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Decolorization effects of six azo dyes by O_3, UV/O_3 and UV/H_2O_2 processes

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Abstract

The decolorization of six azo dyes by means of O_3, UV/O_3 and UV/H_2O_2 processes was performed successfully with all of the tested dyes decolorizing effectively. However, dyes with dis-azo links (i.e. C.I. Direct Yellow 4 and C.I. Acid Black 1) were more difficult to decolorize than mono-azo dyes. During the processes, the pH value changed from 5.4 to 3.5, thus becoming more acidic. The increased acidity resulted from the oxidation and breakdown of the $R^+-(SO_3^-)$ bond in the azo dye structure. The removal of 95% color was the level chosen to compare the energy efficiency. The results showed that UV/H_2O_2 used the most energy, 5–11 times, than that of UV/O_3 process, and 265–520 times more than that of the ozonation process.

Keywords: Ultraviolet; Advanced oxidation processes (AOPs); Azo dye; Decolorization; Ozone; Hydrogen peroxide

1. Introduction

Azo dyes are used extensively in textile dyeing operations and contribute to problems associated with disposal of the wastewater generated from such operations. The textile industry is one of the most important industries in Taiwan's economic development in the last few decades. About 10,000 dyes are used in the textile industry, and the quality and quantity of wastewater from this industry are of concern by the public and government, especially, with the aesthetic character of color discharges into rivers and other surface water bodies. The Taiwan Environmental Protection Agency (EPA) issued strict effluent standards to enforce textile industries to decolorize and eliminate pollutants from wastewater before discharging. Therefore, the technologies of dye wastewater decolorization are of increasing importance.

The industrial manufacturing of azo dyes and textile finishing processes generate wastewater streams contaminated with azo dyes. Some of the azo dyes are difficult to treat by conventional wastewater treatment methods. In a study of USEPA, 11 out of 18 azo dyes passed through the activated sludge process substantially untreated, while the other 4 dyes were significantly adsorbed onto the waste activated sludge, and only 3 dyes were apparently biodegraded [1]. Some studies have shown that azo dyes are very difficult to be biodegraded under aerobic conditions [2,3]. Usually, dye wastewater can be decolorized through conventional treatment such as coagulation with hydrolyzing metals followed by flocculation and filtration (or sedimentation plus filtration). But this conventional process produces a large amount of sludge which needs further disposal. On the other hand, adsorption onto activated carbon is also an effective method for color removal. However, activated carbon may have a very short life, depending on the initial color level and the nature of dyestuff.

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Regeneration of activated carbon is inefficient and very expensive. The other treatment method such as membrane filtration is also effective but very expensive [4]. The operating cost of disinfection of secondary effluent (99.9% faecal coliform reduction) by membrane filtration was much higher than ozone, UV or chlorine.

Advanced oxidation processes (AOPs), such as O_3, UV/O_3 and UV/H_2O_2 are widely used to decompose organic products in industrial wastewater and groundwater. The extensive literature in this field has been reviewed by Venkatadri and Peters [5]. These processes have the potential ability to mineralize most of the organic contaminants into carbon dioxide and water. In AOP systems, the free radicals ($OH^{\cdot}$) are the dominant species contributing to the degradation of organics in wastewater. These processes comprise the activation of hydrogen peroxide, or ozone, with UV light to produce hydroxyl radicals which have a higher oxidation potential (2.8 V) than that of hydrogen peroxide (1.78 V). During the last decade, some investigations have been reported about the successful application of the UV/H_2O_2 process for dye wastewater treatment. Shu et al. [6] demonstrated that the UV/H_2O_2 process was able to decolorize C.I. Acid Red 1 and C.I. Acid Yellow 23. Galindo and Kalt [7] expressed that C.I. Acid Orange 7 could be decolorized in UV/H_2O_2 process with an optimum molar ratio of H_2O_2/dye about 1754. The treatment of various dyes in a UV/H_2O_2 process has also been reported by Ince, Neamtu et al., Cisneros et al., and Mohey El-Dein et al. [8–11]. Ozonation and ultraviolet induced ozonation were studied by Hafez et al. [12] and Shu and Huang [13] for the decomposition of *m*-toluidine azo dye and C.I. Acid Orange 10, respectively. Based on the above literature, advanced oxidation processes are shown to be a feasible treatment for decolorization of azo dyes.

