Desalination and Water Treatment
www.deswater.com

doi:10.5004/dwt.2016.1619

66 (2017) 140–148
March

Case analysis on textile wastewater subjected to combined physicochemical–biological treatment and ozonation

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Received 15 November 2015; Accepted 19 June 2016

ABSTRACT

The large amount of wastewater produced by the textile industry necessitates a cost-effective technology for enhanced wastewater treatment. In this study, a combined processing method was established to enhance discharge water quality. This process incorporated a pretreatment system, a biological contact oxidation unit, an ozone oxidation unit, and an intensive treatment system. Through this treatment approach, the ozonation of textile wastewater was examined to determine the effects of ozone dosage, ozonation time, and color/chemical oxygen demand (COD) of feed wastewater. Results revealed that the color and COD removal rates increased with increased ozone dosage. Color and COD decreased whereas NH_3–N slightly increased with the progress of ozonation. Color removal rate decreased whereas ozone dosage increased with increased feed color and COD. Feed color greatly influenced ozone dosage but not COD. Color removal rate during ozonation can be controlled to 50%–55% at a response time of approximately 2 h. The average ozone dosage was 51 g m^{-3}. After treatment by the combined process, the final discharge water was able to meet the national first-grade emission standard (GB4287-2012). The total removal rates of COD and color reached 95.2% and 95.4%, respectively. The cost of wastewater treatment amounted to only approximately 1.70 Yuan RMB m^{-3} wastewater.

Keywords: Textile wastewater; Physicochemical–biological treatment; Ozonation; Color removal

1. Introduction

The textile industry is a traditional pillar of China's national economy. Fiber processing produced 45.8 million tons of products in 2013, with the gross export amounting to over 250 billion dollars. Hence, China ranks first worldwide in terms of textile production according to the 2013/2014 China Textile Industry Development Report and is apparently a large textile power.

The textile industry involves wide ranging activities, from the preparation of raw materials to ennoblement treatment. Ennoblement is the collective term for pretreatment, dyeing, printing, and finishing of textile materials. These activities demand about 100–200 L of high-quality water per kilogram of textile product and are also highly chemically polluting [1,2]. Water is used as the principal

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medium to remove impurities, apply dyes and finishing agents, and generate steam; after serving these functions, water is then generally discharged as an aqueous effluent [3]. Effluents produced by dyeing and printing (accounting for 80% of the total amount of textile mill discharges) contain a substantial amount of chemicals that are toxic, carcinogenic, and mutagenic to various microbiological or animal species [4]. This discharge is difficult to treat economically because of its characteristics, such as extremes of pH (either alkaline or acidic, depending on the processes used) and temperature, high biological oxygen demand (BOD), high chemical oxygen demand (COD), and high concentrations of suspended solids (SS) [5]. These effluents are also highly colored because of residual dyes [6] and are thus visually unappealing and considered less for downstream reuse [7]. Therefore, the search for an effective method of treating wastewater from the textile industry is highly challenging.

Many attempts have been made to treat textile wastewater by using conventional wastewater treatment methods. These approaches mainly comprise physicochemical and biological treatments [8]. Physicochemical processes could achieve removal efficiencies sufficiently high to meet the limits imposed by locally different regulations. However, such processes are associated with a high production of chemical sludge, considerable consumption of chemicals, and high operational costs [9,10]. Furthermore, the complete decolorization of wastewater containing recently developed dyes is difficult to achieve with methods such as adsorption and chemical coagulation, especially when the dyes are highly soluble [11]. Biological processes are relatively advantageous in terms of low costs and lack of chemical sludge generated. However, this type of approach is insufficient for the decolorization and removal of surfactants and the recalcitrant COD fraction to attain acceptable limits for direct discharge [3]. Therefore, innovative treatment methods of these biorefractory effluents should be developed for effluents that do not satisfy color limits imposed in local regulations.

Ozone is potentially useful for the above purpose because ozonation can enhance the biodegradability of wastewater and achieve successful decolorization [12,13]. Ozone is a powerful oxidant for water and wastewater treatment and can be used to mineralize or partially oxidize pollutants into biodegradable intermediates [14,15]. Although the cost of ozone production has decreased in recent years, ozonation remains costly [16]. Ozone can also be used only as needed to convert biorefractory compounds into biodegradable products [17]. Thus, the combination of chemical oxidation and biological treatment is particularly interesting. This approach may be beneficial in terms of the reduction of ozone dose and treatment costs [2,12]. Nevertheless, a high synergistic effect with the biological component is required to achieve a considerable cost advantage.

The present study developed a combined processing method that comprises a pretreatment stage (adsorption and chemical coagulation), a biological contact oxidation component, an ozone oxidation process, and an intensive treatment system. The removal rates of COD and color in each step of the combined process were analyzed. The effects of ozone dosage, ozonation time, and different color/COD concentrations on ozone-oxidation efficiency were discussed. After treatment with the combined process, the final discharge water satisfied the national first-grade emission standard (GB4287-2012; COD < 80 mg L^{-1}, BOD_5 < 20 mg L^{-1}, color < 50 times, ammonia nitrogen (NH_3–N) < 10 mg L^{-1}, total phosphorus (TP) < 0.5 mg L^{-1}, total nitrogen (TN) < 15 mg L^{-1}, and SS < 50 mg L^{-1}).

