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Simultaneous removal of cadmium ions and phenol from water solution by pulsed corona discharge plasma combined with activated carbon

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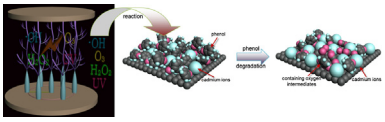
HIGHLIGHTS

- The Cd^{2+} and phenol can be simultaneously removed from PCDP/GAC system.
- A possible removal mechanism for Cd^{2+} and phenol in PCDP/GAC system is proposed.
- The intermediates and decomposition products of phenol are analyzed by GC–MS.

GRAPHICAL ABSTRACT



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ABSTRACT

In this paper, an integrated approach, the pulsed corona discharge plasma combined with granular activated carbon (PCDP/GAC), is used for the simultaneous removal of cadmium ions (Cd^{2+}) and phenol in wastewater. Experimental results show that 69.0% Cd^{2+} and 87.3% phenol can be simultaneously removed from the PCDP/GAC treatment system after 60 min. This removal efficiency is 27.2% and 11.4% higher than in the GAC adsorption system, respectively, and 65.8% and 60.6% higher than in the PCDP treatment system, respectively. An increasing pulsed voltage is favorable for removal of Cd^{2+} and phenol. The removal efficiency of Cd^{2+} and phenol is the highest with oxygen as the feeding gas. At the same input power and an increasing initial phenol concentration, the removal efficiency of Cd^{2+} and phenol decreases. Scanning electron microscopy and energy-dispersive X-ray spectroscopy show that Cd^{2+} amount produced on GAC by the discharge treatment in the PCDP/GAC system is higher than that in the GAC adsorption system. Fourier transform infrared spectroscopy shows that O—H, C—H, and C=C bonds on GAC are cleaved, whereas Ar—O—C and C=O groups increase during the PCDP treatment. A possible removal mechanism of Cd^{2+} and phenol in such a system is proposed.

1. Introduction

The co-contamination of aquatic systems with heavy metals and toxic organic pollutants is a problem of global concern [1]. Heavy metal ions and aromatic compounds from industrial activities, such as plating, metallurgy, and dyeing, are a threat to humans and the environment because of their toxicity and persistence after they have been released into the natural environment [2]. Among these pollutants, cadmium (Cd) is considered as one of the most toxic heavy metals [3]. Some aromatic compounds, such as phenol, are defined as priority pollutants by the US Environmental Protection Agency because of their strong corrosive effects on the skin and the mucosa. These compounds can also inhibit the central nervous system or cause damages to liver and kidney functions. Many traditional methods are applied to remove these two kinds of pollutants, such as electrochemical precipitation, ion exchange, reverse osmosis, and solvent extraction, which are mainly based on applications in single systems that contain either metal ions or organic pollutants [4]. Moreover, these conventional separation techniques have many disadvantages, such as low efficiency, possible production of secondary toxic compounds, and the generation of sludge, which entails high disposal costs [5–7]. Adsorption is considered as one of the most common and effective methods for the

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removal of these pollutants from wastewaters because of the flexibility in design and operation provided by the adsorption process [8]. However, the removal efficiency of adsorption technology in removing heavy metal ions from the co-pollutants (heavy metal ions and organic compounds) is low [9]. In addition, the adsorption process is not the final organic pollutant disposal method, thereby resulting in the liquid-to-solid phase transition of organic substances without degradation. Therefore, the integration of the adsorption process and other technologies has been given increasing attention for the efficient removal of heavy metal ions and toxic organic pollutants [10].

Pulsed high-voltage electrical discharge in water, or pulsed corona discharge plasma (PCDP) in aqueous solutions, has been widely considered as a novel technology for the effective elimination of organic compounds, particularly those with high toxicity and low biodegradability (e.g., phenols, chlorophenols, and organic dyes) [11–13] as well as microorganisms in water (e.g., bacteria and yeast) [14]. The technology comprises many physical and chemical processes, which directly or indirectly decompose and detoxify organic pollutants. The physical processes include UV light emission, shock waves, electrohydraulic cavitation, and pyrolysis within plasma channels as well as supercritical water conditions. The chemical processes simultaneously occur in water, such as free radical reactions, including the formation of short-lived reactive radicals (·OH, ·H, $\cdot O_2$, and $HO_2\cdot$) and the simultaneous production of long-lived molecular species (H_2O_2 and O_3) in an aqueous phase. Numerous studies have demonstrated that organic compounds in wastewater can be satisfactorily removed by the PCDP treatment [15–17].

