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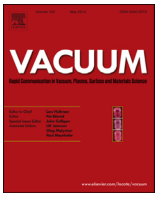

Evaluation and optimization of multi-channel pulsed discharge plasma system for soil remediation

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ABSTRACT

Pulsed discharge plasma is a rapid and high-efficient option for organic pollutants contaminated soil remediation. In order to optimize the coupling of pulsed power supply and plasma reactor, and enhance pollutant removal in soil by pulsed discharge plasma, power circuit parameters and reactor configuration such as adjustable trim capacitance, discharge needle number, and discharge voltage were optimized with respect to energy input, power source efficiency, pollutant degradation, process efficiency, and energy yield. *p*-Nitrophenol (PNP) was chosen as the model pollutant in soil. The experimental results showed that 90.7% of PNP were degraded under the conditions of adjustable trim capacitance 200 pF, discharge needle number 19, and discharge voltage 18.0 kV, with power source efficiency of 62.5%. The increase of adjustable trim capacitance was beneficial for PNP degradation and process efficiency, but not for power source efficiency and energy yield. Higher PNP degradation efficiency was obtained as more discharge needle numbers were employed, as well as for the power source efficiency and energy yield. Increasing discharge voltage benefited PNP degradation, but went against energy yield. The power source efficiency increased firstly with discharge voltage, and then decreased with further increasing discharge voltage. Furthermore, PNP mineralization was also analyzed.

1. Introduction

With the quick development of industrial production and urbanization, lots of relocated or closed industrial enterprises caused serious soil pollution [1–3]. In these polluted sites, a large number of toxic contaminants have been continually entered into plant sites including petroleum hydrocarbon, heavy metals, pharmaceuticals and persistent organic pollutants [4–7]. Among the contaminants, phenols have been highly overused and misused in the manufacture of dyes, pesticides and medicines, resulting in the accumulation in soil and great threat to human health [8–10]. Therefore, remediation of these polluted sites is of great necessity.

Several technologies such as physical method [11], chemical method [12], bioremediation [13], and photocatalysis [14] have been employed to remedy organic pollutants contaminated soils. With the strengthening of industrial standard and the increasing of economic values of lands, the conventional remediation technologies can not meet the requirement of high efficient and rapid remediation due to drawbacks such as second pollution and time-consuming, and thus it is urgent to explore new methods for soil remediation.

Advanced oxidation processes (AOPs), which take advantage of the high oxidizing potential of reactive species obtained from molecular oxygen or water, are the obvious choice. Among the AOPs, non-thermal discharge plasma is a promising approach [15,16]. During discharge plasma processes, the ensuring electron–molecule interactions generate highly reactive non-thermal plasma, which are strongly oxidizing environments due to the presence of large number of chemically active species, such as ozone, H_2O_2, OH and OOH radicals, O atoms, and ions (O_2^-, O_2^+, H_3O^+, O_3^-) [15]. A schematic view of the relevant discharge plasma processes is presented in Fig. 1. Based on its principle for pollutants decomposition, it is suggested that organic pollutants in soil can be removed effectively and rapidly when the polluted soil was placed in discharge plasma region.

In our previous study, pulsed discharge plasma was employed to remedy *p*-nitrophenol (PNP) contaminated soil, and it has been proved that PNP in soil could be degraded effectively and rapidly [17]; however, during soil remediation by pulsed discharge plasma, fundamental knowledge on output characteristics of the pulsed

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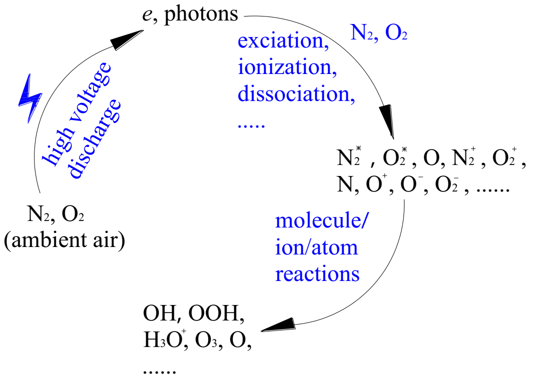

Fig. 1. Schematic view of the relevant discharge plasma processes.