The objective of this study was to determine the feasibility of using O_3, UV/O_3 and UV/H_2O_2 processes as alternative treatment technologies for wastewater of azo dyes in a 100 l (dm^3) pilot scale reactor. The operating parameters such as hydrogen peroxide dosage and pH were evaluated to find the optimal condition for the UV/H_2O_2 process. The 95% color removal was chosen to evaluate the retention time demand and electric power consumption by all of the O_3, UV/O_3 and UV/H_2O_2 reactors. The decolorization rate constant of each azo dye follows the pseudo-first order reaction with respect to the dye concentration.

2. Experimental

2.1. Material

Hydrogen peroxide was obtained from Fluka Chemical (30% w/w). The azo dyes, which included C.I. Acid Orange 10, C.I. Acid Red 14, C.I. Acid Red 18, C.I. Acid Yellow 17, C.I. Direct Yellow 4, and C.I. Acid Black 1, were obtained from Aldrich Chemical Co. and used without further purification. The characteristics of azo dyes are displayed in Table 1. The chemical structures of six azo dyes are shown in Fig. 1. The HPLC grade solvents, methanol and acetic acid were purchased from Fisher Scientific Co. Reagents for the measurement of hydrogen peroxide concentrations were all ACS analytical reagent grade from Fisher Scientific Co. All stock solutions were refrigerated and stored in the dark.

2.2. Apparatus

The azo dye solution was placed in a pilot scale stainless steel reactor, which was 33 cm diameter, 150 cm height, with 100 l (dm^3) holdup volume. At the reactor center, a Canrad Hanovia low pressure mercury lamp, with 25 inches arc length and 5000 W output, was located. The lamp was protected by two wells: one was nitrogen gas to avoid any explosive hazard and the other was for cooling water circulation. The solution was completely mixed using a recycling pump and nitrogen bubbling from the reactor bottom. The pH was monitored. The emitted gas was passed through an activated carbon adsorption column to prevent volatile organic emission in the laboratory. For ozonation and UV/O_3 reaction, ozone was introduced into the bottom of the reactor through a four-head sparger of medium porosity. The exhaust gas was vented from the top of the reactor passing through a pair of absorption bottles and vented into a laboratory hood. Ozone was generated from oxygen by Welsbach T-816 ozone generator. Ozone output flow rate was adjusted by a ball valve at 6.0 $dm^3\ min^{-1}$, and a 99.6% pure dry oxygen stream was

Table 1
The characteristics of six azo dyes studied in this work

Azo dyes	Formula	λ_{max} (nm)	MW	Dye contents (%)
C.I. Direct Yellow 4	$C_{26}H_{18}N_4Na_2O_8S_2$	497	624.56	70
C.I. Acid Black 1	$C_{22}H_{14}N_6Na_2O_9S_2$	618	616.50	85
C.I. Acid Orange 10	$C_{16}H_{10}N_2Na_2O_7S_2$	452	452.38	95
C.I. Acid Red 14	$C_{20}H_{12}N_2Na_2O_7S_2$	515	502.44	50
C.I. Acid Red 18	$C_{20}H_{11}N_2Na_3O_{10}S_3$	506	604.48	75
C.I. Acid Yellow 17	$C_{20}H_{10}Cl_2N_4Na_2O_7S_2$	400	551.30	60

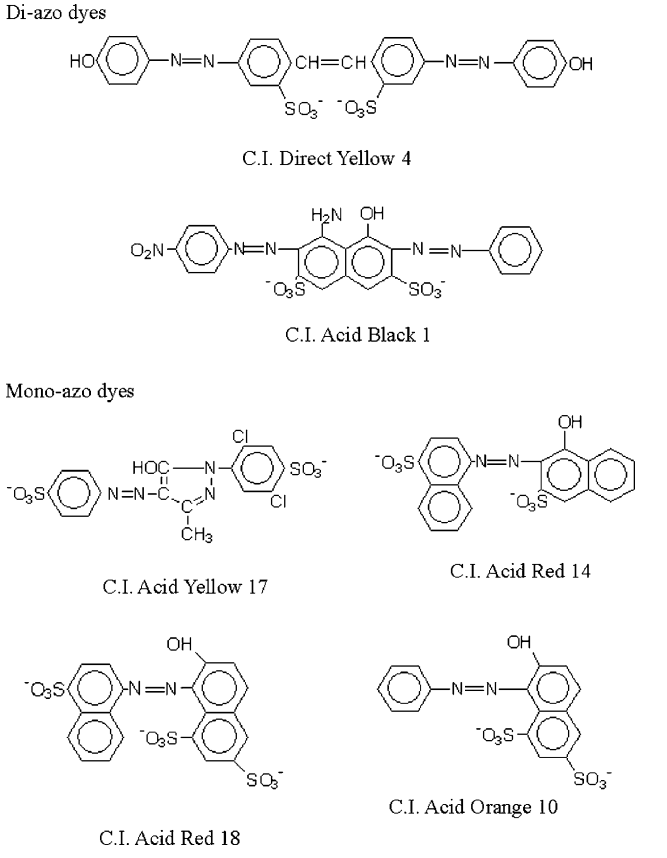

Fig. 1. The molecular structures of six azo dyes studied in this work.

used to produce ozone. The ozone generator was set at 110 W and 105 V for maximum capacity.

