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Degradation of C.I. Acid Orange 7 by ultrasound enhanced ozonation in a rectangular air-lift reactor

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Abstract

The combination of 20 kHz ultrasound and ozone for the degradation of C.I. Acid Orange 7 was studied. The effect of power density, gas flow rate, initial pH, hydroxyl radical scavenger, and initial dye concentration on the decolorization of C.I. Acid Orange 7 was investigated. The decolorization of C.I. Acid Orange 7 fits the pseudo-half-order kinetic model under most of the operating conditions. The decolorization rate increased with the increase of power density and gas flow rate, but decreased with the increasing initial dye concentration. Either pH or sodium chloride has little effect on the decolorization rate, indicating that the low frequency ultrasound enhanced ozonation process for the decolorization of C.I. Acid Orange 7 is mainly a direct reaction rather than radical reaction. Moreover, the decolorization rate increased with sodium carbonate to dye moalr ratio. The synergistic effect of C.I. Acid Orange 7 mineralization by ultrasound enhanced ozonation was significant when the system temperature was raised due to the heat effect of ultrasonic irradiation.

Keywords: Ultrasound; Ozone; C.I. Acid Orange 7; Decolorization; TOC; Synergistic

1. Introduction

The disposal of wastewater containing azo dyes is an environmental concern since the associated color is quite noticeable to the public, and some dyes may have carcinogenic and/or teratogenic effects on public health [1]. Decolorization of wastewaters is one of the significant problems as the dye will be visible even at low concentration. Most of the dyes are found to be resistant to the conventional sewage treatment process as they are designed to resistant chemical and photochemical degradation [2]. Ozone has been used for the treatment of refractory wastewater such as textile industry wastewater [3]. However, there are a few disadvantages which limit industrial applications of ozonation. The first is the high production cost of ozone and the low ozone utilization due to poor mass transfer rate of ozone [4,5]. Furthermore, though ozone could readily decoloralize dye wastewater, it could not mineralize dye wastewater effectively due to the limit oxidizing power of ozone ($E^0 = 2.07$ V). One method of enhancing ozonation reactions is to simultaneously apply ultrasonic irradiation [2,6–23].

Over the last few years there have been a lot of reports on the application of ultrasound for the destruction of aqueous inorganic and organic pollutants such as azo dyes [24–29]. When the aqueous solutions containing the organic pollutants such as azo dyes are exposed to an acoustic field, dissolved gases and water vapor are entrapped by cavitation bubbles, which expand at rarefaction cycles of the bubbles and release extreme temperatures upon adiabatic collapse. Under these conditions, bubble contents are pyrolytically dissociated into free radicals, some of which could diffuse into the aqueous phase [26]. Pyrolytic reactions inside or near the bubble and radical reactions in the solution are regarded as the two major pathways for sonochemical degradation [29]. Sonochemical destruction is particularly effective for volatile substrates as these solutes can be directly combusted within the gas phase of the hot collapsing cavitation bubbles [6]. The effect of ultrasound waves on the hydrophilic chemical oxidation is due to the production of hydroxyl radicals during the cavitation-induced thermal decomposition of water [30]. The rate constant of radical type reaction is relatively too low as approximately only 10% of the hydroxyl radicals generating in the cavitation bubbles could escape to the bulk liquid [31]. Therefore, much effort has been devoted to accelerate the decomposition rates of hydrophilic compounds by means of the combination with other methods.

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The coupling of ultrasonic irradiation with ozonation (US/O_3) may provide a possible advanced oxidation process (AOP), in which hydroxyl radical, a more effective oxidant ($E^0 = 2.87$ V) than ozone, is generated in sufficient quantity to affect wastewater treatment. In the presence of ultrasonic irradiation, ozone is decomposed thermolytically in the vapor phase of a cavitation bubble [32]:

$$O_3(g) +))) \rightarrow O_2(g) + O(^3P)(g) \quad (1)$$

$$O(^3P)(g) + H_2O(g) \rightarrow 2^{\bullet}OH(g) \quad (2)$$

where the symbol ")))" indicates ultrasonic irradiation. These decomposition reactions occur in the gas phase. The reaction products migrate to the interfacial sheath of the bubble where they subsequently react in the aqueous phase [13]. In addition, ultrasonic irradiation has been demonstrated to increase the mass transfer of ozone to aqueous phase by means of increasing mass transfer coefficient [6,11,15,33,34]. Until now, more attention was paid to the employment of conventional reactors such as stirred tank reactors or bubble columns to perform the combine US/O_3 process. However, the reactor with more efficient mass-transfer was seldom used in the coupled US/O_3 process. The airlift reactors have been regarded as a promising type of gas–liquid reactor due to their good mixing with low shear stress and energy consumption as well as their advantages of high gas–liquid mass and heat transfer [35]. At present airlift reactors have been widely used in biotechnological processes, especially in biological wastewater treatment [35]. An airlift reactor is mainly composed of two zones, the riser and the downcomer. The riser lane corresponds to the zone where the gas phase is injected and the fluid and gas travel upwards together; in the downcomer lane the gas and liquid travel downwards [36]. To our knowledge, only Bando et al. conducted the combined US/O_3 process in a rectangular air-lift reactor [17]. However, the effect of various operation conditions on the coupled US/O_3 process in this kind of reactor was not investigated.