2. Material and methods

2.1. Experimental setup

A schematic of the experimental system is shown in Fig. 1. The process includes the following four stages:

2.1.1. Pretreatment

Effluent generated by a textile factory was first filtered using a high-chain-type mechanical grid to dispose large suspended particles. The effluent was then subjected to temperature reduction in a cooling tower and wastewater quantity/quality adjustment in the regulating reservoir. The water was sent to a coagulation–flotation process chamber (reaction tank), in which $FeSO_4$ was used as the coagulation agent and polyacrylamide (PAM) was the flocculant. The organic polymer decolorizing agent used mainly comprised a quaternary ammonium polymer (QAP) as decolorant. Wastewater subsequently entered the primary settling tank, where COD and SS were partially removed by sedimentation.

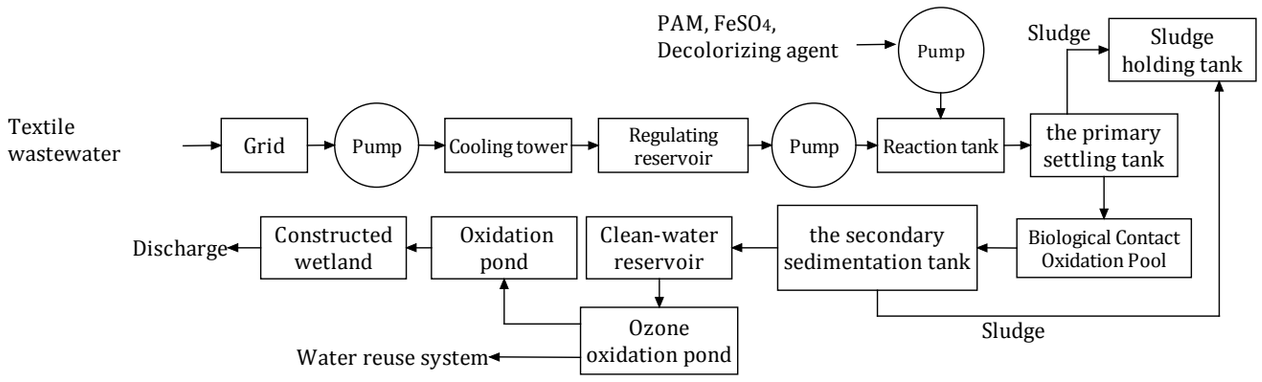

Fig. 1. Schematic of the combined process.

2.1.2. Biological contact oxidation

Wastewater from the primary settling tank was transferred to the biological contact oxidation pool for aerobic treatment. Afterward, the wastewater was sent to the secondary sedimentation tank for sedimentation to remove the SS and aging biofilms obtained from the biological contact oxidation unit.

2.1.3. Ozone oxidation

Wastewater from the secondary sedimentation tank was transferred through the clean-water reservoir to the ozone oxidation pond for further decolorization.

2.1.4. Extensive treatment

After ozonation, some wastewater was transported to the oxidation pond and constructed wetland for intensive treatment. The remaining wastewater was sent to the water-reuse system for advanced treatment.

2.2. Sewage quality

Treated sewage from one textile wastewater treatment plant in the city of Foshan contained dyestuff, auxiliaries, sizing agent, acid–base, fiber impurities, and inorganic salts. The sewage was purple black and malodorous without oil slick. This type of industrial wastewater is difficult to treat. The sewage parameters and analytical methods and instruments are listed in Table 1.

2.3. Setup and operational details of the combined system

2.3.1. Cooling tower, regulating reservoir, reaction tank, and primary settling tank

The cooling tower was mainly used to cool the sewage to below 35°C. The regulating reservoir primarily regulated the quality and quantity of wastewater for pretreatment. The pool was approximately 8.5×10^4 m^3 in size, and the hydraulic retention time was 6 h. In the reaction tank, $FeSO_4$, PAM, and organic polymer decolorizing agent were used as coagulation agent, flocculant, and decolorant, respectively. SS, including mud-and-sand and fibers, were removed from the primary settling tank to prevent their entrance into the follow-up biological contact oxidation pool. This strategy protected the latter pool from clogging and lightened the load. The hydraulic retention time was more than 3 h.

2.3.2. Biological contact oxidation pool, secondary settling tank, and clean-water reservoir

The aerobic treatment of sewage was completed in the biological contact oxidation pool. Semisoft nylon contact filling was used as biofilm carrier. The sewage circulated continuously with a recycle rate of 200%. Organics in the sewage was fully degraded by the aerobe on the biofilm. A plastic micro-porous aerator was used, and the dissolved oxygen (DO) was approximately 2 mg L^{-1}. The biological contact oxidation pool was classified into primary- and secondary-contact oxidation pools, with hydraulic retention times of 4 and 25 h, respectively. Sewage pH was controlled to within 7.6–8.9, and the removal rate of COD reached 80%. The secondary sedimentation tank was mainly used for sedimentation and the removal of aging biofilms and fine SS that were not initially removed. Hydraulic retention time in this tank was approximately 15 h. Sewage can be concentrated and allocated in the clean-water reservoir, and hydraulic retention time was 2.7 h.