2.3. Analyses

Optical absorption spectra on azo dyes were determined by a Cray DMS-300 spectrophotometer. The wavelengths of maximum adsorption for each azo dye were selected for quantitative analysis. The C.I. Acid Orange 10 (AO10) was also analyzed by a LDC/Millton-made HPLC with a 3 μm particle size, 6 mm × 75 mm C_{18} column, based on the proposed method of Bruins et al. [14]. The mobile phase was 50% methanol and 50% water with 1 M acetic acid. Hydrogen peroxide concentration was determined by spectrophotometer method based on the method proposed by Masschelein et al. [15]. By adding 1.0 ml Co(II) reagent and 1.0 ml hexametaphosphate solution to 80 ml sample and making up to 100 ml with saturated bicarbonate solution, a green color is developed for the measurement of the absorbance at 260 nm. The absorbance is then compared with the calibration curve to obtain the hydrogen peroxide concentration. Ozone concentration was determined by iodometric method based on the method proposed by Shechter [16]. By adding 5 ml of sample into a sample vial with 5 ml of 2% neutral potassium solution, a yellow color was developed. After 30 min, measurement of the absorbance at 352 nm was compared with the calibration curve to obtain the aqueous ozone concentration.

2.4. Procedure

In our pilot scale study, 100 dm^3 of aqueous solution, prepared in given dye concentration and hydrogen peroxide dosage, was reacted in the photo-oxidation reactor under UV radiation. Samples were withdrawn at regular time intervals for analysis. For ozonation, 100 dm^3 of aqueous solution, prepared in various dye concentrations and ozone dosage was reacted in the reactor with or without UV radiation.

3. Results and discussion

3.1. Determining optimum operating conditions of UV/H_2O_2 reactor

In our preliminary study, the decoloration of C.I. Acid Orange 10 (AO10) by ultraviolet radiation alone, or by hydrogen peroxide oxidation alone, showed that neither could degrade azo dye significantly. Combining UV radiation and hydrogen peroxide could degrade azo dye in a relatively high reaction rate. The decolorization of AO10 was studied under various conditions such as initial hydrogen peroxide concentration, pH and the experimental results were given in Fig. 2. The experiments were performed at volumetric UV intensity 50 W dm^{-3} (5000 W for 100 dm^3), initial hydrogen peroxide concentration of 7.08 mg-mol dm^{-3} and AO10 concentration of 20.0 mg/l (20.0 mg dm^3, 42.0 μg-mol dm^{-3}) in a 100 dm^3 pilot scale photochemical reactor. In Fig. 2, concentrations of AO10 and H_2O_2 versus time were recorded during the experiment while monitoring the pH change.

The rate of decolorization of azo dyes can be expressed as:

$$\frac{dC_A}{dt} = -kC_A C_{OH^\cdot} \quad (1)$$

where C_A represents concentration of AO10; $C_{OH^\cdot}$ denotes the concentration of hydroxyl radical; t is the reaction time, assuming that the hydroxyl radical concentration reaches equilibrium instantaneously in the presence of excess hydrogen peroxide. Thus, $C_{OH^\cdot}$ can be treated as a constant. The rate expression equation is then simplified into a pseudo-first order kinetic model as follows:

