



# Remediation of *p*-nitrophenol and pentachlorophenol mixtures contaminated soil using pulsed corona discharge plasma



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## ABSTRACT

The feasibility of remediation of *p*-nitrophenol (PNP) and pentachlorophenol (PCP) mixtures contaminated soil was investigated using pulsed corona discharge plasma. The influences of initial pollutant concentration, the existence of the second pollutant, and soil pH on pollutants removal were studied. The results showed that the mixed pollutants could be removed effectively by pulsed corona discharge plasma. For 600 mg kg<sup>-1</sup> mixtures contaminated soil, comprised of 300 mg kg<sup>-1</sup> PCP and 300 mg kg<sup>-1</sup> PNP, 86.0% of PNP and 94.1% of PCP were removed at the specific energy density of 485 J g<sub>soil</sub><sup>-1</sup>, and correspondingly, the energy yield was about 18.3% higher than for treating single PNP contaminated soil (600 mg kg<sup>-1</sup>) and 6.1% lower than for treating single PCP contaminated soil (600 mg kg<sup>-1</sup>). PNP and PCP degradation efficiencies both decreased with increasing initial pollutant concentration. In mixtures contaminated soil, the changes of PCP initial concentration brought obvious influences on both PNP and PCP degradation, as well as on energy yield; however, the changes of PNP initial concentration contributed to more obvious effects on PCP degradation than PNP degradation, and caused little influence on energy yield. The degradation characteristics of mixtures depend on their acid–base equilibrium constants, and alkaline condition was beneficial for both PNP and PCP removal. The differences in degradation, mineralization, dechlorination and denitrification between single pollutant contaminated soil and mixtures contaminated soil were attributed to the different reaction rates of active species with PCP and PNP molecules.

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## 1. Introduction

The improper disposal practices and accidental spillage of organic chemicals have led to serious soil contamination problems. The contamination of soils by biorefractory organic pollutants, such as chlorinated aromatic and nitroaromatic organic compounds, are of environmental concern due to the toxicity and stability exhibited by these pollutants [1–3]. Efforts to explore effective methods to remediate soil contaminated with organic compounds have led to the study of alternative remediation strategies, such as Fenton method [4], bioremediation [5], electrokinetic remediation [6], and photocatalysis [7].

These studies have one common feature, that is, they all use single organic pollutant as the treatment object. The environmental behaviors of single pollutant and soil remediation mechanisms have been investigated widely and detailedly in these studies.

However, the research on the environmental behavior of single organic pollutant would exhibit insufficient evidence as the basis for development of soil environmental standards and capacities, because actually polluted soils are often contaminated with complex mixtures of chemicals. Therefore, more attention should be paid to the research on the environmental behaviors of mixtures in soil.

In general, degradation characteristics of mixtures have great relevance with their intrinsic properties and mutual effects of pollutants, because degradation of one compound may be significantly affected by other compounds in the mixture. It has been reported that the degradation of single 2-chlorophenol and 2-nitrophenol by TiO<sub>2</sub>/UV was faster than that in binary mixtures [8]. The same trends were also observed in the systems containing any two components of phenol, guaiacol, 2-chlorophenol, and catechol [9]. On the contrary, Alber et al. has reported that greater mineralization potential was obtained in bioremediation of mixed soil containing pentachlorophenol (PCP) and *p*-nitrophenol (PNP) than in single contaminated soil in the case of (NH<sub>4</sub>)<sub>2</sub>HPO<sub>4</sub> as nitrogen source [10].

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In our previous studies, pulsed discharge plasma has been employed to remediate single PNP and PCP contaminated soils respectively, and great performances of soil remediation were obtained [11,12]. To realize the remediation characteristics of actual polluted soil adequately by pulsed discharge plasma, it is of great significance to understand degradation characteristics and mutual effects of mixtures in soil. In the present study, the remediation of PCP and PNP mixtures contaminants soil was investigated using pulsed corona discharge plasma. The feasibility for mixtures contaminated soil remediation was evaluated. In order to understand the mutual effects, the degradation characteristics of single pollutant and mixtures in soils were compared. The effects of initial pollutant concentration, the presence of the second pollutant, and soil pH on remediation characteristics were systematically evaluated, and the degradation mechanisms were also discussed.

## 2. Experimental

### 2.1. Materials

PCP and PNP were purchased from the Chemical Plant of Tianjin, China, with above 99% purity. All other organic and inorganic reagents used were analytical grade (Tianjin Fuyu Refinery Chemical Co., Ltd. and Shenyang Chemical Reagent Factory, China) and were used as purchased without further purification.

Soil sample in this study was collected from a suburb of Dalian, China. The pretreatment process, the soil properties and the preparation of single pollutant contaminated soil were the same as our previous research [11]. The mixtures (PNP and PCP) contaminated soil was prepared by adding appropriate amount of PCP, PNP, and acetone solution into a given amount of pretreated soil. The soil was vigorously homogenized, and then placed in the fume hood until acetone evaporated entirely. Soil pH was adjusted with NaOH and H<sub>2</sub>SO<sub>4</sub> solutions as described by Hultgren et al. [13].