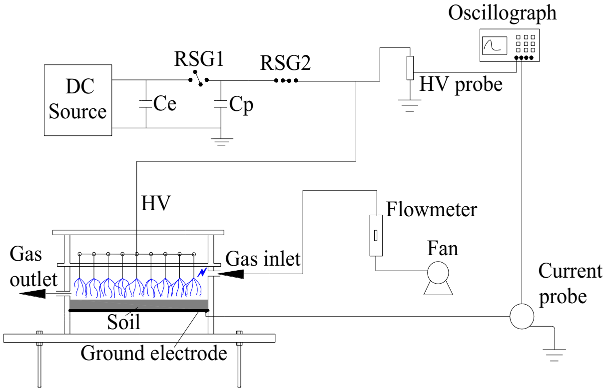

Fig. 2. Schematic diagram of the experimental apparatus (Ce was the storage capacitor, Cp was the adjustable trim capacitor).

power supply, discharge characteristics of plasma reactor, and the relationship of energy input and utilization is still unsatisfactory and not often available. In pulsed discharge plasma system, the effective input of pulsed energy into the discharge reactor and its high-efficient utilization depends on the output characteristics of the pulsed power supply and discharge characteristics of the plasma reactor, and the energy utilization directly affects pollutants removal [18]. Furthermore, power circuit and reactor configuration exhibit obvious effects on the output characteristics of the pulsed power supply and discharge characteristics of the plasma reactor [19–21].

Therefore, in order to evaluate and optimize the coupling of pulsed power supply and plasma reactor, and then improve energy utilization and pollutant degradation, a multi-channel plasma reactor was designed; the effects of power circuit parameters (such as adjustable trim capacitance, voltage output) and reactor configuration (such as discharge needle number) on the output characteristics of the pulsed power supply, discharge characteristics of the plasma reactor, and PNP degradation performance were evaluated in terms of energy input, power source efficiency, energy utilization, and energy constant.

2. Experimental

2.1. Materials

PNP and other reagents were analytical grade and used as purchased without further purification. The soil samples, soil pretreatment processes, and preparation of PNP contaminated soil were the same as our previous research [17]. PNP initial concentration in the soil was 800 mg kg^{-1}.

2.2. Soil remediation system

The schematic diagram of the experimental apparatus was illustrated in Fig. 2, which was similar with our previous work [17]. Specially, high-voltage pulses were generated using the combination of a 0–50 kV adjustable DC power source, a storage capacitor (C_e), an adjustable trim capacitor (C_p) and rotation spark gap switches (RSG1, RSG2). In the power supply system, C_e and C_p were charged respectively by the changes of the rotation spark gap switches position, and then C_p was discharged towards to the reactor, forming pulse discharge. The pulse rise time was less than 100 ns, and the pulse width was less than 500 ns. Stainless-steel hypodermic needles (inner diameter of 0.7 mm and outer diameter of 1.0 mm) were used as high voltage electrode, which were distributed uniformly in a circle of 50 mm diameter, and the needle number was adjustable to change the discharge channel. Stainless-steel plate was used as ground electrode. 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 frequency was 100 Hz in the present research. The peak 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.

In each experiment, PNP contaminated soil samples (approximately 2.0 g, 1.3 mm depth) were spread on the ground electrode. Soil moisture content was about 15%. Air was injected for one side of the reactor and out from the other side with the flow rate of 0.5 L min^{-1}.

2.3. Extraction and analysis

PNP extraction procedure and analysis methods were the same as previous study [17]. The energy stored in adjustable trim capacitance (W, J $pulse^{-1}$) was calculated as follows [22],

$$W = \frac{1}{2} C_p U^2 \tag{1}$$

where C_p was the adjustable trim capacitance (F), and U was peak pulse voltage (V).

The energy input into reactor (E_{in}, J $pulse^{-1}$) was calculated by the following equation [22],

$$E_{in} = \int_0^T u(t) \times i(t) dt \tag{2}$$

where $u(t)$ and $i(t)$ were the discharge voltage (V) and current (A) at time t (s), respectively.

Power source efficiency (θ) was calculated by the following equation [22],

$$\theta = \frac{E_{in}}{W} \times 100\% \tag{3}$$

Energy yield (G, g kWh^{-1}) was defined as the removed PNP divided by input energy,

$$G = \frac{m_{PNP}}{E_{in} \cdot f \cdot t} \tag{4}$$

where m_{PNP} was the amount of PNP removed.

The specific energy density (SED, J g_{soil}^{-1}) was calculated by the following equation,