2.3.3. Ozone-oxidation pond

Ozonation is a highly desirable alternative solution to the problem of color in textile effluents. This process can effectively break down the conjugated double bonds of dye chromophores and other functional groups, such as complex aromatic rings [18]. Thus, ozonation was used to remove residual pollutants after the biological treatment. In the present paper, ozone gas (with a concentration of 100–120 g m^{-3} and flow rate of 80–90 m^3 h^{-1}) was produced in a low-voltage and high-frequency discharge tube. Ozone production proceeded as follows: (i) compressed air (about 0.4 MPa) was produced with an air compressor; (ii) air was separated with an oxygen generator, producing high-concentration oxygen (90% of weight percentage); (iii) a gas mixture of O_3, O_2, and N_2 was generated in the low-voltage and high-frequency discharge tube; and (iv) the O_3, O_2, and N_2 mixture was intro-

Table 1
Pollutant contents of influents from a textile wastewater treatment plant and corresponding discharge standards

Parameter (Unit)	Raw wastewater value	Discharge standard	Analytical methods and instruments
pH (–)	8–11	6–9	Glass-electrode method; Orion 420A+pH meter
COD (mg L^{-1})	400–1,000	80	Fast digestion spectrophotometric method; DRB200 digester/ DR890 chromometer
BOD_5 (mg L^{-1})	200–450	20	Dilution and inoculation method; YSI5100 dissolved oxygen meter
Color (times)	400–1,100	50	Dilution times method;
NH_3–N (mg L^{-1})	3–25	10	Nessler's reagents spectrophotometer; 722 spectrophotometer
TP (mg L^{-1})	1–10	0.5	Ammonium molybdate spectrophotometry; 722 spectrophotometer
TN (mg L^{-1})	25–90	15	Alkaline potassium persulfate digestion UV spectrophotometry; UV-1800 ultraviolet spectrophotometer
SS (mg L^{-1})	50–250	50	Gravimetric method; electronic scales

duced to water with a venture tube mixer. After the ozone reaction, about 30,000 t d^{-1} sewage was sent to the intermediate-water-reuse system, and the rest was sent to the oxidation pond and the constructed wetland.

2.3.4. Oxidation pond and constructed wetland

Biofilm colonization and aeration systems were installed in the oxidation pond for reoxidation settling. Semisoft nylon contact filling was also used as a biofilm carrier, through which organics can be fully degraded by the contained aerobes. Oxygen was transferred with a galvanized steel pipe and aerated through a polyvinyl chloride perforated aerator pipe. The hydraulic retention time was approximately 12 h, and the DO was approximately 7–9 mg L^{-1} (some DO originated from ozone decomposition). The constructed wetland ecosystem exhibited the synergistic effect of physical, chemical, and biochemical reactions. The wetland was expected to achieve efficient wastewater purification through filtration, adsorption, sediment, ion exchange, plant uptake, and microbial decomposition. Some of the plants in the wetland included water hyacinths, water lettuces, canna indica, and cyperus alternifolius. Hydraulic retention time was approximately 4 h.

2.4. Analytical methods

The percentage removal of the parameters evaluated was determined using the following equation:

$$\%\ \text{removal} = \left(\frac{C_0 - C}{C_0}\right) \times 100 \quad (1)$$

where C_0 and C are the inlet and outlet colors, inlet and outlet concentration of COD and NH_3–N.

Table 2
Effluent characteristics after pretreatment

Parameters	Value	Average value
pH	8.8–10.2	9.7
COD (mg L^{-1})	210–500	405
BOD (mg L^{-1})	80–220	158
Color (times)	180–260	230
SS (mg L^{-1})	20–130	48
NH_3–N (mg L^{-1})	11.0–26.1	20
TP (mg L^{-1})	1.0–4.3	2.1
TN (mg L^{-1})	29–38	34

3. Results and discussion

3.1. Pretreatment

Effluent quality varies with time and may include many types of dyes, detergents, sulfide compounds, solvents, heavy metals, and inorganic salts. Their amounts also depend on the effluent-generating process [19]. Pollutants in effluents inhibit microbial growth, so suitable ambient conditions are required to implement subsequent biological contact oxidation processes. Regarding a biological contact oxidation reactor's performance, granulation is a highly important component that determines the wastewater-treatment effect. In the present pretreatment process with grid block, temperature reduction, coagulation, flocculation, and discoloration, COD and color decreased to levels that fitted the process of biological contact oxidation (Table 2). The desired results for COD and color removal in the pretreatment process using $FeSO_4$, PAM, and organic polymer decolorizing agent were achieved (Fig. 2). The removal rate of COD was 26%–63% (average = 40%), and that of color was 53%–74% (average = 65%). Overdosage is likely to cause secondary pollution, so the consumptions of $FeSO_4$, PAM, and organic polymer decolorizing agent in this pretreatment were controlled to 1,070–1,120, 6–7, and 15.0–16.5 g m^{-3}, respectively. The treatment results are shown in Fig. 3.