$$\frac{dC_A}{dt} = -k_{obs}C_A \quad (2)$$

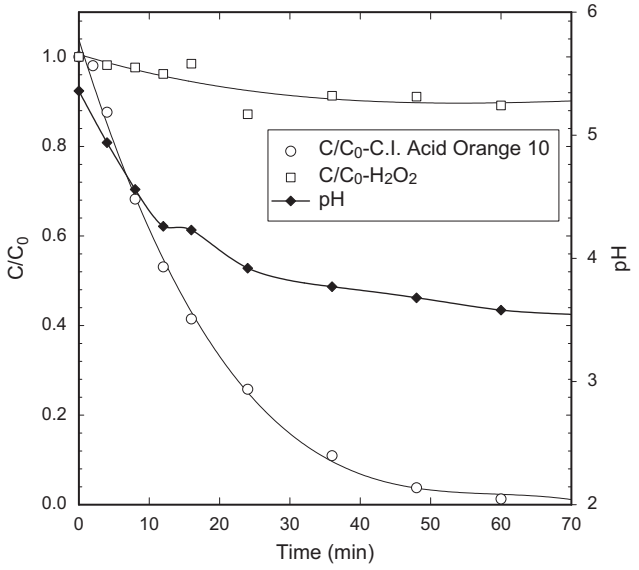

Fig. 2. The normalized concentrations (C_A/C_{A0}) of C.I. Acid Orange 10 and hydrogen peroxide as functions of time in UV/H_2O_2 process. Initial dye concentration was 42.0 μg-mol dm^{-3} (20.0 mg dm^{-3}) and hydrogen peroxide concentration was 7.08 mg-mol dm^{-3} at pH = 5.3.

By integrating both sides, the equation changes as follows:

$$-\ln\left(\frac{C_A}{C_{A0}}\right) = k_{obs}t \tag{3}$$

where k_{obs} is the rate constant of the pseudo-first order kinetic model; C_{A0} expresses the initial concentration of AO10. Based on the above equations, Fig. 3 shows the changes in normalized AO10 of the logarithmic concentration (C_A/C_{A0}) with time with hydrogen peroxide concentration from 2.36 to 9.44 mg-mol dm^{-3}. Observed pseudo-first order rate constants of the UV/H_2O_2 reaction are calculated by linear regression, 0.0251 and 0.0761 min^{-1} for H_2O_2 concentrations of 2.36 and 9.44 mg-mol dm^{-3} min^{-1}, respectively. The square of the relative correlative coefficients (R^2) of experimental results are high from 0.990 to 0.998. Also, the straight lines indicate that the assumption of a pseudo-first order reaction is reasonable as shown in Fig. 3. Fig. 4 shows the changes in normalized AO10 of logarithmic concentration (C_A/C_{A0}) with time while pH changes from 2.4 to 9.7. The observed pseudo-first order rate constants of the UV/H_2O_2 reaction are calculated by linear regression, 0.0644 and 0.0273 min^{-1} for pH 2.4 and 9.7, respectively. The R^2 values are high from 0.989 to 0.998. It was observed that the degradation of AO10 followed the pseudo-first order reaction, so that the pseudo-first order rate constant can be obtained by regression analysis with a reaction time of 60 min, the hydrogen peroxide concentration decreased to about 10% of the original value. On the other hand, the pH change from 5.4 to 3.5 was attributed to the oxidation

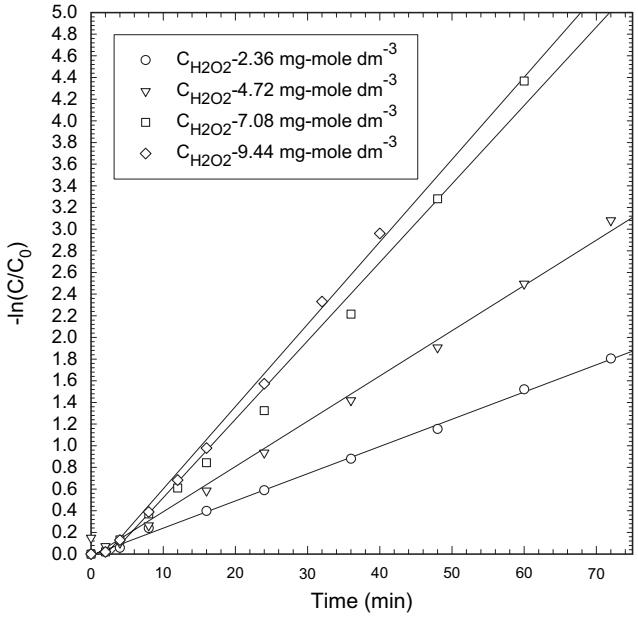

Fig. 3. The first order rate representation of C.I. Acid Orange 10 versus time under various hydrogen peroxide concentration from 2.26 to 9.44 mg-mol dm^{-3} at pH = 5.3 in UV/H_2O_2 process. Initial dye concentration was 42.0 μg-mol dm^{-3}.

and the breakdown of $R^+-(SO_3^-)$ bond in dye structure.

