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Research article

A novel two-level dielectric barrier discharge reactor for methyl orange degradation

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ABSTRACT

A novel pilot two-level dielectric barrier discharge (DBD) reactor has been proposed and applied for degradation of continuous model wastewater. The two-level DBD reactor was skillfully realized with high space utilization efficiency and large contact area between plasma and wastewater. Various conditions such as applied voltage, initial concentration and initial pH value on methyl orange (MO) model wastewater degradation were investigated. The results showed that the appropriate applied voltage was 13.4 kV; low initial concentration and low initial pH value were conducive for MO degradation. The percentage removal of 4 L MO with concentration of 80 mg/L reached 94.1% after plasma treatment for 80min. Based on ultraviolet spectrum (UV), Infrared spectrum (IR), liquid chromatography–mass spectrometry (LC–MS) analysis of degradation intermediates and products, insights in the degradation pathway of MO were proposed.

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1. Introduction

Water pollution is facing grave challenges in modern society. With rapid development of printing and dyeing industry, chemical industry, as well as petroleum industry, large amount of low-degradable organic dye wastewater are discharged, which causes a serious environmental problem. The prominent problems for dyeing wastewater treatment are color removal and degradation of organic compounds (Basha et al., 2012; Martínez-Huitle and Brillas, 2009). Various methods such as physical methods (Ong et al., 2014; Sojka-Ledakowicz et al., 2010), chemical methods (Liu et al., 2013; Patil et al., 2011), biological methods (Isik and Sponza, 2006; Kannan and Sumeet, 2012) and combined methods (Qi et al., 2011; Ou et al., 2015; Shin et al., 2014) have been proposed. However, physical and chemical methods only transfer the pollutants to another phase, which usually induce by-products formation. Biological methods are not effective for the highly colored and low-degradable dyeing wastewater. Therefore, the development of environmentally friendly and efficient methods for wastewater treatment receives great attention.

Plasma oxidation process, with high efficiency and without secondary pollution, is one of the most promising methods for dye wastewater treatment. Various types, such as high-voltage pulsed corona discharge (Wang et al., 2006; Zheng et al., 2012), glow discharge (Ke et al., 2011; Wang et al., 2012a, 2012b), dielectric barrier discharge (DBD) (Reddy et al., 2013; Kim et al., 2013; Li et al., 2014; Liu et al., 2012a, 2012b; Magureanu et al., 2010; Hijosa et al., 2013; Tang et al., 2012; Tichonovas et al., 2013) and gliding arc discharge (Abdelmalek et al., 2005; Merouani et al., 2015; Liu et al., 2012a, 2012b) have been applied for wastewater treatment. Among them, DBD is a commonly used method, with large amount of energetic electrons, UV radiation, OH radicals, O radicals, as well as O_3 and H_2O_2 molecules. Different DBD reactor configurations and the system of organic pollutants were investigated. In recent years, the conventional batch DBD reactors with different electrode configuration (Reddy et al., 2013; Kim et al., 2013; Li et al., 2014) were proposed for various wastewater treatments. Kim et al. (2013) developed a DBD reactor for degradation of veterinary antibiotics, and estimated the relationship between the degradation efficiency and the energy requirements on the basis of an initial concentration of 5 mg/L. Li et al. (2014) investigated acetamiprid removal in wastewater by DBD with the synergistic effect of reactive species and UV/visible lights. The degradation efficiency was 83.48% at 200 min when the discharge power was 170 W and the initial acetamiprid concentration was 50 mg/L. Besides the conventional batch reactors, *ex situ* and *in situ* discharge (Liu et al., 2012a, 2012b),

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as well as coaxial water falling film reactors (Magureanu et al., 2010; Hijosa et al., 2013) were proposed. Liu et al. (2012a, 2012b) compared *ex situ* and *in situ* discharge for the degradation of carbamazepine (CBZ). The results showed that *ex situ* discharge system was more effective than the *in situ* system. Tichonovas et al. (2013) proposed a pilot DBD semi-continuously operated plasma reactor. However, most existing reactors, were applied for low concentration wastewater treatment with a small capacity like 100 or 500 mL, which were difficult to satisfy industrial continuous wastewater treatment requirement.

In the present study, a novel two-level dielectric barrier discharge reactor was designed which could continuously degrade wastewater with high concentration and large processing capacity. The viability of the novel two-level dielectric barrier discharge reactor was tested during the degradation of MO dyeing wastewater. The degradation mechanism and the influence factors that affect the degradation of MO dyeing wastewater in the DBD reactor had been also researched.

