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Removal of Acid Black 1 from water by the pulsed corona discharge advanced oxidation method

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ABSTRACT

Water pollution is a major global problem. Organic pollutants are one of the major groups of toxic and carcinogenic contaminants that, due to their complex structures, show resistance to biodegradation processes. The pulsed corona discharge (PCD) advanced oxidation technology, with a point-to-point configuration of the electrodes immersed in the aqueous solution, was used for decomposing Acid Black 1 (AB1) as a representative of synthetic organic azo dyes. The effects of electrical field frequencies (60 and 120 Hz) and electrode gap spaces (2, 4 and 6 mm) on decomposition of AB1 were investigated. The largest decomposition achieved was 99.93% with optimal conditions of electrical field frequencies of 120 Hz and an 8 mm electrode gap space. Also, the effect of the catalytic properties of titanium dioxide nanoparticles on the treatment process of the PCD reactor was investigated. Different concentrations of TiO_2 NPs (0.075, 0.3, 0.08 and 3 gL^{-1}) were tested. A concentration of 0.08 gL^{-1} TiO_2 was found to be the optimal concentration that increased the dye degradation from 52.62% (with no titanium dioxide nano particles) to 94.14% in 15 min. However, a higher concentration of TiO_2 showed the adverse effect and decreased the degradation level of AB1. Finally, the pulsed corona discharge as an oxidation process was compared with the photocatalytic processes of UV, UV/H_2O_2 and UV/TiO_2 in terms of removing AB1 from an aqueous solution, and promising results were reported.

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1. Introduction

Synthetic dyes are commonly used as a source of coloring agents in textile, leather, dyeing, cosmetics, paper and food-processing industries due to their resistance to biodegradation and color flexibility. Azo dyes, characterized by having the azo group, and consisting of two nitrogen atoms (—N=N—) as the chromophore in their molecular structure, represent about 70% of all dyes used in the industrial applications [1]. Azo dyes have advanced thermal and optical properties due to their chemical nature, molecular size, and structure. The discharge of dye-containing effluents into water bodies cause serious environmental and health problems. The effluents are toxic and carcinogenic with a complex structure mainly based on aromatic amines that also generate by-products of their partial degradation [1]. AB1 (also called Amido Black10B, or Naphtol Blue Black) is one of the azo dyes that is widely used in industries [2].

Different techniques (e.g., coagulation, electro coagulation and flocculation [3,4], ion exchange [5], membrane separation [6,7], biosorption, bioaccumulation [1,8] and oxidation [9]) have been used with varying degrees of success for the removal of dyes and other contaminants from water and wastewater. Major disadvantages of these methods, especially for dye removal, include partial degradation, generation of toxic sludge, high cost and technical limitations [10]. Therefore, there is a critical need to develop methods to efficiently treat dye-containing wastewaters before discharging them into the environment. Recently, advanced oxidation processes (AOPs) have attracted interest as efficient methods for removing organic dyes from wastewater. The main mechanism of advanced oxidation processes is an in situ generation of very strong oxidizing agents (e.g., $OH^{\bullet}$, $H^{\bullet}$, $O^{\bullet}$, H_2O_2, H_2, O_2, O_3) as well as chemical and physical processes that provide subsequent reaction with the pollutants, resulting in the decomposition of dissolved molecules in water via oxidation reactions [11]. This process reduces the target compounds' toxicity or increases their biodegradability. The common point in most advanced oxidation processes is the production of hydroxyl radicals ($OH^{\bullet}$) intermediates, which are highly reactive and non-selective oxidants with oxidation potential (E_0) of 2.8 V that can initiate oxidation

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Table 1
Oxidizing potentials of reactive species at 25 °C.

Oxidants	E_0: electrochemical potential (V)
Chlorine	1.36
Hydrogen peroxide (H_2O_2)	1.78
Ozone (O_3)	2.08
Atomic oxygen (O)	2.42
Hydroxyl radical ($OH^\bullet$)	2.80
Fluorine	3.06
TiO_2^+	3.20

processes and oxidize the aromatic ring and double-bound structures of contaminants [12]. The reaction rate constants between hydroxyl radicals and organic species are in the range of 10^8–$10^{10}\,M^{-1}\,s^{-1}$ [13]. Oxidizing potentials of reactive species commonly used for wastewater treatment at 25 °C is shown in Table 1. Ono and Oda [14] reported that high-voltage electric discharges in water by a pulsed streamer corona cause a formation of ozone (O_3), a Hydroxyl radical ($OH^\bullet$) and a hydrogen radical ($H^\bullet$) from oxygen and water as follows:

$$\text{Initiation and Propagation}: e^- + H_2O \rightarrow e^- + H^\bullet + OH^\bullet \quad (1)$$

$$\text{Ionization}: e^- + 3O_2 \rightarrow e^- + 2O_3 \quad (2)$$

$$\text{Streamer Propagation}: O + H_2O \rightarrow 2OH \quad (3)$$

The dissolved ozone (O_3) in water can in turn decompose to form hydroxyl radicals ($OH^\bullet$) or react directly with the pollutants. Major active species involved in the degradation of organic pollutants using a pulsed streamer corona discharge are hydroxyl radicals ($OH^\bullet$) and a hydrogen peroxide (H_2O_2) [14].

