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Removal of C.I. Reactive Red 2 by low pressure UV/chlorine advanced oxidation

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ABSTRACT

Azo dyes are commonly found as pollutants in wastewater from the textile industry, and can cause environmental problems because of their color and toxicity. The removal of a typical azo dye named C.I. Reactive Red 2 (RR2) during low pressure ultraviolet (UV)/chlorine oxidation was investigated in this study. UV irradiation at 254 nm and addition of free chlorine provided much higher removal rates of RR2 and color than UV irradiation or chlorination alone. Increasing the free chlorine dose enhanced the removal efficiency of RR2 and color by UV/chlorine oxidation. Experiments performed with nitrobenzene (NB) or benzoic acid (BA) as scavengers showed that radicals (especially $^{\cdot}OH$) formed during UV/chlorine oxidation are important in the RR2 removal. Addition of HCO_3^- and Cl^- to the RR2 solution did not inhibit the removal of RR2 during UV/chlorine oxidation.
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Introduction

Textile wastewater is an important contributor to environmental pollution of water in many countries. In China in 2012, textile industry discharges into waterways accounted for 11% and 9% of the total wastewater volumes and chemical oxygen demand discharges from all industrial sources, respectively (China Statistical Yearbook on Environment, 2013). Wastewater from dyeing and finishing processes plays a dominant role in pollution from the textile industry, because of the high concentrations of COD, salinity, colors, and toxic substances in these waste streams (Solís et al., 2012). Therefore, control of wastewater pollution from these sources has received increasing attention around the world.

Azo dyes are widely used in dyeing and finishing processes, and are present in 60%–70% of all commercial dyes (Işık and Sponza, 2005). Azo dyes are strong colorants, and have high bio-recalcitrance and toxicity because of their nitrogen–nitrogen double bonds (Cai et al., 2015). Untreated azo dye effluents are toxic and mutagenic to aquatic organisms in receiving waters (Panda and Mathews, 2014). Therefore, treatment technologies for removal of azo dyes are important.

Because azo dyes are bio-recalcitrant (Brown and Hamburger, 1987), chemical oxidation processes are frequently used to

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remove color from secondary effluents. Ozonation and Fenton oxidation are useful for removing some dyes. Ozonation quickly decolorizes colored wastewater *via* oxidative cleavage of the conjugated bonds in dye molecules (Tehrani-Bagha et al., 2010). Idel-aouad et al. (2011) found that the heterogeneous Fenton process using Fe(II)-exchanged synthetic Y Zeolite provided good performance in decolorization of a C.I. Acid Red 14 solution at pH 3.13–7.27. However, ozonation is very expensive, and Fenton oxidation produces large quantities of iron-containing sludge. Recently, the ultraviolet (UV)/H_2O_2 process has been considered as a promising technology for azo dye removal. UV/H_2O_2-generated hydroxyl radicals can react non-selectively with a broad range of organic compounds (Narayanasamy and Murugesan, 2014; Kasiri and Khataee, 2012). The principal advantage of the UV/H_2O_2 technology is that no sludge is created after treatment (Behnajady et al., 2006). However, H_2O_2 is expensive.

Recently, UV/chlorine oxidation has been reported as a novel advanced oxidation process (Feng et al., 2007, 2010). During UV/chlorine oxidation, free chlorine, including HOCl and OCl^-, absorbs UV photons and produces several radicals, including hydroxyl ($\cdot OH$) and chlorine ($\cdot Cl$) radicals (Nowell and Hoigné, 1992). The molar absorption coefficients of HOCl and OCl^- at 254 nm have been determined to be 59 ± 1 and 66 ± 1 L/(mol · cm) respectively (Feng et al., 2007), whereas the molar absorption coefficient of H_2O_2 at 254 nm is only 18.4 L/(mol · cm) (Stefan et al., 1996). During UV irradiation, HOCl has a much smaller $\cdot OH$ scavenging rate than H_2O_2 (Watts and Linden, 2007). The yield of $\cdot OH$ during the UV/chlorine oxidation process is affected by the oxidation conditions, including pH and concentrations of the pollutants. The $\cdot OH$ radicals produced have been used for removing pollutants such as nitrobenzene, trichloroethylene, and iopamidol (Watts and Linden, 2007; Wang et al., 2012; Sichel et al., 2011). Furthermore, $\cdot Cl$ radicals formed during the UV/chlorine oxidation process may also play an important role in pollutant degradation. In one study (Fang et al., 2014), the $\cdot Cl$ radicals removed 62%–65% of the benzoic acid. Reaction rates of $\cdot Cl$ with some compounds containing electrophilic functional groups, such as acetic acid, benzoic acid, and phenol, are much higher than those of $\cdot OH$ (Fang et al., 2014). However, the removal of azo dyes using UV/chlorine oxidation, and especially under high salinity and alkalinity conditions, has not been reported.

