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ORIGINAL ARTICLE

Removal of caffeine from water by combining dielectric barrier discharge (DBD) plasma with goethite



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Abstracts In this research, dielectric barrier discharge plasma was developed to cooperate with goethite for removing caffeine in aqueous solution. Goethite was characterized by X-ray diffraction and scanning electron microscopy. The effects of input power, initial concentration and catalysts concentration on the removal efficiency of caffeine were evaluated. Furthermore, the degradation pathways of caffeine were also discussed preliminarily. In the case of caffeine concentration at 50 mg L⁻¹, the degradation efficiency of caffeine was improved from 41% to 94% after 24 min on the conditions of input power of 75 W by combining goethite catalysts (2.5 g L⁻¹), while the energy efficiency could be enhanced 1.6–2.3 times compared to the single DBD reactor. The reaction mechanism experiments demonstrated that attack by hydroxyl radical and ozone was the main degradation process of caffeine in aqueous solution. These studies also provided a theoretical and practical basis for the application of DBD-goethite in treatment of caffeine from water.

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1. Introduction

Caffeine (C₈H₁₀N₄O₂) is one of the most consumed stimulating substances, being present in cola drinks, coffee, tea and energy drinks [1]. Caffeine has a high water solubility (*K_s* value

is more than 10,000 mg L⁻¹) and low octanol–water partition coefficient, which make it easy to dissolve in aqueous environment [2,3]. Due to the low efficiency of conventional wastewater treatment process, caffeine has been detected in many surface water and ground water. Caffeine has been found at concentrations of respectively 0.03 to 1.28 nmol L⁻¹ in Switzerland [4], while concentrations of 0.05 to 0.5 nmol L⁻¹ were found in Germany [5]. Caffeine is also detected in groundwater with concentrations ranging from 0.05 to 1.2 nmol [6]. Many treatment technologies are introduced to remove caffeine from wastewater, including activated carbon adsorption, biodegradation, ozonation, photo-Fenton, photo-oxidation, electrochemical oxidation, and photo-

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electrolysis [7–9]. However, the low efficiency, incomplete degradation, and high energy consumption limit the wide application of these technologies.

Non-thermal plasma (NTP) has been reported as efficient technique for the removal of pollutants in air, but this technique was not applied until now for the removal of water pollutants on a large scale [10,11]. Reddy et al. [12] combined CeO_2 , $\text{Fe}_2\text{O}_3/\text{CeO}_2$ and $\text{ZrO}_2/\text{CeO}_2$ catalysts with non-thermal plasma reactor for the degradation of phenol in water. The results confirmed that mineralization of phenol was enhanced by the catalyst addition up to 43.7%. The similar results were obtained on the removal of a model pharmaceutical compound sulfamethoxazole (SMX) [13], it had been observed that $\text{ZrO}_2/\text{CeO}_2$ promoted the degradation and mineralization of SMX. They [14] also designed a dielectric barrier discharge reactor for the degradation of crystal violet and investigated effect of several operating parameters on the removal efficiency. Among these techniques, dielectric barrier discharge plasma was based on the use of a discharge electrode plate covered with a dielectric barrier of quartz [15]. Electric discharges generated in water induced different physical and chemical effects like high electric fields, UV irradiation, over-pressure shock waves, and the formation of active species like atomic oxygen O, hydroxyl radical $\cdot\text{OH}$, ozone radical ions, hydrogen peroxide, hydroperoxyl radicals, and ozone are generated, which are capable of oxidizing organic pollutants [16–18]. However, some disadvantages of single DBD plasma, high energy consumption and low mineralization efficiency, limit the wide spread use of this technology.