### 2.2. Treatment of contaminated soil

The schematic diagram of the experimental apparatus was illustrated in Fig. 1, which was similar with previous work [11]. The reaction system consisted of a pulsed high-voltage power supply and a reactor vessel. Briefly, the high voltage electrode comprised of 19 stainless-steel hypodermic hollow needles with inner diameter of 0.7 mm and outer diameter of 1.0 mm, and the needles were distributed uniformly in a multi-pore plate. The ground electrode was a wire netting (200 mesh). The distance of adjacent needle was 12.5 mm and the distance between the high voltage electrode

and the ground electrode was 16 mm. The pulse voltage and current were measured with a Tektronix TDS2014 digital oscilloscope equipped with a Tektronix P6015A high voltage probe and a Tektronix A6021 current probe, and then the electric power was calculated through the integral of pulse voltage and current under time. The pulse discharge voltage, pulse frequency and pulse-forming capacitance  $C_p$  were 18 kV, 50 Hz and 200 pF, respectively. The input energy per pulse was 0.018 J.

In each experiment, the soil sample (approximately 5.0 g, 3.1 mm depth) was spread on the ground electrode. Prior to discharge treatment, the moisture content of soil was adjusted to about 15% with deionized water. Air was injected into the reactor with the flow rate of 0.5 L min<sup>-1</sup>, and then passed through the pores in the plate into the plasma region.

### 2.3. Extraction and analysis

After each discharge experiment, the soil sample was mixed with 50 mL deionized water in an airtight sealed conical flask, which was shaken on a shaker at 300 rpm for 6 h, and then centrifuged at 3000 rpm for 15 min, and finally the extract was collected.

PNP and PCP concentrations were analyzed by HPLC system (SCL-10ACP, Shimadzu, Japan) equipped with a Hypersil ODS (25 μm, 4.6 × 250 mm) reverse phase column, which were the same as previous research [11,12]. The recoveries were 90.1–95.3% for PNP and 83.4–87.4% for PCP ( $n = 5$ ). The methods for degradation intermediates identification could be seen in previous research [11,12].

Cl<sup>-</sup>, NO<sub>3</sub><sup>-</sup> and NO<sub>2</sub><sup>-</sup> were detected by ion chromatography (SCL-10ASP, Shimadzu, Japan) equipped with Shimpack IC-A3 column (4.6 mm × 150 mm) and COD-10AVP conductance detection. The mobile phase consisted of a mixture of 8.0 mmol L<sup>-1</sup> *p*-hydroxybenzoic acid, 5.5 mmol L<sup>-1</sup> Bis-Tris, and 50.0 mmol L<sup>-1</sup> boric acid solutions with a flow rate of 1.0 mL min<sup>-1</sup>. The mineralization was determined by a Total Organic Carbon analyzer (TOC-5000A Shimadzu). COD was measured by the potassium dichromate method.

The specific energy density (SED, J g<sub>soil</sub><sup>-1</sup>) can be calculated by the following equation:

$$SED = \frac{E \cdot f \cdot t}{m_{soil}} \quad (1)$$

where  $m_{soil}$  is the total amount of treatment soil (g),  $E$  is the input energy per pulse (J),  $f$  is the pulsed frequency (Hz), and  $t$  is the treatment time (s).

The energy yield ( $G$ , g kW h<sup>-1</sup>) is defined as the mass of removed PCP or PNP ( $m$ ) divided by consumed energy.

$$G = \frac{m}{E \cdot f \cdot t} \quad (2)$$

Denitrification efficiency and dechlorination efficiency are defined as described in our previous studies [11,12].

## 3. Results and discussion

### 3.1. Effect of PCP initial concentration on mixtures degradation

In order to evaluate the mutual effects of target pollutants, the effect of PCP initial concentration on mixtures contaminated soil remediation was firstly investigated, and as control experiments, remediation experiment of single PCP contaminated soil was also conducted.

Fig. 2 showed the evolution of PCP degradation efficiency and energy yield with treatment time under different initial concentrations in single PCP contaminated soil. PCP degradation efficiencies

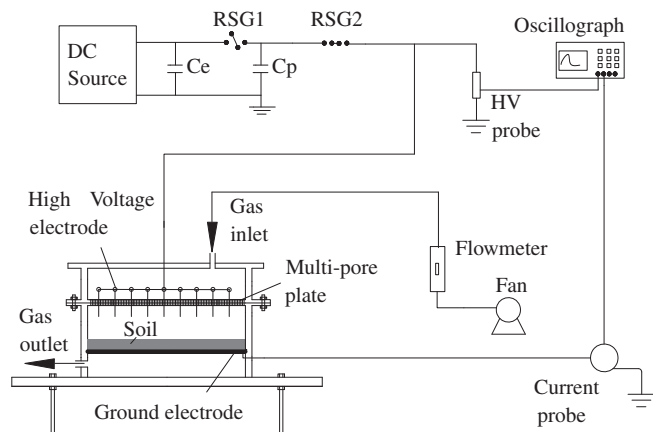


Fig. 1. Schematic diagram of the experimental apparatus.

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