3.2. Biological contact oxidation

Biological contact oxidation is a new type of wastewater treatment method resistant to the effects of high load, high microbial concentration, and lack of sludge backflow. This technology is widely used in wastewater treatment [20]. Hence, we chose biological contact oxidation as an important step to treat COD concentration in textile wastewater. Biological contact oxidation was effective in removing COD at an average rate of 80% (Fig. 4(a)).

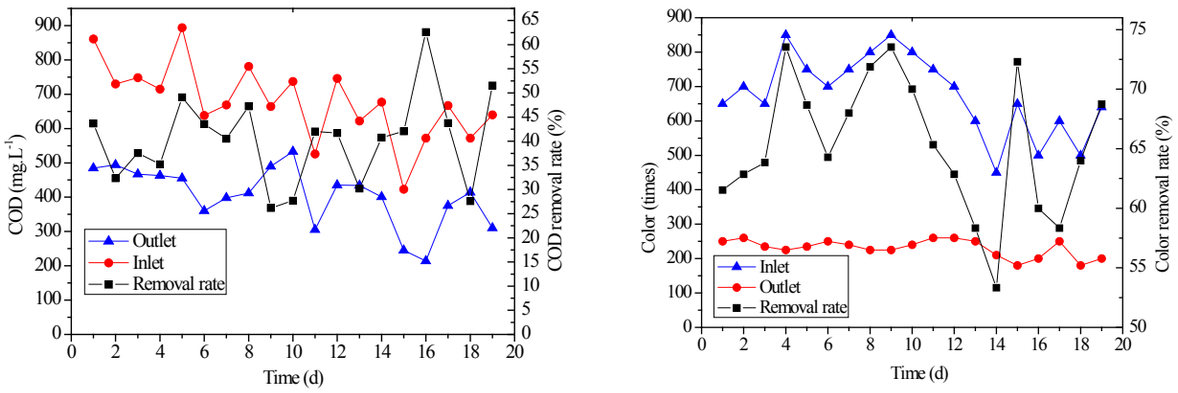

Fig. 2. (a) COD and (b) color removal in pretreatment.

After this treatment, COD levels in the sewage averaged at 85 mg L^{-1}, which approached that of the discharge standard (COD < 80 mg L^{-1}). Compared with COD removal, color removal in this treatment was only 47% (average = 120 times; Fig. 4(b)), which far exceeded the limit of the discharge standard (color < 50 times). Apparently, auxiliary processes were necessary to further treat the sewage and reach the standard.

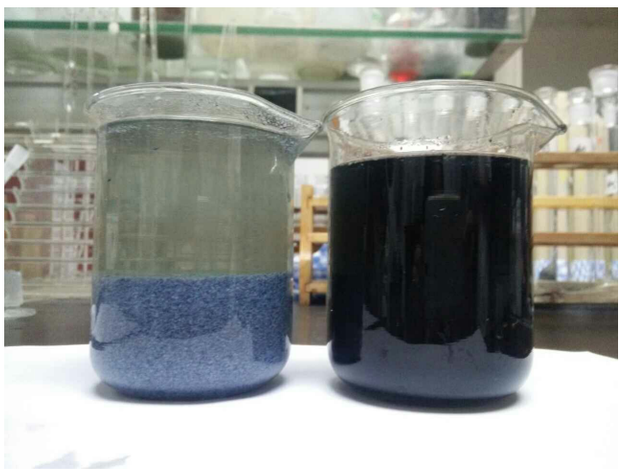

Fig. 3. Control of textile wastewater with and without the addition of $FeSO_4$, PAM, and organic polymer decolorizing agent.

3.3. Ozone oxidation

3.3.1. Effect of applied ozone dose on color and COD removal

Fig. 5(a) shows that the removal rate of color constantly increased with increased ozone dosage. With feed colors of 110 and 120 times, removal rate increased from 33% and 21% to 79% and 78%, respectively, with increased dosage from 13 to 125 g m^{-3}. With increased ozone dosage, the removal rate of COD also increased (Fig. 5(b)). However, the removal rate of COD was much lower than that of color. With increase ozone dosage from 13 to 125 g m^{-3}, the removal rate of COD increased from 10% to 36%. The lower removal rate of COD than that of color can be explained by the incomplete oxidation of organic materials.

