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Effect of chemical parameters on pyrene degradation in soil in a pulsed discharge plasma system

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ABSTRACT

Degradation of pyrene in soil in a net-to-net pulsed discharge plasma (PDP) system was reviewed. Effect of main chemical parameters, including air flow rate, pyrene concentration, initial pH and soil moisture content on pyrene degradation was examined. The obtained results show that 87.9% of pyrene could be removed under the condition of 60 min reaction; increasing of air flow rate within 1 L min^{-1} was favorable for degradation; pyrene removal was decreased with the increase of initial pyrene concentration; oxidation of pyrene was more evident in acidic soil; enhancement of soil moisture content has no benefit on pyrene degradation.

Introduction

Polycyclic aromatic hydrocarbons (PAHs) are listed as priority pollutants by USEPA due to its teratogenicity, carcinogenicity, mutagenicity and toxicity [1]. Nowadays, because of the various applications of PAHs, high concentration of wasted PAHs is found in air, water, as well as soil. Especially for the soils near coal tar, creosote, manufactured gas plants (MGP) and wood treatment facilities, which can be polluted by PAHs easily by leaking, accidental chemical spilling, slag, coal disposal and incomplete combustion of fossil fuel [1–3]. Accumulation of PAHs in plants and underground water caused by the leakage of PAHs from soil can lead to severe risk to human health. A variety of technologies, such as chemical, biological, physical, thermal processes and their combinations have been carried out accordingly to remediate PAHs contaminated soils [4]. However, during the development of these technologies, some problems emerge: biological process needs much time to finish the remediation process and also requires rigid conditions for keeping the growth of microorganisms [5,6]; conventional chemical methods may result in secondary pollution and the whole process costs much [7,8]; electro-kinetic technology is limited by the solubility and desorption of organic pollutants in soils [9,10]. Therefore, feasible remediation method needs to be provided and investigated for the remediation of PAHs-contaminated soils.

Recently, advanced oxidation processes (AOPs), as a novel method, have been paid great attention on pollution control because of its ability to oxidize refractory organic contaminants deeply and rapidly. As one of lately developed AOPs, low-temperature discharge plasma has aroused considerable interest due to its higher removal efficiency and environmental compatibility. Pulsed discharge plasma (PDP), which has received extensive concern in recent years, is one kind of low-temperature discharge plasmas [11–13]. It has been concluded that a variety of active species, such as •OH, •H, •O, $•HO_2$ and O_3, can be generated in a PDP system to oxidize the organic contaminants [14,15]. In addition, physical processes, including strong electric field, ultraviolet light, and shock waves, can also make contribution to the organic pollutants degradation. Plentiful studies have confirmed that PDP can be applied in wastewater treatment successfully [16,17]. Some investigations also review the feasibility of PDP on the remediation of persistent organic pollutants (POPs) contaminated soils. Researches carried out by Wang et al. [18–20] demonstrate the possibility of pentachlorophenol, p-nitrophenol and phenol's removal from soil in a pulsed discharge plasma system with multi-need-to-plate electrode. In these studies, pentachlorophenol degradation efficiencies can arrive to 92% and 77% after 45 min of discharge treatment at 14.0 kV, under oxygen and air atmosphere respectively, and the removal efficiency of p-nitrophenol was 78.1% in plasma system under the condition of 20 kV pulsed discharge

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voltage. O_3, •OH, •O, and H_2O_2 are considered as the main active species for organic compounds degradation [19]. These results confirm the possible application of PDP on the treatment of PAHs contaminated soils.

On account of the serious soil pollution, toxicity of PAHs and characteristics of PDP technology, a PDP system with net-to-net electrode was set up in this research and was then used for the remediation of PAHs-contaminated soils. One of the typical PAHs, pyrene, was chosen as the target pollutant in present study to explore the effects of some critical chemical parameters, including air flow rate, pyrene concentration, initial pH values of the soil and soil moisture content on pyrene degradation in the PDP system. The research would set up a new PDP system, which is favorable for soil remediation and provide some fundamental data for the further application of the system. Energy efficiency of pyrene removal in the PDP system was also calculated in each section of the research to explain the effect of PDP on pyrene degradation in soil.

Experimental

Materials

Pyrene (Aldrich, purity 98%) was used as the representative of PAHs in the study. Other organic and inorganic reagents used in the research were analytical grade and were not further purified after purchase.

Soil sample was collected from Nanshan, a scenic spot in Zhenjiang. The collected soil was passed through a 10 mesh standard sieve after 7-day air drying. The measured soil properties were as follows: organic matter content is 8.86 g kg^{-1}; bulk density is 1.12 g cm^{-3}; pH is 6.04; moisture content is 6.9%; cation exchange capacity is 6.57 cmol kg^{-1}.

