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Degradation and mechanism analysis of bisphenol A in aqueous solutions by pulsed discharge plasma combined with activated carbon

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ABSTRACT

Degradation of bisphenol A (BPA), an industrial chemical and suspected endocrine disruptor, in a pulsed discharge plasma (PDP) system combined with activated carbon (AC) was investigated in this study. The experimental results showed that the degradation rate of BPA could reach 95.3% after 60 min discharge plasma treatment at 20 kV discharge voltage and the degradation process fitted the first-order kinetic model; degradation rate of BPA was higher in acid solution than that in neutral or alkaline solution; compared to air and nitrogen, the degradation rate of BPA was highest when oxygen was bubbled into the reaction system; the results of spectrum detection indicated the synergistic effect of PDP and AC and the important role of ·OH and ·O for the degradation of BPA; the removal of TOC could be 36.8% at the optimal condition; longer discharge plasma treatment time resulted in lower pH value and higher conductivity of treated solution; the increase of absorbance at 240–260 nm in UV–Vis spectra of the BPA solution illustrated the production of intermediates during the degradation process; the degradation pathway was proposed based on the research results and references. Scanning electron microscope (SEM) analysis showed that the surface of the AC after PDP treatment was smoother and AC could be generated by pulse discharge under a certain condition.

1. Introduction

Bisphenol A (BPA) is a kind of typical endocrine disruptor with estrogenic activity and can carry a high health risk even at the low dose of 0.05 mg per kg body weight [1,2]. As an important derivative of phenol and acetone, BPA has been widely used for the production of polycarbonate plastic, epoxy resin and flame retardant. However, the BPA can be transferred from the products to environment and then dispersed in water, which can inevitably cause damage to human beings. Many studies have shown that BPA has certain embryotoxicity and teratogenicity which can raise the risk of ovarian cancer, prostate cancer, asthma and leukemia [3–5]. In addition, BPA can damage liver and pancreatic β cell, which can lead to the disorder of thyroxine and fat [6]. The effect of BPA on human body is cumulative and irreversible. It is therefore imperative to remove BAP in aqueous solution.

In recent years, many methods have been employed to remove BPA, including ozone oxidation [7], ultrasonic radiation [8], Fenton oxidation [9], photocatalysis oxidation [10], electrochemical oxidation [11]. However, there are some shortcomings for these methods. For example, the equipment and running cost of ozone oxidation is high and the utilization of ozone is lower. The mineralization of BPA by ultrasonic radiation is not complete and the treatment cost is higher. Besides, the reaction condition is limited by the operating parameters, dissolved gas and co-existing effect, which can account for the strictly reaction condition of ultrasonic radiation. Fenton oxidation for BPA degradation is dependent on the solution pH, higher solution pH is not beneficial to generate ·OH and the required pH value is less than 3.5 generally. In addition, the redundant of ferrous ion in the reaction system can increase chemical oxygen demand (COD) of the treated water, which can lead to secondary water pollution. For the degradation of BPA in a photocatalytic system, it is difficult to recycle the photocatalyst and the utilization of light is lower. The energy consumption of electrochemical oxidation is relative higher with a higher cost. Besides, the hydrogen evolution reaction and the hydrogen evolution side reaction can be introduced during degradation process, which can result in the electrodes corrosion. Therefore, it is significant to develop some new technologies for BPA removal in water.

Technology of pulsed discharge plasma (PDP) is a kind of advanced oxidation technologies (AOTs). PDP integrates electron, ion, neutral compounds and free radical [12–16]. Therefore, various physical and chemical effects existing simultaneously to oxidize organic compounds directly or indirectly in a PDP system. Studies have shown that kinds of

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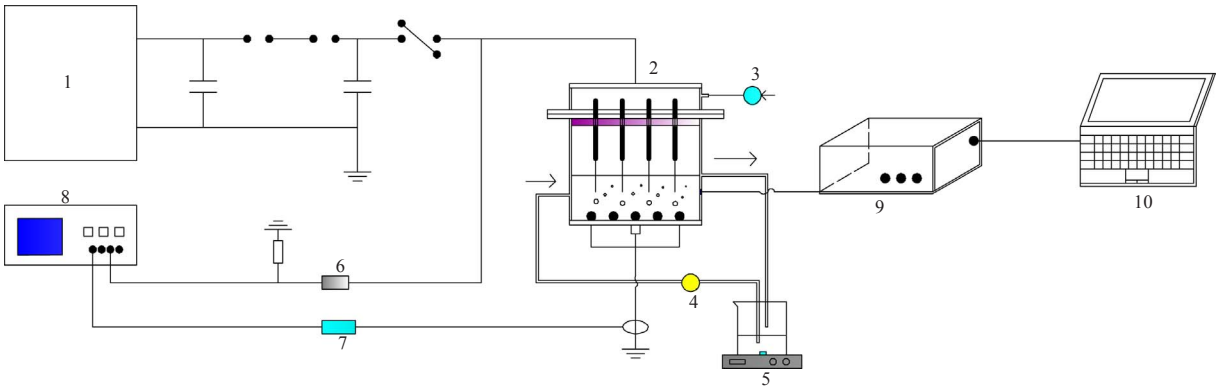