3.3.2. Effect of ozonation time on the removal of color, COD, and NH_3–N

Fig. 6 shows that color decreased with prolonged ozonation time. At an ozone dosage of 60 g m^{-3}, the colors of 100, 120, and 140 times dropped to 38, 47, and 58 times, respectively, at 140 min. Ozonation time was prolonged

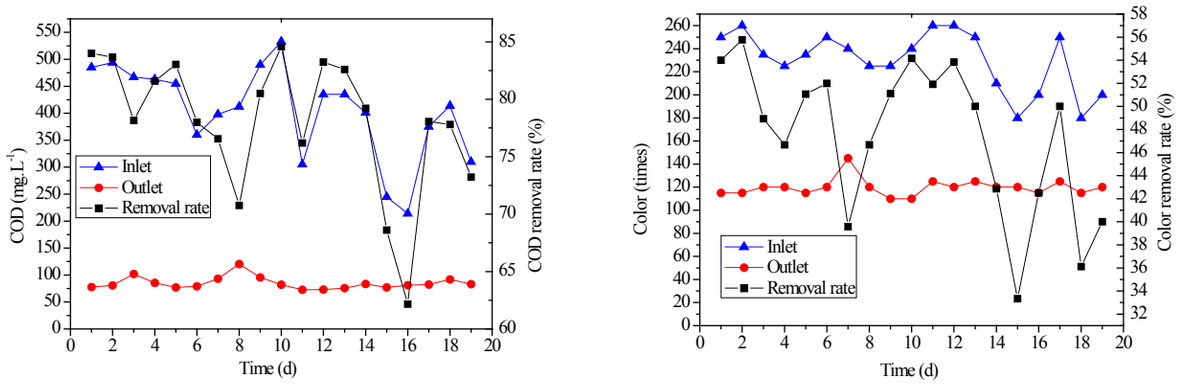

Fig. 4. (a) COD and (b) color removal in biological contact oxidation.

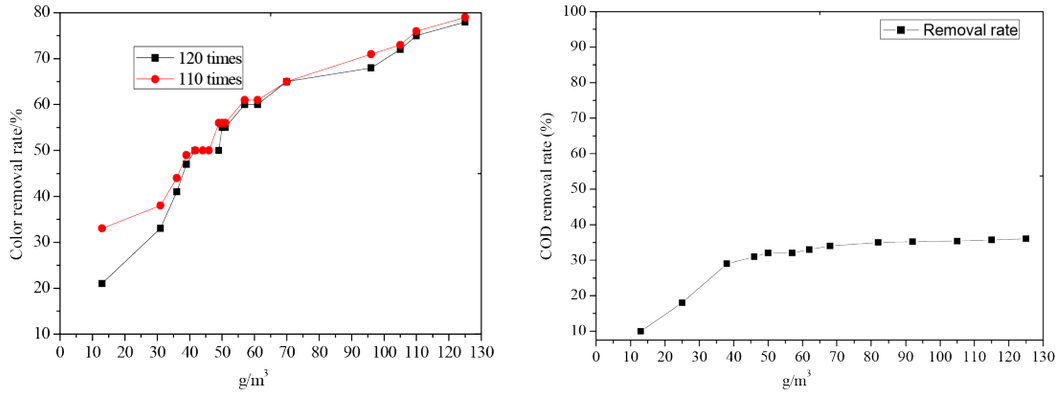

Fig. 5. Effect of ozone dosage on the removal rate of (a) color and (b) COD.

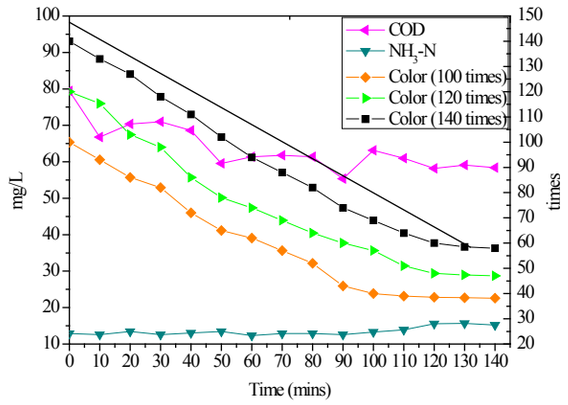

Fig. 6. Effect of ozonation time on color, COD, and NH_3–N removal.

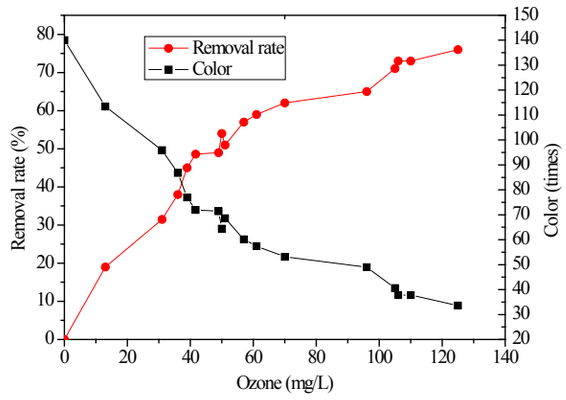

Fig. 8. Effect of ozone dosage on color removal.