In order to determine the optimum operating conditions of H_2O_2 concentration and pH for AO10 decolorization in the UV/H_2O_2 reactor, the rate constants were plotted with these two parameters as

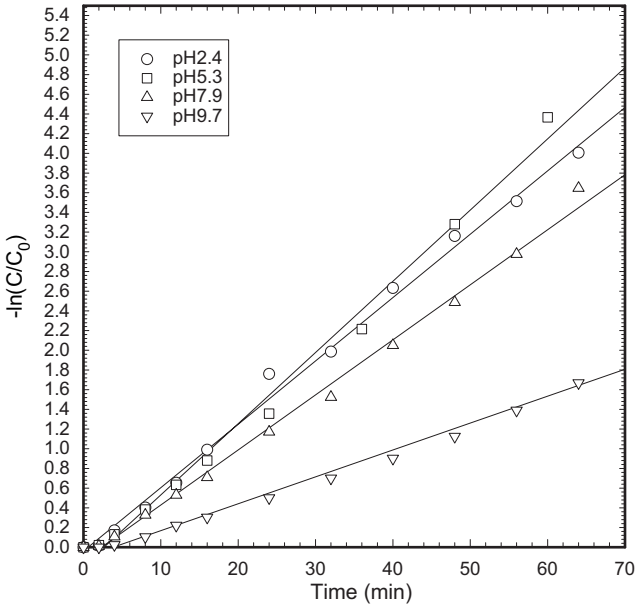

Fig. 4. The first order rate representation of C.I. Acid Orange 10 versus time under various pH from 2.4 to 9.7 in UV/H_2O_2 process. Initial dye concentration was 42.0 μg-mol dm^{-3} and hydrogen peroxide concentration was 7.08 mg-mol dm^{-3}.

shown in Fig. 5. It indicates the optimum hydrogen peroxide dosage while the rate decolorization constants increase linearly with increasing H_2O_2 concentration up to 7.08 mg-mol dm^{-3}. Thus, an H_2O_2 concentration of 7.08 mg-mol dm^{-3} was selected for decolorization of six target azo dyes for this UV/H_2O_2 reactor.

The pH is also one of the major factors which affects treatment efficiency. The dye solution original pH is 5.3, adjusted by adding 1 N NaOH to give pH 9.7 and 7.9, or by adding 1 N HCl to give pH 2.4. Meanwhile, the AO10 concentration was 42.0 μg-mol dm^{-3} and initial hydrogen peroxide concentration was 7.08 mg-mol dm^{-3} in the UV/H_2O_2 reactor as shown in Fig. 5. It was observed that the decolorization rate showed a peak efficiency at pH 5.3 and so this pH was selected for the decolorization condition of six target azo dyes in the UV/H_2O_2 reactor. Ultimately, from the above data, the optimal conditions obtained for UV/H_2O_2 reactor were initial concentration of H_2O_2 of 7.08 mg-mol dm^{-3}, pH at 5.3 with initial AO10 concentration of 42.0 μg-mol dm^{-3}.

3.2. Decolorization of azo dyes in UV/H_2O_2 process

A 100 dm^3 reactor with six azo dyes (20.0 mg dm^{-3}) including two dis-azo dyes and four mono-azo dyes were tested in UV/H_2O_2 reactor for decolorization. The volumetric UV intensity at 50 W dm^{-3}, initial hydrogen peroxide concentration of 7.08 mg-mol dm^{-3} and azo dye concentration of 20.0 mg dm^{-3} (from 19.9 to 42.0 μg-mol dm^{-3}) is shown in Fig. 6. Six azo dyes in the same concentration were decolorized. Results showed that dis-azo dyes, C.I. Direct Yellow 4 (DY4) and C.I. Acid Black 1 (AB1), were the most difficult ones to be decolorized in the UV/H_2O_2 reactor. The mono-azo dyes were much easier to be treated in comparison with the dis-azo dyes. The retention time for 95% removal was 92 min for DY4, but 26 min for C.I. Acid Yellow 17 (AY17).

3.3. Ozonation of C.I. Direct Yellow 4 (DY4)

The decolorization of DY4 with initial concentration of 20.0 mg dm^{-3} was tested with ozone using an oxygen flow rate of 6.0 dm^3 min^{-1} as shown in Fig. 7. This follows pseudo-first order reaction, too. Also, the reaction was accompanied by the release of organic and inorganic acids to decrease pH from 5.8 to 3.4 with a retention time of 16 min and the concentration of DY4 decreased from 20.0 mg dm^{-3} to less than 0.1 mg dm^{-3}.