Fig. 1. Schematic diagram of the experimental apparatus: 1. DC power, 2. Reactor, 3. Gas flow meter, 4. Peristaltic pump, 5. Stirrer, 6. High-voltage probe, 7. Current probe, 8. Oscilloscope, 9. Spectrograph, 10. laptop.

organic compounds in aqueous solution, such as phenols, p-chlorophenol, sulfate and dyes can be removed by the PDP effectively [17–20]. However, there are some problems restricting the application of the technology. For example, active species generated in the discharge process could not be fully utilized in the degradation process. Furthermore, ultraviolet light or other physical effects produced in the discharge channel have not been efficiently used, which decrease the energy utilization of the oxidation process. Hence, research on the synergistic process of PDP with other technologies for pollutants degradation has become a hot topic. Activated carbon (AC) has been widely used as an adsorbent for wastewater treatment due to its extended large specific surface area and high pore structure [16,21,22]. Studies have reported that AC can accelerate ozone decomposition and induce the formation of ˙OH in the oxidation system [23,24]. Grymonpré is the first research who introduced AC into a PDP system and the result improved that the overall removal of phenol could be improved by the addition of AC [25,26]. Subsequently, many groups have focused on the research of the synergistic behavior of PDP/AC for organic compounds removal. Zhang et al. and Hao et al. have investigated the catalysis of granular AC in the non equilibrium plasma water treatment reactor [24,27] and reported that the removal of organic compounds could be increased significantly in the presence of AC due to the adsorption and catalysis initiated by the basic functional groups on the granular AC surface and the higher specific surface area of the AC. Zhang et al. and Jiang et al. combined discharge plasma with AC fiber and verified the more advantages of the combined processes for pollutant abatement [28,29]. Recently, some other carbon materials, such as multiwalled carbon nanotubes and mesoporous carbon, were also applied in a discharge plasma system and proved that the performance of discharge plasma was significantly improved due to the more ˙OH formation in the synergistic system [30,31].

Although researches have shown that discharge plasma combined with AC can further enhance degradation of organic compounds, the degradation process of organic compounds by the synergistic effect has not been clarified sufficiently. Furthermore, most researches have focused on the organic compounds degradation and surface properties of AC. The effect of AC addition on the formation of ˙OH, however, has not sufficiently studied. In addition, other active species (such as ˙O, and H_2O_2,), which are playing major role on organic compounds degradation, have not been investigated thoroughly. It is therefore necessary to study the amount of these active species before and after AC addition in the discharge plasma system, and then to verify the synergistic effect of plasma and AC.

In this study, a combined system of PDP (multi-needle-to-plate electrode geometry) and AC was set up to investigate the degradation of BPA in aqueous solutions in the synergistic system. The degradation of BPA was investigated by varying experimental conditions of the discharge voltage, the initial solution pH value and the gas bubbling varieties. The relative emission spectrum intensities of ˙OH and ˙O was detected to illustrate the synergistic effect of PDP and AC and the oxidization of the two radicals. Mineralization of the BPA was studied by testing the TOC of the solution under different treatment time. UV–Vis spectra and change of pH value and conductivity were also explored to propose the degradation mechanism of BPA. Lastly, surface properties of AC were inspected to account for the effect of discharge on the AC structure. The research would expand the application of the PDP/AC and provide more detail mechanism analysis for the synergistic effect.

2. Experimental

2.1. Material

AC used in present study was purchased from Sanniu AC Factory in Liyang, Jiangsu, which was firstly sieved into uniform size of 2–3 mm. Powder was then removed by rinsing the AC with deionized water and the pure AC was dried at 105 °C for 24 h. The preprocessed AC was finally stored in a desiccator before use. The initial concentration of BPA used in this study was controlled at 20 mg/L and the corresponding pH value and conductivity were about 6.9 and 20 μS/cm, respectively. Initial pH value of the BPA solution was adjusted with 0.1 mol/L NaOH and 0.1 mol/L HCl. Conductivity of the BPA solution was adjusted with 0.1 mol/L NaCl solution. All chemicals used in the study were of an analytical grade unless special instructions.

2.2. Experimental setup

The schematic diagram of PDP system was shown in Fig. 1, which was similar with our previous research [32,33]. The PDP system was composed of a power supply, an electric monitoring system, a circulatory reaction system and an emission spectrum detection system. The power supply was made up of a DC power supply, a storage capacitance, an adjustable trim capacitance and a rotating spark-gap switch. The frequency of the power supply was 0–150 Hz adjustable and the pulse voltage was 0–50 kV adjustable. The C_P was 2 nF. The electric monitoring system included a voltage probe (Tektronix P6015A), a current probe (Tektronix P6021) and an oscilloscope (Tektronix TDS3032B). The circulatory reaction system was a connection of a reactor, a peristaltic pump (Longer pump BT600-2J) and a magnetic stirrer (Ronghua 85-2). The total height and the inner diameter of the reactor were 140 mm and 80 mm, respectively. The high voltage anodes were seven syringe needles (type 12#, length 100 mm), which were located in the upper of the discharge chamber. One needle was in the center and the other six needles distributed uniformly around a circle of 30 mm radius. The ground cathode was a stainless steel plate (80 mm diameter, 3 mm thickness), which was located on the bottom of the discharge chamber. AC was put on the stainless steel plate. Air was

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