



ORIGINAL ARTICLE

Plasma treatment by gliding arc discharge of dyes/ dye mixtures in the presence of inorganic salts



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Abstract The gliding arc discharge plasma (GAD) technique was used for discoloration and degradation of an anthraquinone dye Alizarin red S (ARS), an azo dye Orange G (OG) and their mixture. The influence of chloride and sulfate salts on the efficiency of treatment has also been studied. The aim of this study is the discoloration and degradation by GAD of ARS, and OG and their mixture.

The GAD plasma generated between two moist air metallic electrodes connected to a high voltage produced chemical species and highly reactive molecules $\cdot\text{OH}$, $\text{NO}\cdot$, $\text{O}\cdot$, O_2 , $\text{HO}_2\cdot$, $\text{H}\cdot$, H_2O_2 , O_3 ... and other excited-state neutral molecules. The reactions of discoloration for ARS and OG followed pseudo-first-order kinetics. The apparent kinetic constants of bleaching were found equal to 0.014 and 0.013 min^{-1} and degradation of 0.008 and 0.010 min^{-1} , respectively for the ARS and OG. For mixtures, the rate of discoloration was 80% in 120 min of treatment for mixtures III, IV and V, while the rate of COD reduction was fast in the first 30 min since it varied between 35% and 40% for all mixtures but stabilized at 68% toward the end of the treatment. The addition of chloride salts and sulfate showed a decrease in the effectiveness of treatment.

GAD treatment is perfectly suited to dyes treatment. It is an alternative method that is part of green chemistry and sustainable development objectives.

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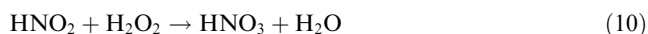
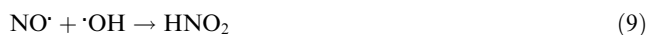
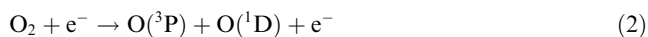


1. Introduction

Anthraquinone and azo dyes, representing two classes of dyes commonly used in the textile industry, have bio-refractory and carcinogenic characters (Bouzaida et al., 2004; Dai et al., 1995). Their treatment before release into the wild is necessary. The organic and conventional treatments (coagulation/flocculation, adsorption and biodegradation, activated carbon, reverse osmosis and ultrafiltration, ... (Vandevivere et al., 1998) became ineffective due to high number of benzene cycles generated, high stability and resistance of new output molecules (Konstantinou and Albanis, 2004). Effluents containing these types of dyes are characterized by a strong color, highly fluctuating pH, high chemical oxygen demand (COD) and biological oxygen demand (BOD) and severe biotoxicity (Shen et al., 2001; Cisneros et al., 2002a,b; Karkmaz et al., 2004). The dye's strong color causes considerable damage to the effluent (Mozia et al., 2005). Inorganic salt presence makes the coloration important and the effluent rich and stable.

In recent years, advanced oxidation processes (AOPs) that generate hydroxyl radicals $\cdot\text{OH}$ in sufficient quantity are more efficient in water treatment. Common AOPs, including application of technologies, such as Fenton reaction (Alshamsi et al., 2007), photo-Fenton process, electro-Fenton process, ozonation, photo catalysis, non-thermal plasma, irradiation by ultrasonic (Chacon et al., 2006; Panizza and Cerisola, 2009; Uilson de Souza et al., 2010; Bansal et al., 2010; Abdelmalek et al., 2004; Onat et al., 2010; Sayan, 2006), are all applied to investigate the degradation of the anthraquinonic and azo dyes. Gliding arc discharge, an initially proposed tool by Czenichovski (1994) has been found especially suitable for degrading dyes, which are the most-used textile colorants and were generalized by other authors (Abdelmalek et al., 2006; Doubla et al., 2008). This technique produces non-thermal plasma at atmospheric pressure generated by an electric arc supplied by a plasma gas, generally air. Treatment effectiveness is due to the formation of chemical species, neutral molecules and molecules in excited states such as highly reactive $\cdot\text{OH}$, $\text{NO}\cdot$, $\text{O}\cdot$, $\text{O}_2\cdot$, $\text{HO}_2\cdot$, $\text{H}\cdot$, H_2O_2 , O_3 ... The most reactive species is the hydroxyl radical, which has an oxidation potential E° ($\cdot\text{OH}/\text{H}_2\text{O}$) = 2.8 V vs. Normal hydrogen electrode (NHE) can attack the most part of organic molecules with rate constants usually in the order of 10^6 – 10^9 $\text{M}^{-1} \text{s}^{-1}$ (Burlica et al., 2004; Abdelmalek et al., 2008; Andreozzi et al., 1999).