3.4. Comparison of three processes for decolorization of C.I. Direct Yellow 4

Fig. 8 gives the results of DY4 decolorization in different oxidation processes. The decolorization of DY4 by ozonation and UV enhanced ozonation are more effective than UV/H_2O_2 process. The two decolorization curves of O_3 and UV/O_3 almost overlapped implying that the presence or absence of UV radiation hardly affects the decolorization rate. However, it takes

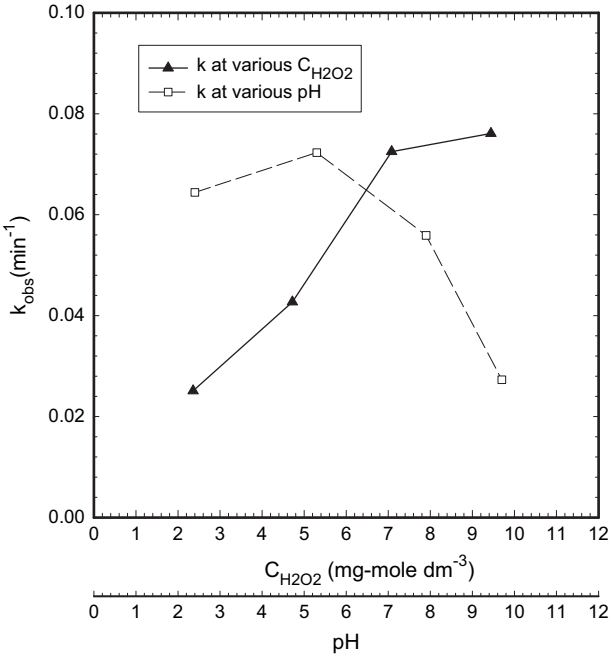

Fig. 5. The observed first order rate constants versus initial hydrogen peroxide concentration and pH in UV/H_2O_2 process. Initial dye concentration was 42.0 μg-mol dm^{-3}.

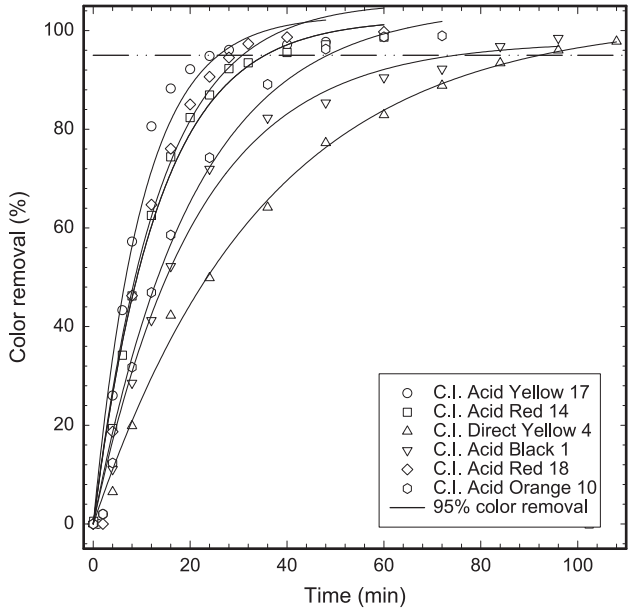

Fig. 6. Color removal efficiencies versus time for six azo dyes in UV/H_2O_2 process. Initial dye concentration was from 19.9 to 42.0 μg-mol dm^{-3} (fixed at 20.0 mg dm^{-3}) and hydrogen peroxide concentration was 7.08 mg-mol dm^{-3} at pH = 5.3.

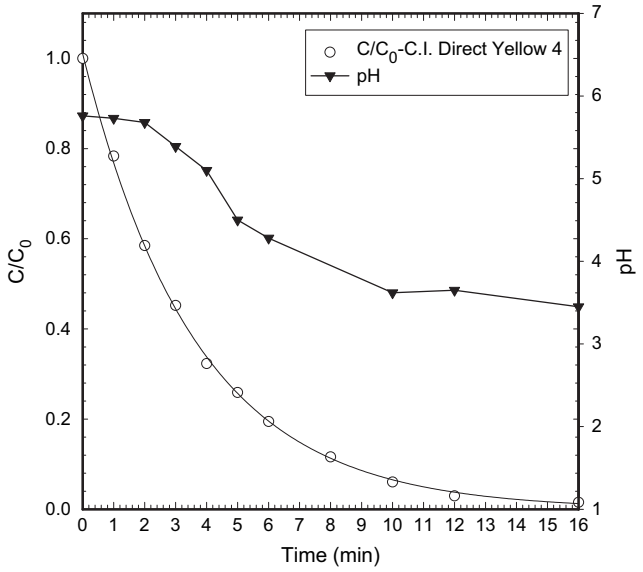

Fig. 7. The normalized concentrations (C_A/C_{A0}) of C.I. Direct Yellow 4 as functions of time in ozonation process at initial pH of 5.8. Initial dye concentration was 22.42 μg-mol dm^{-3} (20.0 mg dm^{-3}) and oxygen flow rate of 6.0 dm^3 min^{-1}. The ozone generator condition was set at 110 W and 105 V.

more than 90 min to decolorize DY4 with UV/H_2O_2 process and 11 min with O_3 and UV/O_3. Similar results are obtained for the other five azo dyes.

