were used for evaluating the biological treatability of TPD wastewater. By comparison with domestic and other industrial wastewater, it was evident that the substrate removal from TPD wastewater was inferior to that from domestic wastewater and much more readily than that from most of the industrial wastewater mentioned in this paper. Above all, TPD wastewater had better biological treatability than traditional textile wastewater. The expected effluent quality strongly related to COD_N and the process kinetics rather than total COD. In order to have a high quality of effluent, further treatment or other process should be combined or integrated with the aerobic process.

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