Goethite is one of the most thermodynamically stable iron oxide [19] and formed close-packed array of O^{2-} and OH^- anions with Fe^{3+} . During last decades, iron minerals have drawn significant interest for their potential application in the field of wastewater treatment due to their excellent absorption and catalytic capacities [20,21]. Recently, Lin et al. [22] found that more than 99% of 2,4,4-trichlorobiphenyl was degraded in a goethite-catalyzed Fenton-like system due to the increase in $\cdot\text{OH}$ radical concentration. Zhou and co-workers [23] designed a novel heterogeneous sonophotolytic goethite/oxalate Fenton-like system (SP-FL) for removal of sulfamethazine. Compared with Fenton-like system, they found that SP-FL system could achieve synergistic degradation of antibiotic sulfamethazine. However, no relevant information (to our best knowledge) was provided regarding degrading of PPCPs combined with dielectric barrier discharge plasma with goethite catalysts.

In the present study, the degradation of aqueous caffeine was conducted using a DBD plasma with goethite catalysts for the first time. In order to provide a theoretical and technical reference for the application of caffeine degradation by catalysts-plasma, the effect of goethite on pollutants degradation was investigated and its degradation mechanism was discussed. Furthermore, the degradation pathways of caffeine were also discussed preliminarily.

2. Experimental

2.1. Materials

Caffeine ($\text{C}_8\text{H}_{10}\text{N}_4\text{O}_2$) was purchased from Sigma–Aldrich, goethite (FeOOH) was kindly provided by Zhenjiang Chemical

Reagent Co. Ltd, all other chemicals and solvents were of analytical grade and without any purification (Aladdin Inc.), and all solutions were prepared in ultrapure water ($R = 18.25 \text{ M}\Omega$), which was purified by a Milli-Q Gradient system (Millipore, Molsheim, France).

2.2. Experimental reactor and method

The schematic of plasma-catalyst system (Fig. 1(a)) was described by our previous literature [24]. The plasma system consisted of a high voltage alternating current (AC) power source (CTP-2000 K, Nanjing Suman Electronics Co., Ltd., China) and a plasma reactor. The electrodes were in air and it was a filamentary discharge. The actual discharge phenomenon is shown in Fig. 1(b). The discharge voltage and current were measured by an oscilloscope (TDS3032, Tektronix), which was displayed in Fig. S1. The equivalent voltage and current per delivered could be integrated by the area of corresponding state variables, and input power was calculated by the following equation.

$$P = \frac{1}{T} \int_0^T U(t) \times I(t) \times dt \quad (1)$$

The main device used to treat the caffeine solution was DBD reactor containing a quartz reaction still and electrodes (Fig. 1(c)). The distance between catalysts and electric barrier was about 1 cm. The reaction still was placed in the center of two aluminum electrodes. The goethite catalysts were placed into the quartz reaction still and suspended in the solution. The goethite concentration was adjusted by adding different quantity of goethite. The liquid circulation system was driven by two peristaltic pumps with a flow rate of 50 mL min^{-1} .

In this study, 100 mL caffeine solution with the initial concentration of 50 mg L^{-1} was pumped into the plasma reactor and was made to flow through goethite in the reaction still with reaction duration of 24 min. The residence time (RT) of plasma reactor was about 18 s. Each sample (1.5 mL) was taken every 4 min. Actually, most oxidants (e.g. $\cdot\text{OH}$, $\cdot\text{H}$) produced in the discharge plasma always exit for an extremely short time except for O_3 and H_2O_2 . In order to avoid the effect of residual oxidants on the toxicity test, $2 \text{ mg Na}_2\text{S}_2\text{O}_3$ was added into 100 mL reaction solution to quench the oxidants (e.g. O_3 , H_2O_2). Prior to the experiment, the reaction was still placed in dark for 60 min to ensure that the adsorption–desorption equilibrium of caffeine on the surface of goethite.

2.3. Characterization

Crystal phase of samples were obtained by X-ray diffraction (XRD, D/Max-rB, Japan Rigaku Corporation) with $\text{Cu K}\alpha$ radiation source of wavelength 0.1541 nm in the range of 3° to 70° with a scanning speed of 4° min^{-1} .

The specific surface area, the pore volume and the pore diameter of the catalysts and other samples were determined by the N_2 adsorption–desorption isotherms (NOVA 3000e) at the liquid-nitrogen temperature.

The morphologies of the goethite was analyzed by a scanning electron microscopy (Hitachi S - 3400N II).

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