The probable reactions mechanisms taking place are listed as follows:



Previous emission spectroscopy study carried out on glidarc humid air plasma confirmed the simultaneous presence of $\cdot\text{OH}$ and $\text{NO}\cdot$ radicals (Benstaali et al., 2002).

Therefore, the aim of this study is to use the same technique and investigate the discoloration and degradation by gliding arc discharge (GAD) produced reactive species on aqueous solutions of an anthraquinone dye, Alizarin red S (ARS), an azo dye Orange G (OG) and their mixture in different proportions. The study also focused on the influence of chlorides and sulfates often present in textile wastewaters. Both dyes are extensively used in the textile industry and are considered mutagenic poisons, suspected of being carcinogenic (Zucca et al., 2008; Lachheb et al., 2002; Xu and Li, 2010).

The advantage of the GAD technique we are using is that it is very efficient, generates reactive $\cdot\text{OH}$ radicals able to degrade highly resistant pollutants and does not require additional chemicals or catalysts which can be seen as a clean technology.

2. Materials and methods

2.1. Materials and reagents

Alizarin red S (ARS), formulae $\text{C}_{14}\text{H}_7\text{NaO}_7\text{S}$ (9,10 dihydroxy-9,10 dioxo-2 anthracenesulfonic acid) and azo dye Orange G (OG), $\text{C}_{16}\text{H}_{10}\text{N}_2\text{Na}_2\text{O}_7\text{S}_2$ (7 hydroxy-8 phenyl azo-7,3-naphthalenedisulfonic acid), were purchased from Acro Organics. The chemical structure and absorption spectra of ARS, OG and their mixtures of different composition are shown in Fig. 1a–c. All solutions were made from analytical-grade chemicals. Distilled water was used to make the dye solutions of desired concentration. The pH of the aqueous dye solution is 6.5 for ARS and OG.

2.2. Plasma-producing apparatus

The experimental apparatus of the glidarc plasma used is shown in Fig. 2. Compressed gas is led through a bubbling water flask to get water-saturated. The gas flow then is driven between two semi elliptic electrodes connected to a 220 V/10 kV high voltage Aupem Sefli transformer. It produces an alternative potential difference of 10 kV and a current intensity of 100 mA, the power used was 1 kW, with a very interesting energy cost. An electric arc formed between two diverging electrodes raised to a convenient voltage difference at the minimum gap. The arc is pushed away from the ignition point by the feeding gas flow, sweeps along the maximum length of the electrode gap and forms a relatively large non thermal plasma zone. A new arc then appears and develops according to the same procedure. The resulting plasma is actually quenched at atmospheric pressure and quasi-ambient temperature. The diffusion process in the liquid is improved by conversion in the liquid phase due to the airflow and magnetic stirring.

A 160 mL solution is placed into the 500 mL Pyrex closed reactor equipped with a cooling system to avoid evaporation. The treatment is done with predetermined functioning parameters. The gas flow is fixed at: $Q = 800$ L/h, the minimum gap between the electrodes: $e = 2$ mm, the diameter of the nozzle: $\Phi = 1$ mm and the distance between the electrodes and the target liquid surface: $d = 3$ cm. The solution is permanently magnetically stirred and the temperature is maintained at 20 ± 2 °C in the cryostat.

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