The application of pulsed electrical discharges has proven to be efficient for degrading organic compounds in aqueous solutions [14]. One of the important factors that determines the efficiency of pulsed electrical discharge processes is its electrodes configuration. Previous works used different electrode configurations such as the Ring-to-Cylinder [15]. Point-to-Plane [16], Needle to Plate [17], Wire-to-Plate [18] and Point-to-Point electrodes [19] to remove bacteria, dyes, and other organic contaminants from aqueous solutions. The Point-to-Point electrode configuration has been used for removing bacteria and complex organic materials such as proteins from water [20]; however, to the best of our knowledge, no literature has reported that this electrode has been used to remove dye from water.Several studies have shown that AOPs such as UV/O_3 [21], UV/H_2O_2 [22], sonolysis in the presence of catalysts [23], pulsed-electrical discharge technology [24] and photocatalysis such as UV/TiO_2 [25] can be effective for the removal of dyes from aqueous solutions. In UV/H_2O_2 process, Hydroxyl radicals ($OH^\bullet$) can be formed mainly when H_2O_2 is photo-dissociated by UV radiation in the range of 200–280 nm. On the TiO_2 surface, photogenerated electrons and holes produce various active oxygen species such as O_2^-, $OH^\bullet$, $^\bullet HO_2$ and $O^\bullet$ [26]. Shu et al. [27] demonstrated the complete decolorization of azo dye Acid Black 1 (AB1) by the UV/H_2O_2 processes. They showed that AB1 solution can be completely decolorized under the optimal hydrogen peroxide dosage of 21.24 mmol L^{-1} and UV dosage of 1400 W/l in less than 1.2 min.

In this research, first the application of pulsed field electrical discharge (i.e., pulsed corona discharge) as one type of AOP on the removal of Acid Black 1 from water was studied at different electrode configurations and electrical field frequency. Then, the application of pulsed corona discharge was compared with other types of AOPs like photocatalytic processes including (a) ultraviolet radiation (UV), (b) ultraviolet radiation (UV) with the addition of hydrogen peroxide (H_2O_2), and (c) ultraviolet radiation (UV) with the addition of TiO_2 for the removal of AB1 from water.

2. Materials and methods

The commercial AB1 (80% of dye) product was purchased from Sigma–Aldrich (Sigma Chemical Co., USA) and used without further purification. A 10 mg/L stock solution of AB1 representing the synthetic organic azo dyes was prepared by dissolving the required amount of analytical grade dye in deionized water without further pH adjustment. The initial pH of the solution (pH 5.2) was measured by using a Fisher scientific accumet® model 15 pH meter according to standard laboratory operating procedures. Hydrogen peroxide (H_2O_2), 30% (wt.%) (Certified ACS) was purchased from Fisher Scientific Inc (Pittsburgh, PA, USA). The concentration of H_2O_2 was chosen following previous studies [27]. Anatase nanopowder titanium dioxide 99% with 10 nm size was purchased from US Research Nanomaterials Inc., (Houston, TX, USA) and was used without further purification.

The generation of ozone was determined by the colorimetric indigo dye method [28]. All concentrations were measured by using UV–vis spectroscopy (Varian Cary 3E, Agilent, Santa Clara, CA, USA) at the wavelength of 618 nm. The 618 nm wavelength was reported as the maximum absorbance wavelength of AB1 [29]. Specific UV absorbance can be used to monitor the change in aromatic nature of organic compounds [29]. Prior to the measurement of AB1 absorbance at different time intervals, a calibration curve was obtained by using the standard AB1 solution with the known concentrations. The samples were taken out by a pipette and the absorbance measurements were finished within less than 1 min because the reaction was likely to continue after sampling and possibly change the composition [29].

The degradation (decolorization) $\eta(\%)$ was calculated as [30]:

$$\text{Degradation}: \eta(\%) = \frac{C_0 - C}{C_0} \times 100$$

Here, C_0 and C are the initial and the final concentrations of AB1 solution, respectively.

The treated samples were taken from the reactor at different time intervals and they were filtered by Millex-HV 0.45 μm PVDF filter (millipore) purchased from Fisher Scientific Inc. (Pittsburgh, PA, USA). The spectroscopic monitoring and Dissolved Organic Carbon (DOC) measurements were done by subtracting the remaining DOC_t from the initial DOC_0 [31]. This was prepared in order to investigate the occurrence of mineralization (See, below equation) and the changes in aromatic nature of AB1 in both pulsed corona discharge and UV- photocatalysis.

$$\text{Mineralization Efficiency}(\%) = \left[1 - \left[\frac{DOC_t}{DOC_0}\right]\right] \times 100$$

3. Experimental set-up

3.1. Pulsed corona discharge (PCD) reactor

In this study, a reactor suitable for batch-scale non-thermal pulsed corona streamer discharge (voltage 15 Kv) provided by Scientific Utilization Inc. (Huntsville, AL, USA) with point to point electrodes configuration, was used to investigate the removal of AB1 from water (see Fig. 1). The reactor was able to produce an electrical discharge in aqueous solution as current was applied across cylindrical electrodes (9.5 mm in diameter). The cathode electrode was a copper composite with an air-feed connection and the anode electrode was composed of stainless steel.

The reactor had the water flow rate of 4.7 (liter/min) and air bubbles with a pressure of 0.5 (psi). The air release was included in the design to avoid the pump head cavitation and gas accumulation in the system and also to enhance the degradation efficiency (%) [32]. A Roto-Flow TM 1K081B stainless-steel pump with a Fluid-O-