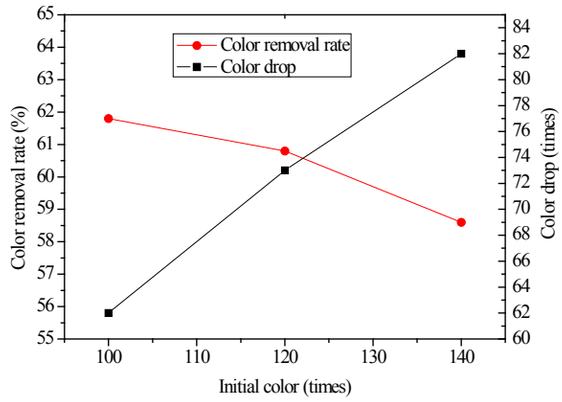

Fig. 7. Effect of feed color on color drop and its removal rate.

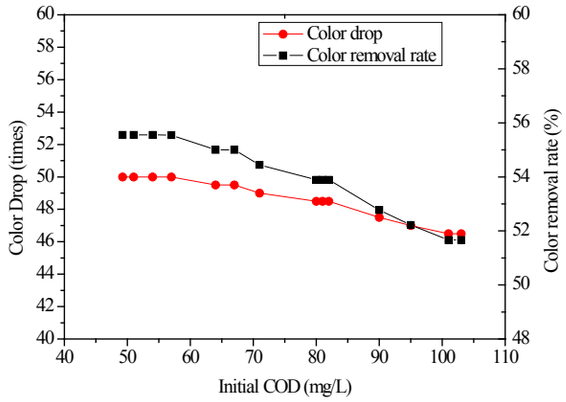

Fig. 9. Effect of feed COD on color drop and its removal rate.

with increased feed color. When the feed color was 100, 120, and 140 times, the curve began to change slowly after 100, 120, and 130 min, respectively. COD continued to fluctuate with prolonged ozonation time but finally decreased from 79.4 to 54.7 mg L^{-1}, whereas NH_3–N slightly increased from 12.9 to 13.8 mg L^{-1}. The increase in COD may be due to the dye molecules being oxidized by ozonation. The results were small-molecule organic fragments such as acetic acid, aldehydes, and ketones, which are not completely mineralized under the oxidative conditions described, thereby contributing to increased COD with prolonged ozonation time [21]. Wang et al. [22] reported that these small molecules significantly contribute to COD and cannot be completely removed by ozonation. However, components that contribute to COD can be easily biodegraded in the subsequent oxidation pond. The slight increase in NH_3–N may result from the production of NH_3 through the inadequate ozonzation of nitrogenous organic compounds. Fig. 4(b) reveals that the average color of sewage after biocontact oxidation was 120 times. Thus, ozonation time was more suitable to control at approximately 2 h.

3.3.3. Effect of feed color and COD on color removal

Color drop increased and color removal rate decreased with increased feed color (Fig. 7). At 60 g m^{-3} ozone dosage and 80 mg L^{-1} COD, color drop increased from 62 to 82 times, and color-removal rate decreased from 61.8% to 58.6% with increased feed color from 100 to 140 times. Furthermore, 100 times of sewage color can be reduced to 38 times with only 60 g m^{-3} ozone dosage. However, 106 g m^{-3} ozone dosage was needed to reduce 140 times of sewage color to 38 times (Fig. 8). Hence, feed color exerted a great influence on ozone dosage [5,21], and pretreatment and biological treatment were necessary before ozone oxidation.

Color drop and color removal rate decreased with increased feed COD, and ozone dosage increased with increased feed COD. However, COD content in feed water showed no apparent influence on color drop, color removal rate, and ozone dosage. At 50 g m^{-3} ozone dosage and 90 times color, color drop decreased from 50 to 46.5 times, and color removal rate decreased from 55.6% to 51.7% with increased COD from 49 to 103 mg L^{-1} (Fig. 9). When COD was 103 mg L^{-1} and color was 90 times, 53.8 g m^{-3} ozone dosage decreased the color to 40 times. Compared with sewage containing 49 mg L^{-1} COD, only 3.8 g m^{-3} extra ozone dosage was needed.

3.3.4. Color and COD removal in ozone oxidation

After biocontact oxidation, color became 115–145 times, with a mean of approximately 120 times (Fig. 4(b)). The aver-

age removal rate of color in the oxidation pond and constructed wetland was approximately 33.8% (Fig. 11(b)). Hence, color in the effluent would reach the discharge standard (<50 times) if the removal rate of color in ozonation can be controlled to 50%–55%. The color and COD removal rates during ozonation are presented in Fig. 10. After ozonation, color remained at 45–65 times (mean ≈ 57 times), with an average removal rate of 52.65. By contrast, COD was 42–73 mg L^{-1} (mean = 61 mg L^{-1}), and its average removal rate was 27%. Ozone consumption was ~51 g m^{-3} on average, which was much lower than that of a previous report (ozone dose = 135 g m^{-3}) [3].

3.4. Intensive treatment: oxidation pond and constructed wetland

Fig. 11 shows that COD and color can be effectively removed using intensive treatments. COD in the effluent decreased to 23–45 mg L^{-1}, with an average removal rate of ~44.1%. Meanwhile, color dropped to 20–45 times, with an average removal rate of ~33.8%.