The objective of this study was to evaluate the removal efficiency of azo dyes by low pressure UV/chlorine oxidation. The effects of UV irradiation doses, chlorine doses, pH values, and ion concentrations on azo dye removal during UV/chlorine oxidation were investigated. The roles of radicals during the UV/chlorine oxidation were also evaluated.

1. Materials and methods

1.1. Chemicals and materials

The commercial azo dye C.I. Reactive Red 2 (RR2) (CAS No. 17804-49-8) was obtained from Jiaying Chemical Company (Jiaying Chemical Company, Shanghai, China) and used without further purification. Sodium hypochlorite solution (13% available chlorine) was purchased from J&K Company (J&K, Beijing, China). Acetonitrile was of HPLC grade. Na_2SO_3 was of analytical grade. All solutions were prepared with ultrapure water (18.25 MΩ) from a Milli-Q purification system (Milli-Q, Millipore, USA).

1.2. RR2 degradation by UV/chlorine oxidation

The UV/chlorine oxidation experiments were conducted in a UV-C irradiation system with an 80-W UV-C lamp at 254 nm (80 W, Light Sources, USA) at 298 K. Free chlorine was added to RR2 solutions (20 mg/L initial RR2, 100 mL) in 120-mL beakers with an inner diameter of 10 cm to give free chlorine doses of 0–20 mg/L. Then the mixture was placed under the UV light and continuously stirred using magnetic agitators at ambient temperature (25 ± 2°C); a schematic diagram is shown in Fig. 1. The quantum yield was calculated at 0.740 ± 0.021 and the incident photon flux (I_0) was determined to be 0.325 μEinstein/sec using KI/KIO_3 actinometry (Bolton et al., 2011). The corresponding average UV fluence rate (E_{avg}) was about 2.0 mW/cm^2. The UV fluence was calculated as the fluence rate multiplied by the exposure time (Rosenfeldt et al., 2006). After UV/chlorine oxidation, Na_2SO_3 was used to remove residual chlorine in the solutions for subsequent RR2, color, and total organic carbon (TOC) analysis. All the experiments were conducted with at least duplicate measurements.

Experiments were conducted to evaluate the effect of pH on RR2 removal. The pH value of the dye solution was kept at 4, 7, 7.5, 8 or 9 by addition of phosphate buffer (final concentration 20 mmol/L).

To evaluate the effect of radicals on RR2 removal, NB and BA at final doses of 10 mg/L and 50 mg/L, respectively, were added into a RR2 solution containing free chlorine (5 mg free chlorine/L). NB was used to quench $\cdot OH$ during UV/chlorine oxidation, while BA was a scavenger of $\cdot OH$ and $\cdot Cl$.

1.3. Analytical methods

The RR2 concentrations in the solutions after UV/chlorine oxidation were determined by high performance liquid chromatography (LC20, Shimadzu, Japan) with detection at 538 nm. A reversed-phase column (ODS-C18, Bonna-Agela China, Tianjin, China) was used for separation, with a mobile phase of 50% acetonitrile and 50% ultrapure water at a flow rate of 0.5 mL/min.

The color, UV absorbance spectrum, and TOC values of the dye solutions after UV/chlorine oxidation were determined using a color analyzer (SD-9012, Xin Rui, China), ultraviolet–visible (UV–Vis) spectrophotometer (UV-2450, Shimadzu, Japan), and TOC analyzer (TOC-L, Shimadzu, Japan).

2. Results and discussion

2.1. Degradation of azo dye during UV/chlorine oxidation

2.1.1. *Removal of azo dye and its color*

The changes in dye concentration and color of RR2 solutions after UV/chlorine oxidation, after only UV irradiation, and after only chlorination are shown in Fig. 2. After only UV

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