3.5. Comparison of three oxidation processes for decolorization of azo dyes

Fig. 9 shows the color removal of six different azo dyes by UV/O_3 process using initial dye concentration of

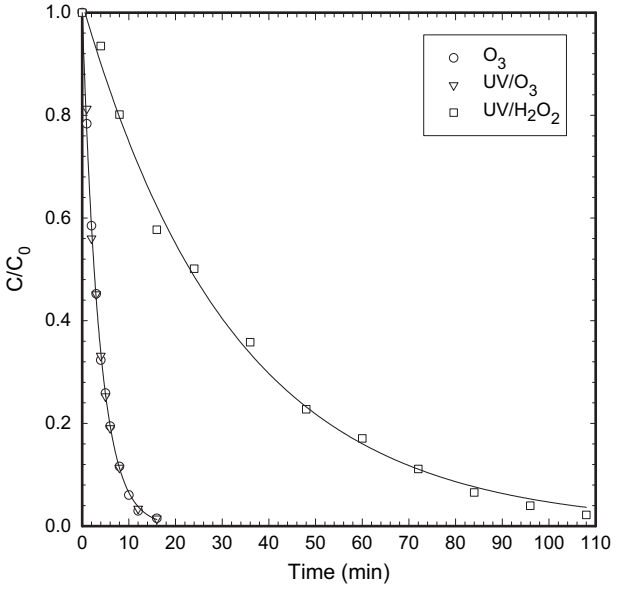

Fig. 8. The comparison of decolorization of C.I. Direct Yellow 4 under UV/H_2O_2 process, ozonation and UV/O_3 processes at initial dye concentration of 22.42 μg-mol dm^{-3}.

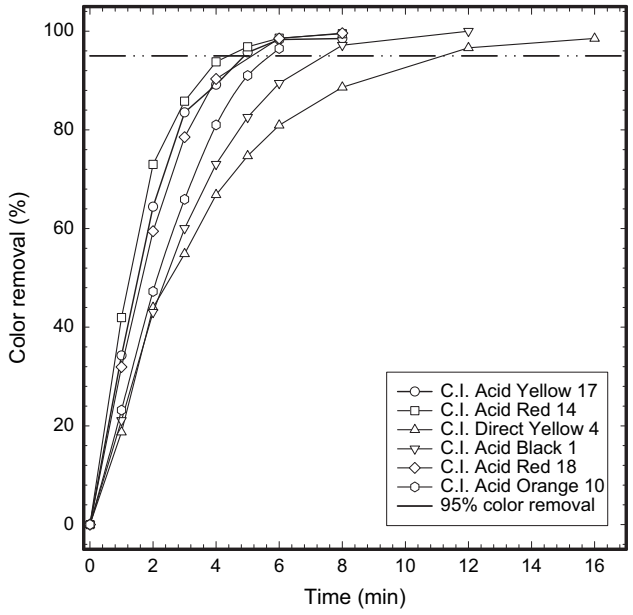

Fig. 9. The color removal of six azo dyes by UV/O_3 process. The initial dye concentration was from 19.9 to 42.0 μg-mol dm^{-3} (fixed at 20.0 mg dm^{-3}) and oxygen flow rate was 6.0 dm^3 min^{-1}. The ozone generator condition was set at 110 W and 105 V.

20.0 mg dm^{-3} (from 19.9 to 42.0 μg-mol dm^{-3}) and oxygen flow rate of 6.0 dm^3 min^{-1}. More than 95% of color can be removed in less than 11.5 min of all six azo dyes. The dis-azo dyes AB1 and DY4 are the most resistant to be decolorized by UV/O_3 process, as well as the UV/H_2O_2 process. Fig. 10 summarizes the pseudo-first order rate constants of decolorization of six azo dyes by O_3, UV/O_3 and UV/H_2O_2 processes with each dye having an initial concentration of 20.0 mg dm^{-3}.