3.5. Review of the combined process

The combined process showed good performance in wastewater treatment (Figs. 12–14). After treatment by the combined process, the discharge water attained 80 mg L^{-1} COD, 20 mg L^{-1} BOD, 50 times color, 10 mg L^{-1} NH_3–N, 0.5 mg L^{-1} TP, 15 mg L^{-1} TN, and 50 mg L^{-1} SS. The total removal rates of COD and color reached 95.2% and 95.4%, respectively (Table 3). Biological oxidation showed the highest removal rate of COD, whereas pretreatment showed the highest removal rate of color among the four treatment units. Cost was considered in the ozonation process. Hence, ozone dosage decreased, and color removal rate was lower than that in the pretreatment.

3.6. Total cost

This project was based on original physicochemical–biological treatment engineering, and ozonation was used to replace the anaerobic process for color removal. Thus, the costs of new equipment and structures must be discussed.

3.6.1. Investment on new engineering

Five ozonators and two oxygen generators costed 9.5 million RMB, and the construction of one ozone contact oxidation pond costed 1.7 million RMB. The investment totaled 11.2 million RMB.

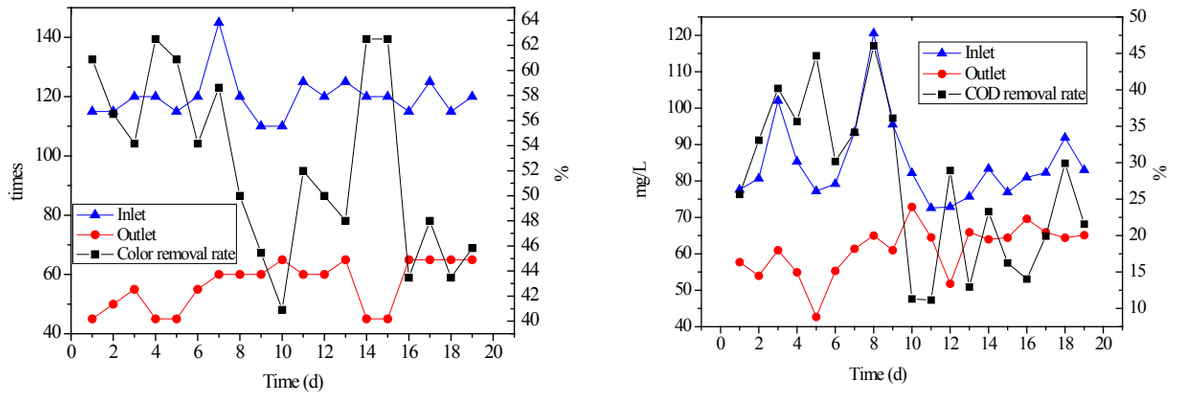

Fig. 10. (a) Color and (b) COD removal in ozone oxidation.

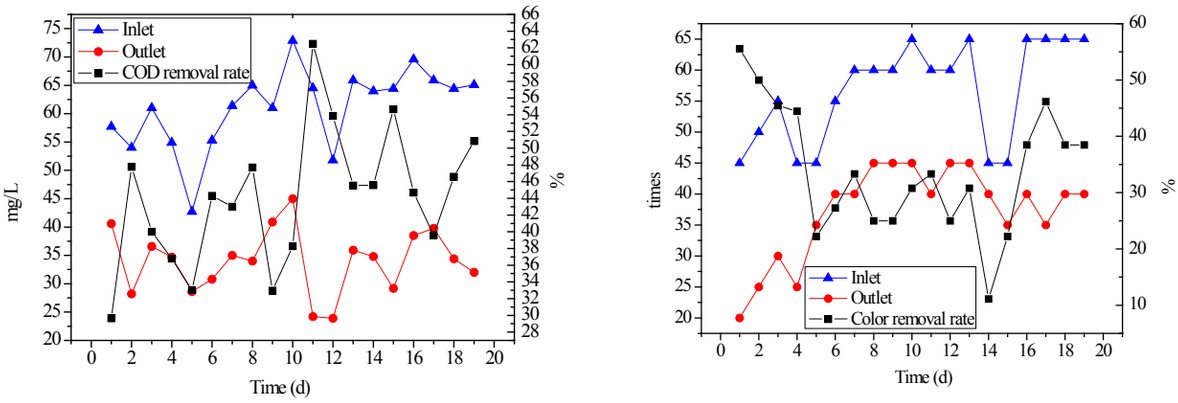

Fig. 11. (a) COD and (b) color removal in intensive treatment.

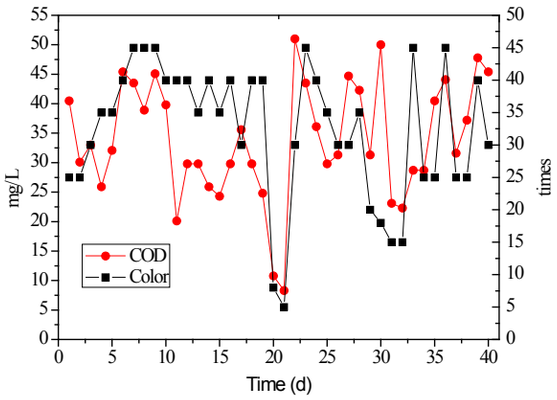

Fig. 12. COD and color concentration in discharge wastewater.