Adsorbents are the concentration centers of pollutants, and each adsorbent can create a high concentration on its surface and its surrounding adjacent area by adsorption. As the discharge process begins, high energy electrons as well as various active radicals (·OH, ·H, ·O, $\cdot O_2$, and $\cdot HO_2$) and active molecules (H_2O_2 and O_3) are produced. The organic pollutants in water or on the adsorbent may be excited, ionized, and dissociated by high-energy electrons. Simultaneously, various active radicals and molecules as well as physical effects (such as strong electric field, UV light, and shock waves) that are generated concomitantly during the discharge process can also act on organic pollutants in water or on the adsorbent. Given the synthetic effects of high-oxidative active species and physical processes, organic pollutants in water and on the adsorbent may be removed effectively and rapidly, thereby releasing adsorption sites of the adsorbent and increasing the adsorption capacity of the adsorbent for heavy metal ions.

In this study, we designed a bench-scale experiment to investigate a co-contamination water treatment technique, which integrated the PCDP treatment and the activated carbon (AC) adsorption process for the simultaneous removal of heavy metal ions and organic pollutants in wastewater. Phenol was chosen as a model aromatic co-pollutant of cadmium ions (Cd^{2+}) because of its relatively wide occurrence in Cd^{2+}-contaminated sites. The two main aspects examined were as follows: (1) the effect of discharge voltage, type of feeding gas, and initial phenol concentration on the removal efficiency of Cd^{2+} and phenol; and (2) Cd^{2+} reduction and phenol degradation in wastewater. A possible mechanism for removal of Cd^{2+} and phenol in such a system was also proposed. It is expected to provide a high efficient method for wastewater treatment with co-pollutants (heavy metal ions and organic compounds).

2. Materials and methods

2.1. Materials

All chemicals were of analytical agent grade. Cadmium chloride ($CdCl_2$) was used to produce heavy metal ions which were purchased from Shanghai Tingxin Chemical Factory in China. Phenol was supplied by Tianjin Fuchen Chemical Reagent Factory in China. The test solutions containing single Cd^{2+} or phenol were prepared by diluting 1.0 g/l stock solutions of Cd^{2+} or phenol to the desired concentrations. The granular activated carbon (GAC, 1.5 mm diameter, 3.0–5.0 mm length) used in this study was columned coal-based carbon and manufactured by Shenyang Chemical Reagent Factory, China. The GAC pretreatment procedure was presented in S1 of Supplementary material (SM).

3. Experimental methods

The schematic diagram of experimental apparatus for the PCDP/GAC treatment system was illustrated in Fig. 1a. The system consisted of a pulsed high-voltage power supply and a reactor vessel. The structure of the pulsed high-voltage power supply could be seen in Wang et al. [18]. The pulse frequency and pulse-forming capacitance C_p was 40 Hz and 3.3 nF, respectively. The three dimensional graph of the reactor vessel was shown in Fig. 1b and its detailed introduction was presented in S2 of SM. The pulse peak voltage (U_p) and current delivered to PCDP combined with GAC system (PCDP/GAC system) were measured with oscilloscope (Tektronix TDS1012B-SC) equipped with voltage probe (Tektronix P6015A) and current probe (Tektronix P6021). The typical pulsed voltage and current waveforms obtained in the experiment were shown in Fig. S1 of SM. The input energy was calculated through the integral of pulse voltage and current under time.

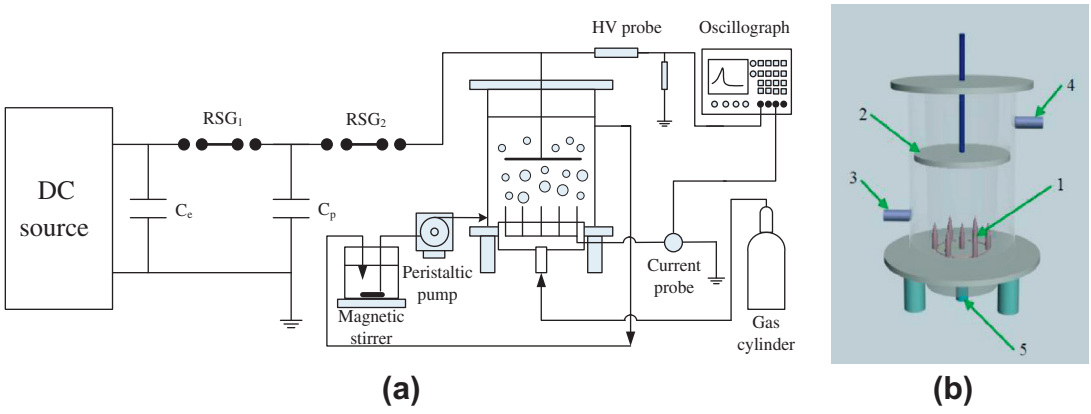

Fig. 1. Schematic diagram of experimental apparatus: (a) system graph, (b) three dimensional graph of the reactor vessel ((1) high voltage multi-needle electrode, (2) ground plate electrode, (3) wastewater inlet, (4) wastewater outlet, and (5) gas inlet).