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Cold atmospheric plasma discharge induced fast decontamination of a wide range of organic compounds suitable for environmental applications

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ABSTRACT

A fast and facile approach for decomposition of model organic dyes through high voltage corona discharge in solution was presented. A comparative study of the significant potential of this technique for degradation of 8 different types of standard organic compounds such as Phenol Red (PR), Methyl Orange (MO), Diamine Green B (DGB), Pyrogallol Red (PYR), Bromocresol Green (BCG), Bromochlorophenol Blue (BCB), Naphtol Green B (NGB) and Crystal Violet (CV) has been performed. The decomposition rate of dyes under corona discharge was evaluated by measuring the changes in absorption peak of model dye at different discharge times using ultraviolet–visible absorption spectroscopy (UV–vis). The results revealed that corona discharge at ambient air significantly enhanced the degradation rate of standard dyes. In addition, a possible mechanism for degradation of dye molecules under corona discharge process was proposed based on our observations. It has been found that this process, which is based on the most efficient energetic species, is much more powerful than other conventional methods such as photocatalytic materials.

1. Introduction

The environmental problems created by the pigment, dyes and colorants are rising in the industrial and household wastewater as a result of their extensive use and relative stability toward degradation [1]. These dyes, (especially textile dyes) are engineered to be resistant to all kinds of treatments without fading. They need to sustain either alkaline or acidic environment; they need to withstand washing with soaps and bleaching agents, microbiological fading, be resistant to light and ultraviolet irradiation etc. [2]. Obviously, the better stability of dyes achieved in the consumer products where the worse problem they cause in the wastewater stream when subjected to decolorization or degradation [3].

Due to the worldwide concern about the treatment of wastewaters a large number of investigations are going on to find more efficient procedures for the treatment of wastewaters [4,5]. Generally there are various physical and chemical routes for dye wastewaters treatments such as adsorption on inorganic or organic matrices, decomposition by photocatalytic materials, advanced oxidation processes (AOPs), microbiological or enzymatic decomposition, etc. [6–12]. Due to the inherent drawbacks of physical, chemical and photochemical approaches to dye removal, the use of corona discharge method for the treatment of textile wastewaters has received much attention as a more cost effective alternative [13–17].

Studies for treatment of organic dyes are largely centered on the development of a fast, efficient and cost effective removal processes. Li et al., have used high-voltage corona discharges in atmospheric air for bacterial inactivation and shown that the chemical and biological effects had a potential correlation with specific energies of corona treatment and ozone produced by corona discharges [18]. Similarly Birmingham and Hammerstrom showed that atmospheric pressure nonthermal plasma can efficiently deactivate bacteria in gases, liquids, surfaces and also can decompose hazardous chemicals [19].

In the present work, the effect of atmospheric corona discharge on degradation of several model dyes was studied. In addition, the effect of discharge duration on decomposition rate of standard dyes were evaluated and compared.

2. Experimental details

A Tesla coil composed of several distinct parts was used to create a corona discharge. A high voltage supply provided by transformer

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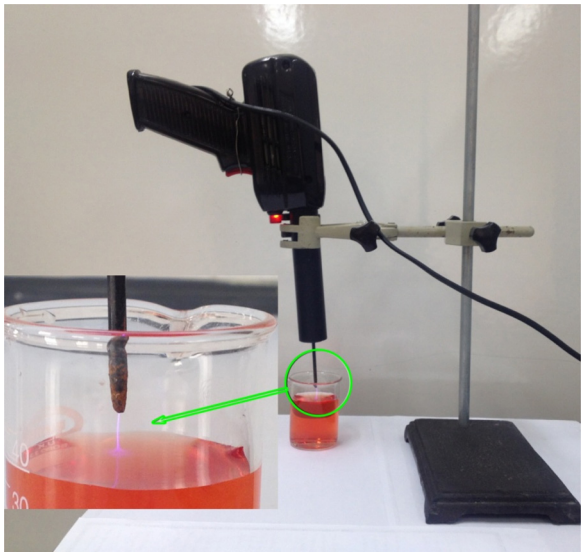

Fig. 1. Experimental set up showing the corona discharge process.

