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Decomposition and decoloration of dyeing wastewater by hydrothermal oxidation

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A B S T R A C T

The objective of this research was to investigate the decomposition and decoloration of textile wastewater by hydrothermal treatment. We studied the hydrothermal oxidation of methyl orange aqueous solution using a flow-type reactor packed with MnO₂ catalyst. Hot steam (vapor) and subcritical water (liquid) were used in the reactor as the decomposition media at the reaction temperature and pressure of 200–300 °C and 1 or 10 MPa, respectively. When methyl orange solution with 300 mg/L of total organic carbon (TOC) was used, nearly complete decomposition of methyl orange was realized in hot steam at 300 °C, 1 MPa, 10 s, and 1.5 oxygen supply ratio, i.e., a much higher TOC decomposition rate was obtained at a lower reaction pressure and shorter residence time than those obtained with subcritical water oxidation. The kinetic modeling of hydrothermal oxidation of the model wastewater containing methyl orange was also investigated. The kinetic model to calculate TOC conversion at a given residence time was proposed based on the regression from the complete set of data. In the case of hot steam oxidation, a second-order kinetic model was used to describe the TOC conversion. However, a two-step first-order kinetic model (a rapid first reaction followed by a slow second reaction) was needed to describe the kinetics of subcritical water oxidation. Both reaction rate expressions were successfully used to estimate the TOC concentrations at any residence time at 90% confidence level.

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1. Introduction

Water pollution adversely affects the environment and it is becoming increasingly serious. Industrial plants generate wide varieties of water pollutants, some of which are too difficult and costly to treat; for example, wastewaters containing synthetic colors, including dyes and pigments. Such wastewaters are high in both color and organic content.

At present, more than 100,000 different dyes are synthesized on the industrial scale and utilized by textile companies. Generally, dyes are classified based on their chemical structures. Acid dyes are one of the most widely used dyes in the textile industry.

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They are water-soluble anionic dyes, containing acidic groups along with one or more azo groups, and are applied to all kinds of natural fibers such as wool, cotton, and silk as well as synthetic fibers such as polyesters, acrylic, and rayon. Dyestuff manufacturing processes and the subsequent dyeing and cleaning processes used in textile manufacturing plants release a large amount of dyes into the environment as wastewater. The release of such wastewaters to water bodies is highly deleterious to the environment, because they prevent the penetration of sunlight to water layers, thereby decreasing the photosynthetic activity in the water bodies [\[1\]](#page--1-0) and threatening their health. The global textile industry consumes more than $10⁴$ ton dyes/year and discharges about $10³$ ton dyes/year into water bodies [\[2\].](#page--1-0)

Therefore, it is essential that such wastewaters are treated before they are discharged into water bodies. Several types of conventional and traditional techniques have been reported for their treatment. Some conventional methods for treating dyeing wastewaters include adsorption [\[3\],](#page--1-0) flocculation [\[4\],](#page--1-0)

Fig. 1. Flow-type reactor system.

electrochemical methods $[5]$, membrane separation $[6]$, ozonation [\[7\],](#page--1-0) and advanced oxidation using $UV/H₂O₂$ [\[8\].](#page--1-0) However, such approaches are expensive for larger effluent streams. Moreover, most azo dyes are generally chemically stable and resistant to degradation.

Some advanced oxidation treatments for dyeing wastewater are wet air oxidation (WAO) and catalytic wet air oxidation (CWAO), which are performed using subcritical water. These processes are considered as pretreatment steps for the conventional biological treatment of dyeing wastewaters. Oxygen and hydrogen peroxide $(H₂O₂)$ are generally used as oxidants in such treatments. In the literature, the treatment efficiencies of different dyes by WAO have been reported as 50–90% removal rate of chemical oxygen demand (COD) or total organic carbon (TOC) at the reaction temperatures of 150–300 ◦C and residence times of 30–240 min in batch or continuous flow reactors $[9-11]$. In CWAO, Al-Cu, CoAlPO₄-5, CeO₂, and Pd–Pt/Al₂O₃ are used as catalysts. The initial 1 g/L of reactive dyes could be completely removed in 20 min with 10 g Al–Cu at atmospheric pressure and 80 \degree C. CoAlPO₄-5 effectively decreased COD values in a dye solution (Bestcolor-21, dark green) at 135 ◦C and 1.0 MPa, with color and COD removals being as high as 95% and 90%, respectively. The $CeO₂$ catalyst demonstrated nearly 100% color removal above 135 ◦C, and COD removal of above 95% at 165 ◦C. The effluent from the dyeing industry was successfully treated in a continuous flow reactor by CWAO on the 0.2 wt% Pd–1.0 wt% Pt/Al₂O₃ catalyst with H_2/O_2 mixture and complete removal of both TOC and color was achieved [\[11–14\].](#page--1-0)