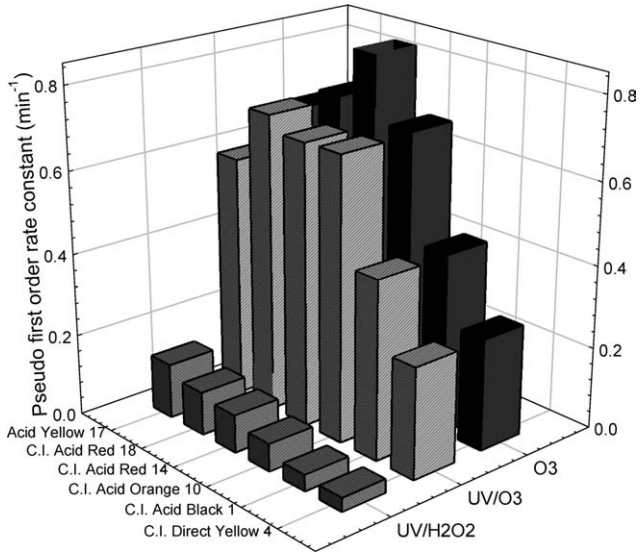

Fig. 10. The pseudo-first order rate constants of six azo dyes under UV/H_2O_2 process, ozonation and UV/O_3 processes at initial dye concentration of 20.0 mg dm^{-3}.

Table 2
Retention time, power and chemicals needed for 95% removal of six azo dyes

Azo dyes	Retention time (min)			Power (10^{-3} kWh)			Chemicals		
	O_3	UV/O_3	UV/H_2O_2	O_3	UV/O_3	UV/H_2O_2	O_3 (oxygen, l)	UV/O_3 (oxygen, l)	UV/H_2O_2 (H_2O_2, ml)
C.I. Direct Yellow 4	11	11.1	92	20.2	945.4	7666.7	66	66.6	60
C.I. Acid Black 1	6.8	6.9	75	12.5	587.7	6250	40.8	41.4	60
C.I. Acid Orange 10	4.3	4.4	48.5	7.9	374.7	4041.7	25.8	26.4	60
C.I. Acid Red 14	3.2	4.4	36.5	5.9	374.7	3041.7	19.2	26.4	60
C.I. Acid Red 18	4.1	4.1	30.8	7.5	349.2	2566.7	24.6	24.6	60
C.I. Acid Yellow 17	4.4	5.1	25.7	8.1	434.4	2141.7	26.4	30.6	60

The dis-azo dyes (AB1 and DY4) were shown to be difficult to be decolorized than other mono-azo dyes in these oxidation processes and their observed first order rate constants are very small. Especially, for DY4, where the rate constants were less than that of one half of mono-azo dyes such as C.I. Acid Red 14 and C.I. Acid Red 18. Thus, it can be seen that the chemical structures of azo dyes affect the decolorization efficiency. The more the azo links, the more difficult the decolorization was in the processes of O_3, UV/O_3, or UV/H_2O_2.

The retention time and power demand of 95% color removal are tabulated for the six azo dyes in Table 2. The retention time demand for 95% color removal by UV/H_2O_2 process is extremely larger than that by the O_3 and UV/O_3 processes. The power demand of 95% color removal for each azo dye is higher for UV/H_2O_2. It is from 5 to 11 times and from 265 to 520 times higher than UV/O_3 and O_3 processes, respectively. In addition, the power demand of O_3 to remove 95% color is very small. In Table 2, the chemicals needed for the three oxidation processes are also tabulated. Pure oxygen gas demand is from 19.2 to 66.0 dm^3 in both the ozonation and UV/O_3 processes. The hydrogen peroxide dosage of 60.0 ml for experiments is constant for UV/H_2O_2 process. Therefore, suitable color removal parameters can be selected in terms of retention time, power demand and reagent dosages for the three oxidation processes.

4. Conclusions

The decolorization rates of six azo dyes have been determined in O_3, UV/O_3 and UV/H_2O_2 processes. The selected dyes were all efficiently decolorized by the three processes. The results showed that the three oxidation processes are feasible pretreatment methods for complete decolorization of dye wastewater from textile industries. On the other hand, the rate of decolorization of azo dyes to 95% can be achieved in 10–110 min depending on the condition. C.I. Direct Yellow 4 (DY4) is the most difficult to be decolorized no matter which oxidation process is used. However, the most efficient dyes to be decolorized are C.I. Acid Red 14 (AR14) by ozonation, C.I. Acid Red 18 by UV/O_3 process, and C.I. Acid Yellow 17 by UV/H_2O_2 process. Also, dis-azo dyes, C.I. Direct Yellow 4 (DY4) and C.I. Acid Black 1 (AB1), are more difficult to be decolorized than mono-azo dyes. For example, the decolorization of DY4 was achieved in 110 min by UV/H_2O_2, yet only about 16 min by UV/O_3, and O_3 process. Estimating the power consumption to reach 95% color removal, DY4 consumes 7666.7×10^{-3} kWh by UV/H_2O_2 process, AR14 only consumes 5.9×10^{-3} kWh by O_3 process. Thus, there are about 1000-fold difference between the minimum and maximum energy demands.

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