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Degradation of aryl-azo-naphthol dyes by ultrasound, ozone and their combination: Effect of α-substituents

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Abstract

Lab-scale degradation of azo dyes with ultrasound (300 kHz), ozone and both was investigated using an aryl-azo-naphthol dye-C.I. Acid Orange 8. It was found that in all schemes color decay was faster than UV absorbance, and the rates followed pseudo-firstorder kinetics except for the decay of UV-254 band by ozone. Sonication alone was sufficient for decolorization, but not for UV absorption abatement or mineralization. Ozonation was more effective than ultrasound in bleaching, but not as much for the mineralization of the dye. Combined operation of ultrasound and ozone improved the rate of bleaching and UV absorption decay and remarkably enhanced the mineralization of the dye. This was attributed to increased mass transfer of ozone in solution and its decomposition in the gas phase to yield hydroxyl radicals and other oxidative species. The effect of α -methyl substituent at the aryl carbon of the dye was found to decelerate the rate of degradation as a result of weakened intramolecular hydrogen bonding. © 2005 Elsevier B.V. All rights reserved.

Keywords: Aryl-azo-naphthol; Chromophore; Hydroxyl radical; Electrophilic; Sonolysis; Ozonation; Mineralization; α -substituent; Intramolecular hydrogen bonding

1. Introduction

Textile dyehouse effluents contain wasted dyestuff in quantities directly related to the degree of imperfect fixing, and thus are difficult to treat by conventional processes involving biological and chemical methods. When these effluents are discharged into water without proper treatment, the dyes are transported over long distances because of their high solubility, and some are hydrolyzed or anaerobically metabolized to form carcinogenic end products [1,2]. Moreover, dyeing mills consume such large amounts of water that in-plant water recycling, which requires proper decontamination of all effluents, is a crucial step in dyehouse wastewater management [3].

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Lab-scale application of advanced oxidation processes (AOPs) using a source of ultraviolet light and oxidizing chemicals such as ozone, hydrogen peroxide and Fenton's reagent are found extremely effective for decolorizing dyeing mill effluents [4–6]. AOPs are based on in situ generation of very reactive free radicals such as the hydroxyl radical ('OH), which owing to its non-selectivity is an exceptionally powerful oxidant. The use of ultrasound to generate hydroxyl radicals is a relatively novel technique in AOP, and the method has attracted attention particularly for its decolorization effects [7–12].

The phenomenon responsible for hydroxyl radical formation during sonication of water is cavitation, which consists of the formation, growth and expansion of micro bubbles within the compression-rarefaction cycles of acoustic waves [13]. When these bubbles expand to sufficiently large sizes, they collapse by violent implosions to release extreme temperatures and pressures at

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local hot spots in the liquid. Under these conditions, molecules of water entrapped in the gaseous bubble interiors dissociate into hydroxyl and hydrogen radicals, which undergo a number of chain reactions including the generation of H_2O_2 [14].

The use of ozone in AOP is widely popular with the advantage that ozone reacts with organic compounds either directly at acidic/neutral conditions and/or indirectly by decomposition at highly alkaline pH [15]. The rate-limiting parameter in decolorization of azo dye solutions by ozone is the mass transfer rate of ozone in the solution [16]. Combined operation of sonolysis and ozonation renders synergistic effects in organic matter destruction [17–19]. The synergy is partly due to exhydroxyl radicals formed cess upon thermal decomposition of ozone in the gas phase (Reactions (1) and (2)), and partly to increased mass transfer of ozone in solution by mechanical effects of ultrasound [17,18]:

$$\mathbf{O}_3+))) \to \mathbf{O}_2(\mathbf{g}) + \mathbf{O}(^3\mathbf{P})(\mathbf{g}) \tag{1}$$

$$O(^{3}P)(g) + H_{2}O(g) \rightarrow 2 OH$$
 (2)

The objective of this study was to investigate and compare the effects of ultrasound (US), ozone (O₃) and their combination (US/O₃) on the degradation of aryl-azonaphthol dyes. The test dye C.I. Acid Orange 8 (AO8) selected from commercial dyestuff was a mono-azo water soluble dye that is widely consumed in the textile industry for its bright color and good fastness properties [20]. The impact of dye structure regarding α -methyl substituents was assessed by testing a similar dye (C.I. Acid Orange 7) that had no α -methyl attachment.

2. Materials and methods

C.I. Acid Orange 8 (MW = 364.4 g mol^{-1}) was obtained from Aldrich in 65% purity and used without further purification. The molecular structure of the dye in equilibrium between its azo and hydrazone tautomers is given below:

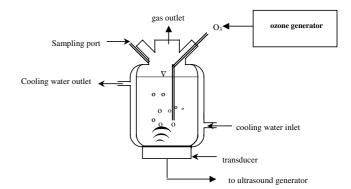


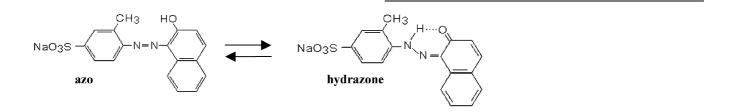
Fig. 1. Schematic diagram of the experimental set-up.

300 kHz as generated by electricity at 25 W. Argon was injected during all single US operations to enhance bubble formation and to compensate for degassing effects. The ultrasonic power input in the reactor was 0.184 W ml⁻¹ as determined by calorimetry [21]. Ozone was generated electrically from dry pure oxygen at a maximum rate of 31 min^{-1} using an Ozone Lab-100 model generator. A schematic diagram of the set-up is presented in Fig. 1.

2.1. Procedure

A 3 mM stock solution was made by dissolving 0.168 g of AO8 in 100 ml of boiling deionized water during magnetic stirring, and stored in the dark at 4 °C prior to use. Dye solutions of various strengths were prepared from the stock by proper dilutions with deionized water followed by 2-h aeration. The pH of all test solutions after saturation with air was 5.0 and no pH adjustment was made in any test scheme.

The procedure consisted of selection of the initial dye concentration and optimum ozone input; and exposure of 100 ml air-saturated test solutions to US, O_3 and US/O₃ combination, respectively at optimized conditions. In all schemes, samples were collected at various times during operation and analyzed for color, UV absorbance, total organic carbon (TOC) and H₂O₂.



The experiments were carried out in a 150 ml glass reactor surrounded by a cooling jacket and consisting of a plate type transducer emitting ultrasonic waves at The performance of the systems for color decay was assessed by the degree of reduction in the maximum absorbance of the dye in the visible band during 30 min operation, while aromatic and naphthol ring degradaDownload English Version:

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