Pulsed discharge plasma system

As shown in Fig. 1, the pulsed discharge plasma system used in the research consists of three parts: a pulsed high-voltage power supply, a reactor and an electrical parameters measurement system. High-voltage pulse discharge were produced by combining a 0–50 kV adjustable DC power supply, a storage capacitor (Ce), an adjustable trim capacitance (Cp) and rotation spark gap switches (RSG1, RSG2). The pulse rise time was less than 100 ns, and the pulse width was less than 500 ns. The pulse frequency and Cp were 0–150 Hz adjustable and 250 pF, respectively. The reactor (shown in Fig. 2) was comprised of a plexiglass™ cylinder and a couple of net-to-net electrodes. The total cylinder height was 255 mm, the inner diameter was 110 mm, and the thickness of the reactor was 5 mm. The net-to-net electrodes were consisted of two parts, and each part was made of a stainless-steel net (200 mesh) wrapped around a resin disc with several uniformly distributed pores on it. The distance between the high-voltage electrode and the ground electrode was controlled at 10 mm. The carrier gas used in the system was air, which was introduced into the reactor from the top of the cylinder and was exported from the bottom of the reactor. The flow rate of air was controlled by adjusting a flow meter. The peak pulse voltage and current were measured with a TektronixTDS3032B digital oscilloscope, a Tektronix P6015A high-voltage probe and a Tektronix P6021 current probe, and the energy consumption was calculated by the integral of pulse voltage and current wavelength.

Experimental method and analytic procedure

Preparation of soil samples

Pyrene contaminated soil samples used in this paper were got by adding a certain amount standard pyrene acetone solution into a given amount of pretreated soil. The soil was then put in a vapor-bathing constant temperature vibrator and shaken at the speed of 250 rpm for 4 h. The homogenized soil was finally put in a fume hood until 100% acetone evaporated entirely, which was confirmed by weight loss method.

The concentration of pyrene reviewed in the investigation was 100 mg kg^{-1}, 200 mg kg^{-1} and 300 mg kg^{-1}. The initial pH of the 100 mg kg^{-1} pyrene contaminated was about 6 and the soil moisture content was around 5%. pH values of the soils were evaluated by measuring the pH of the mixture of soil and distilled water at a ratio of 1:2.5 (soil:water = w/w). The change of the soil pH values were realized by adding H_2SO_4 and NaOH. The change of soil moisture content was achieved by adding certain amount of deionized water into the soil.

Extraction procedure of pyrene from soil

Firstly, adding the soil sample into an acetone-dichloromethane mixed solution (v/v = 50/50) and putting the mixture in an ultrasonic apparatus for 1 h extraction. The mixture was then centrifuged at 1500 rpm for 10 min and the supernatant was collected for further purification by going through a glass column filled with silica gel (200–300 mesh). The effluent liquid after washing by acetone-hexyl hydride mixed solution (v/v = 50/50) was then dried

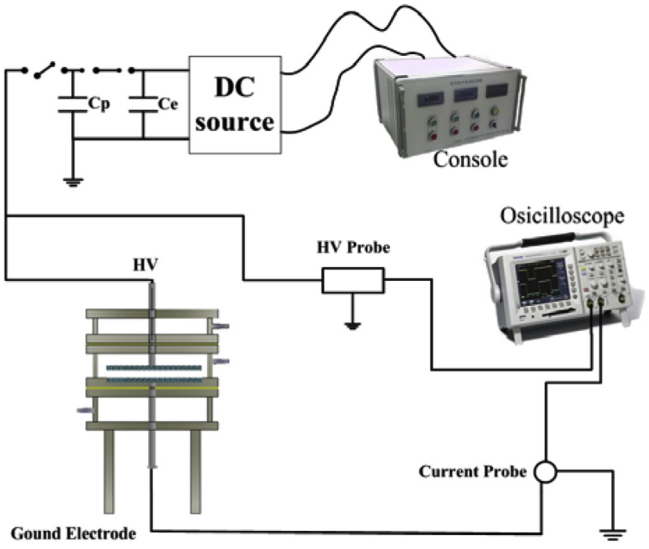

Fig. 1. Schematic diagram of the experimental system.

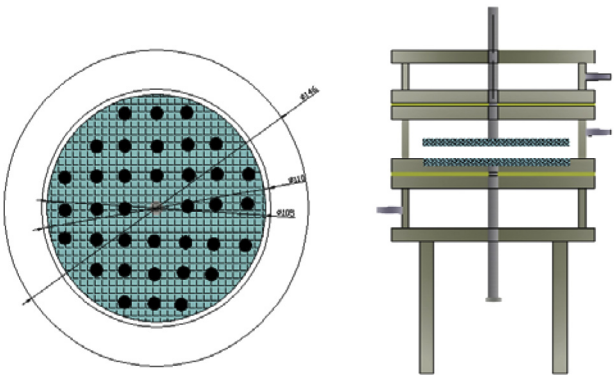

Fig. 2. Top view and section view of the reactor.