The supercritical and subcritical water oxidation (SCWO and Sub-CWO, respectively) methods show excellent treatment ability, allowing clean and efficient decomposition of many aqueous organic wastes. The SCWO method seems to be a promising alternative for conventional treatments of textile wastewater. Söğüt and Akgün [\[15\]](#page--1-0) investigated the decomposition of an aqueous solution of a model azo dye pollutant (C.I. Disperse Orange 25) using H_2O_2 as an oxidant by SCWO in a continuous flow reactor. They achieved the decomposition rate of up to 98.52% in very short residence times of 4–12 s. Somayeh et al. [\[16\]](#page--1-0) studied the decomposition and decoloration of acid orange 7 (AO7) in subcritical water. The operating temperature was 180–374 ◦C at a wide range of residence time of 10–400 s. AO7 completely decomposed at higher temperatures or when longer residence times were used.

However, to the best of our knowledge, reaction kinetics of dyeing wastewater in steam/subcritical water with a catalyst have not been reported till now. The objective of this research was to investigate the decomposition and decoloration of textile wastewater by hydrothermal treatment. In this research, an aqueous solution of methyl orange was selected as a model for textile wastewater under steam/subcritical water conditions.

2. Experimental

2.1. Continuous apparatus

Fig. 1 shows a flow-type experimental setup. The two feed streams were pumped in two different lines and then separately preheated (reaction temperature). Two syringe pumps (Model 500D, Teledyne Isco, USA) were used to pump the feed to the reactor at the required flow rates (0.96–12 mL/min), respectively, which depended on the required residence times (1.5–1800 s) in the reactor. The reactor used was type-316 stainless steel tube, with the following dimensions: 3/4 in. of diameter; 1.65 mm of thickness; 700 mm of length; and 117.2 mL of inner volume. The central 400 mm part of the reactor was filled with $MnO₂$ particles as catalyst. Other spaces of the reactor were filled with 3 mm $\varphi\alpha$ -alumina balls (Sumitomo Chemical Co., Ltd., Japan). The preheating coil for methyl orange aqueous solution was made of a 1/8 in. spiral tube of type-316 stainless steel with a length of 2000 mm and an inner volume of 11.24 mL, and the preheating coil for H_2O_2 aqueous solution was made of a 1/16 in. spiral tube of type-316 stainless steel with a length of 2000 mm and an inner volume of 1.86 mL. The reactor and preheating coil were placed in three PID-controlled electric furnaces (ARF-50K, Asahi-Rika Works Co., Ltd., Japan). The temperatures of the reactor and preheating coil were measured by using K-type thermocouples. H_2O_2 was used as the oxidant source and it was completely decomposed to oxygen and water in the preheater section above 200 ℃. After preheating, oxygen and the feed of the model wastewater were reacted on the $MnO₂$ catalysts. The effluent from the reactor was rapidly cooled by using a cooling coil kept in a glass water bath (W400 \times D250 \times H280 mm). Then, the particles were filtered out by using a 0.5 - μ m inline filter (Swagelok Co., USA). The effluent was depressurized by a back-pressure regulator (Tescom Co., USA) and the liquid product was collected in a graduated cylinder (30 cc, AS ONE Co., Japan).

2.2. Calculation of residence time

The residence time in the reactor packed with the $MnO₂$ catalyst was calculated by using the following equation:

$$
t = \frac{V_{\text{MnO}_2} \times \rho_{\text{SC}}}{F_{\text{T}} \times \rho_{\text{L}}}
$$
 (1)

where $V_{\rm MnO_2}$ is the volume (mL) of the MnO₂ catalyst, $\rho_{\rm SC}$ is the density (g/mL) of fluid at the reaction pressure and temperature, F_T is the total volumetric flow rate (mL/s) by considering the volumes of both the model wastewater and the oxidant fed to the system, and ρ_L is the density (g/mL) of fluid under feed pump conditions. Since the investigations were performed with diluted aqueous solutions Download English Version:

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