During the US/O_3 process, the system temperature would be raised due to the heat effect of ultrasonic irradiation. The temperature effect on sonochemical reaction and chemical reaction is different. The reactor cooling was usually provided in most of the reports as the largest sonochemical effects are observed at lower temperatures [31]. Martins et al. [18] observed that the combined US/O_3 process for the decolorization of pararosaniline dye was less efficient than ozonation alone due to the temperature increase when no reactor cooling was provided. However, Lall et al. [15] and He et al. [22] reported that the decolorization rate was enhanced by increasing the temperature in the coupled US/O_3 process. Our previous work also demonstrated the synergistic effect of the combined US/O_3 process for the decolorization of methyl orange was more significant when the system temperature was raised from 16 to 28 °C [20]. In this case the cooling water would not be needed and the operation cost for cooling water would be saved. Therefore, the objective of this study was to treat C.I. Acid Orange 7, a model azo dye, by the coupled US/O_3 process in a rectangular airlift reactor. The effects of operating conditions such as power density, gas flow rate, initial pH, hydroxyl radical scavenger, and initial dye concentration on the decolorization efficiency were investigated. The synergistic effect of the combined US/O_3 process for the mineralization of C.I. Acid Orange 7 was also explored.

2. Materials and methods

Stock solution of C.I. Acid Orange 7 was prepared in distilled water before each run. H_2SO_4 or NaOH was used to adjust the initial pH (pH_0) of the dye solution. The stock solution was fed into a rectangular air-lift reactor made of plexy glass (see Fig. 1). This reactor consists of a square column (50 mm × 30 mm) with the height of 120 mm, divided into a riser and a downcomer section by a plexy glass baffle (width: 30 mm; thickness: 4 mm; total height: 55 mm). The riser-to-downcomer cross-sectional area ratio was equal to 1.56. The baffle was located at a distance of 5 mm from the bottom of the reactor. The gas distributor at the bottom of the riser was a perforated tube with six orifices of 1 mm diameter.

Sonication was performed with a KS-250 ultrasonic generator (250 W, 20 kHz, Ningbo Kesheng Instrument Co., China) equipped with a titanium probe transducer. The tip of the probe was 1 cm in diameter and was placed 1.5 cm into the liquid layer. The sonication was administered in pulses with a 50% duty cycle. The acoustic power (P) was determined calorimetrically [37].

Ozone was bubbled into the solution using an ozone generator (XFZ-5QI, China). The gaseous ozone concentration was monitored by the iodometric method with potassium iodide solution [38]. A predetermined amount of aliquot was removed with pipette at different time intervals. The absorbance of the solution was measured using a Shimadzu UV-1600 spectrophotometer after dilution with distilled water. Total organic carbon (TOC) was determined using Multi N/C®2100, Analytik Jena AG.

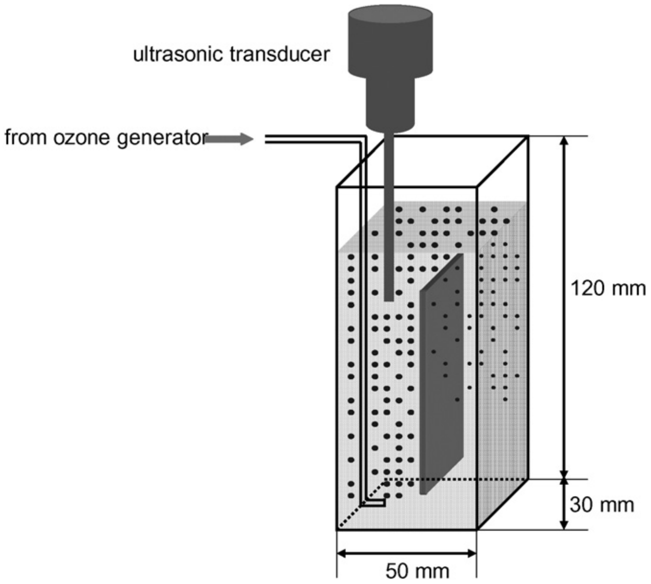

Fig. 1. The experimental set up.