2. Experimental part

The schematic diagram of the two-level DBD reactor was shown in Fig. 1(a). The DBD plasma reactor, which was operated at atmospheric pressure, comprised two coaxial quartz tubes, two stainless steel disk electrodes and one stainless steel mesh electrode. Wastewater flowed up through the inner quartz tube towards the top of the tube, then falling down on the outside surface of the tube, making a water film. When the voltage was applied between two stainless steel disk electrodes, wastewater elongated the stainless steel disk electrode in the water to the water surface in the inner quartz tube because of its electrical conductivity, so discharge was generated between the stainless steel disk electrode cover with dielectric barrier above the water and the water surface in the inner quartz tube. The gas-phase reactive species formed during the discharge and transferred into the wastewater, so that the wastewater could be degraded to some extent. Similarly, when the voltage was applied between the stainless steel disk electrode in the water and the stainless steel mesh electrode wrapped at the outside of the outer quartz tube, water in the inner quartz tube served as the ground electrode because of its electrical conductivity. Discharge was formed in the annular region between the two coaxial quartz tubes. That was to say, the water film was in direct contact with the discharge where two coaxial quartz tubes represented the dielectric barrier. Therefore, two-level continuous discharges (shown in Fig. 1(b)) generated with high space utilization efficiency and large contact area between the plasma and the wastewater. In reactor, the diameter of the stainless steel disk electrode was 50 mm and the discharge gap of the first level was

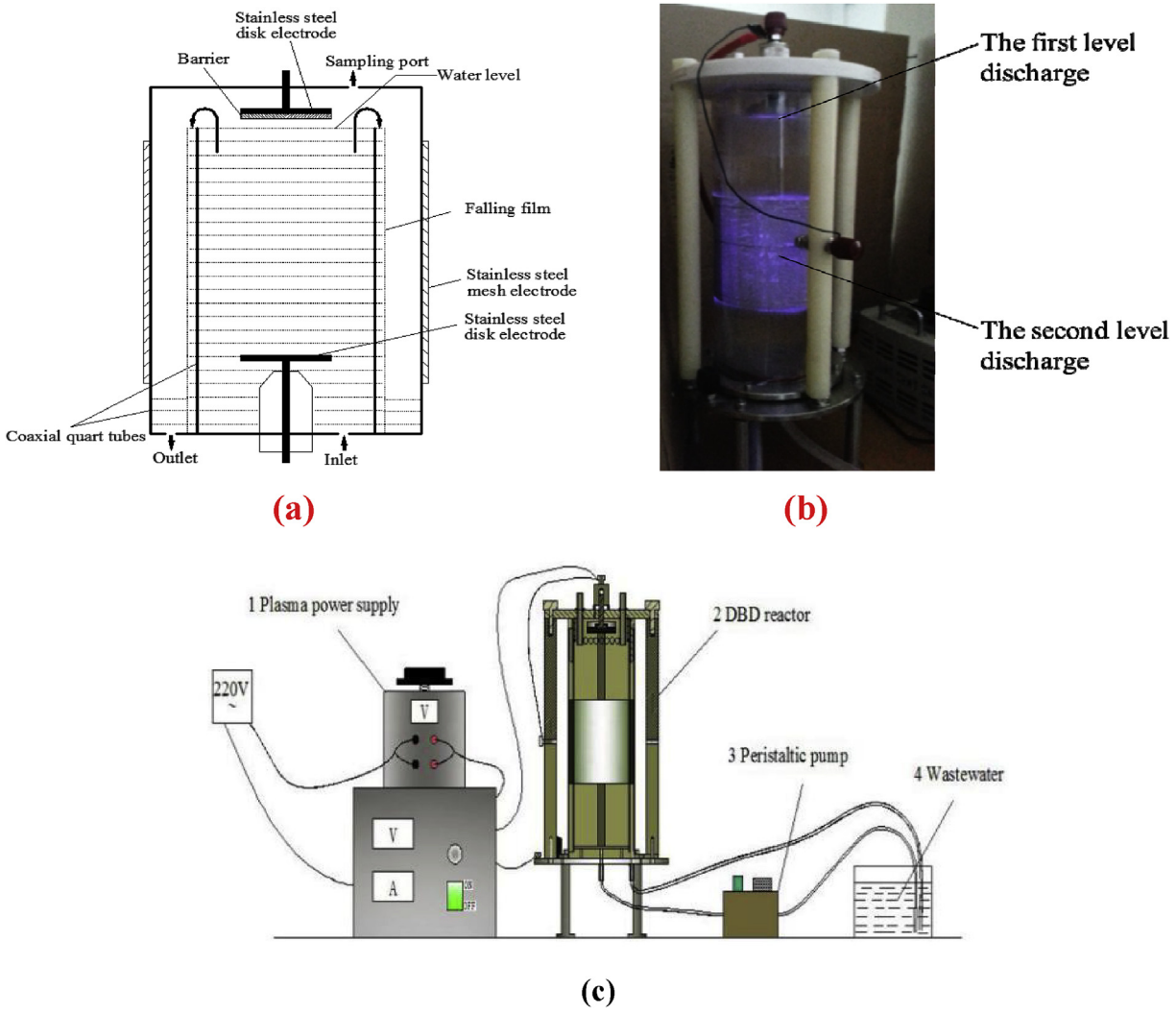

Fig. 1. Schematic (a) and actual diagram (b) of the two-level DBD reactor, processing flow chart (c) of wastewater.

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