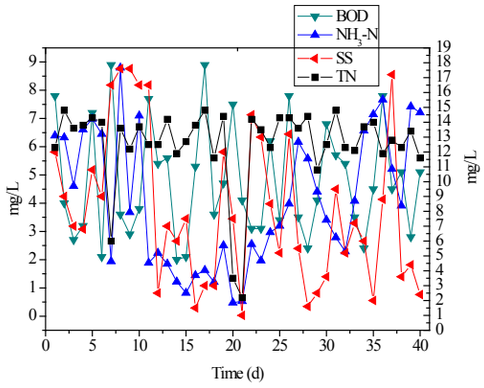

Fig. 13. BOD, NH_3–N, SS, and TN concentrations in discharge wastewater.

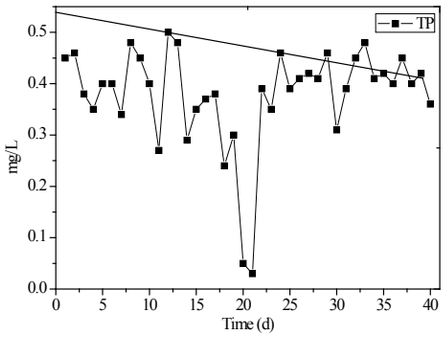

Fig. 14. TP concentration in discharge wastewater.

3.6.2. Operating cost

During ozonation, ~51 g O_3 m^{-3} wastewater and 12 kW·h kg O_3 power were consumed on average. Thus, an additional operating cost of approximately 0.37 RMB m^{-3} emerged for a power price of 0.6 RMB kW·h^{-1}. The total operating cost of treating textile wastewater in 2014 is displayed in Table 4 (water and power bills, chemical cost, sewage discharge fees, sludge discharge fees, etc.), reaching 1.70 RMB m^{-3}.

Table 3
Summary of results of the combined process (indicated by removal rate [%])

Treatment	COD	Color
Pretreatment	40	65
Biological contact oxidation	80	47
Ozone oxidation	27	52.6
Intensive treatment	44.1	33.8
Total	95.2	95.4

Table 4
Cost of the combined process

Unit	Data
Sewage production (10^4 m a^{-1})	1,521.33
Electric rate (Yuan kW·h^{-1})	0.6
Power (kW·h a^{-1})	25,766,235
Electricity cost (Yuan a^{-1})	15,459,741
$FeSO_4$ consumption (t a^{-1})	16,849
PAM consumption (t a^{-1})	93
QAP consumption (t a^{-1})	249
Chemical cost (Yuan a^{-1})	4,617,952
Recycled water (t a^{-1})	82,525
Fresh water (t a^{-1})	332
Water cost (Yuan a^{-1})	180,628
Sludge treatment cost (Yuan a^{-1})	5,054,400
Emission charges (Yuan a^{-1})	605,851
Machine maintenance cost (Yuan a^{-1})	15,965
Unit sewage treatment cost (Yuan m^{-3})	1.70

4. Conclusions

The combined processing method was successfully used to treat textile wastewater. The method comprised a pretreatment system, a biological contact oxidation unit, an ozone oxidation unit, an oxidation pond, and a constructed wetland. Through this treatment approach, the effects of ozone dosage, ozonation time, feed color, and COD on ozone oxidation efficiency were discussed. Furthermore, color and COD removal in the combined process were analyzed. The following conclusions were drawn from the experimental results.

With increased ozone dosage, color and COD removal increased. However, high color removal and low COD removal rate were obtained by ozonation. The lower removal rate of COD compared with that of color can be explained by the incomplete oxidation of organic materials. Color and COD generally decreased with prolonged ozonation time, although COD occasionally increased during ozonation. However, NH_3–N slightly increased because of the incomplete ozonation of nitrogenous organic compounds, which then produced NH_3. With increased feed color and COD, removal rate of color decreased, whereas ozone dosage increased. Feed color greatly influenced ozone dosage, but the influence of COD was not evident.

Color removal rate in the ozonation process could be controlled to within 50%–55% at a response time of approximately 2 h through the combined effects of ozone dosage and ozonation time on the removal rates of color and COD, as well as on such removal rates in each step of the combined process. The average ozone dosage was 51 g m^{-3}.

The desired effect was achieved in the treatment of textile wastewater by using this combined process. After treatment, the discharge water attained 80 mg L^{-1} COD, 20 mg L^{-1} BOD, 50 times color, 10 mg L^{-1} NH_3–N, 0.5 mg L^{-1} TP, 15 mg L^{-1} TN, and 50 mg L^{-1} SS. The total removal rates of COD and color reached 95.2% and 95.4%, respectively. The economic investment entailed for this combined processing method was low (~1.70 Yuan RMB m^{-3} wastewater).

Acknowledgment

The authors would like to acknowledge the financial support of the Major Science and Technology Projects of Guangdong Province "Foshan City green low-carbon urban construction technology integration and demonstration" (No. 2012A010800003).

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