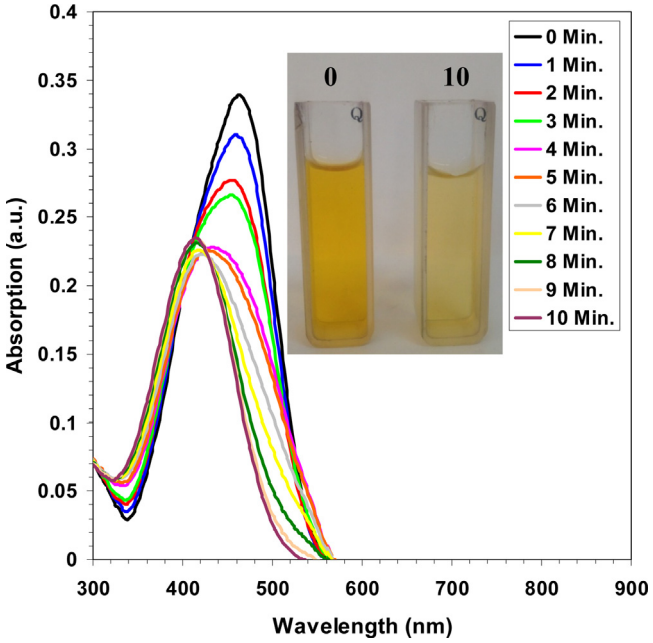

Fig. 2. Degradation rate of MO at different discharge durations from 1 to 10 min.

was used to generate the spark gap. Transformers charge the high voltage capacitors to fire the spark gap. Then energy in the primary coil is transferred into the secondary coil by a magnetic coupler where results in an extremely high voltage at the top of the secondary coil which is the last place for the electricity before it jumps into the air. Fig. 1 shows the experimental set up used in this work. Corona discharge instrument works with AC 220–240 V and 50–60 Hz. This instrument has a sharp electrode which allows electrical field to concentrate around the electrode. The parameters of corona discharge were as follows: voltage: 360 V, current: 0.5 A, pressure: 760 Torr, working gas: air and temperature: 25 °C. The facility has a sharp electrode at the end for boosting the electrical field. In this situation the electrical field is concentrated around the electrode. Inset of Fig. 1 shows a typical corona discharge for a PYR dye. The primary concentration of all model dyes was the same (10^{-4} M). In this concentration of dye, all dye powder easily dissolves in DI water without any aggregation and the solution remains stable for a couple of months. The most of dye molecules used in this research have chromophore structures with alternating double and single bonds such as C=C and C=O (carbonyl), as well as azo group —N=N— or nitro group ($-NO_2$). The detailed structure of each dye and the corresponding chromophores are presented on Fig. S1–S8 in Supplementary information (SI).

2.1. Material characterization

Degradation rate of all organic dyes was performed using a double beam Optizen POP spectrophotometer (Mecasys Company, Korea) in the range of 200–900 nm wavelengths by measuring the maximum absorption peak of individual model dyes at different discharge duration times. DI water was supplied from a Millipore water purification system (Direct-Q-3). The conductivity of all solution was almost the same and equal to 0.055 μs.

3. Results and discussion

3.1. Crystal structure

Dyes are aromatic compounds which have the ability to absorb light in the visible wavelengths ranging from 400 to 700 nm. The colors of dyes and pigments are due to the absorption of visible light by the compounds, which involves aryl rings and have delocalised electron systems. The dye molecules have a chromophore group which is responsible for absorption of light. In addition to chromophores, most dyes also contain groups known as auxochromes (color helpers) such as carboxylic acid, sulfonic acid, amino, and hydroxyl groups. While these are not responsible for color, their presence can shift the color of a colorant and they are most often used to influence dye solubility [20,21].

Fig. 2 shows the degradation rate of MO at different discharge durations from 1 to 10 min. As is clear from the figure the maximum absorption peak of MO (located around 465 nm) is decreasing due to exposure to corona discharge process. This obviously shows the ability of corona discharge in decomposition of this model dye at ambient conditions. The inset of Fig. 2 is the optical image of samples at the beginning and end of discharge process. The untreated MO sample has a clear orange color while the treated sample shows a very pale orange color. In fact, the position and shape of the absorption band affect the appearance of the observed color of dyes. Many compounds absorb light in the ultraviolet (UV) region, with some absorptions extending into the violet (400–430 nm) region. Thus, these compounds appear yellowish to the eye—i.e., the perceived color is complementary to the absorbed color. Progressive absorption into the visible region gives orange (430–480 nm), red (480–550 nm), violet (550–600 nm), and blue (600–700 nm); absorption at 400–450 and 580–700 nm gives green. In this research a wide range of dye molecules with maximum absorption peak ranging from 430 to 720 nm was chosen in order to evaluate the capability of this technique for decontamination and dye removal [22,23].

Changes in absorption spectrum of NGB are depicted on Fig. 3. The degradation rate of NGB under corona discharge is more remarkable than MO and after about 10 min the absorption value almost reaches to zero. Moreover, optical image of the sample at the beginning and end of corona discharge process clearly verify degradation of this model dye.

Changes in absorption spectrum of DGB are also illustrated in Fig. 4. Compared with NGB and MO, the degradation efficiency seems to be less in the case of DGB. In fact, the maximum changes in abortion peak of DGB are noticeably smaller than NGB and MO. This difference is clearly observable in the optical image of the mentioned dye at the beginning and end of discharge process (inset of Fig. 3) which may be related to the differences in the chemical sta-