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Technical Note

Degradation of phenol in mists by a non-thermal plasma reactor

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ABSTRACT

A link tooth wheel-cylinder non-thermal plasma reactor was set up to investigate the degradation of phenol in the mists. In addition, the decomposition efficiency of phenol, TOC removal, and byproduct formation were investigated. The stable discharge was achieved in both air and the mist condition. The decomposition efficiency and TOC removal increased with increasing the input power. For the input power of 3.6 W, the phenol decomposition and TOC removal reached 90% and 47%, respectively. Phenol degradation byproducts were identified as small molecular organic acids, including formic acid, acetic acid, and oxalic acid. Their masses in the trapped solutions first increased and then decreased slightly with increasing the input power. Therefore, the biodegradation capacity of the phenol degradation byproducts can be improved.

1. Introduction

Many processes, for example, wet coke-quenching (WCQ) in coking plants, paint spraying in the surface treatment industry, water cooling in the metallurgical industry, and hair removal by boiling in abattoirs, are sources of mists. These mists usually contain multiple contaminants such as volatile phenols, aliphatic hydrocarbons, and ammonia. For example, in the WCQ process, when untreated coking water is used to quench a ton of coke, 3 g of benzopyrene, 69 kg of ammonia, 52 kg of volatile phenols, 2 kg of total cyanide, 0.5 tons of water, and 25–410 g of coke powder are emitted in the mist (Xi and Guo, 2003; Zhao, 2007). Consequently, the control of mist pollution is of considerable importance in environmental protection.

At present, the conventional methods used to control mist pollution are relatively undeveloped. For WCQ mists, inertial impaction is mainly used to remove water and coke powder (Gao, 2005), without treatment of other pollutants. Scrubing is a basic method for removing particulate matter in waste gases from paint spraying. Only a few treatment technologies combine scrubing with treatment of volatile organic compounds, and these processes have very low efficiencies (Qi et al., 2005). Emission of the mists produced in the metallurgical industry and in abattoirs is not controlled. Therefore, the development of technologies for mist purification is therefore needed.

Non-thermal plasma treatment, an advanced oxidation process with potential applications in purification of environmental contaminants, has been widely studied for more than three decades. Earlier published work reported that non-thermal plasmas based on production of oxidative radicals could be applied to removal of a wide variety of gaseous contaminants, such as volatile organic compounds in indoor air (Fan et al., 2009; Schmid et al., 2010), NO_x and Hg in flue gas (Jeong and Jurng, 2007; Wang et al., 2009). Wastewater treatment by non-thermal plasmas, with varying degrees of efficiency, has also been widely investigated (Malik et al., 2001; Hao et al., 2007). Much research has been carried out on the removal of phenol in aqueous solutions using plasma reactors (Faungnawakij et al., 2006; Sato et al., 2008). Generally, the OH radical is considered to be the dominant radical in the oxidation of aqueous phenol (Wang et al., 2008). In addition, decomposing organic compounds by spraying a water solution of organic compounds as droplets into non-thermal plasma in gas phase was investigated using a pulsed-corona discharge reactor. The results showed that spraying as water droplets into the discharge in air is effective for faster water treatment (Yasushi et al., 2008; Tsutomu et al., 2010).

In this study, phenol was chosen as the target pollutant, and mists were produced by evaporation of phenol solutions. A link tooth wheel-cylinder non-thermal plasma reactor was constructed to investigate phenol degradation under discharge and its decomposition efficiency, TOC removal and byproduct formation.

2. Experimental section

A schematic diagram of the experimental system is shown in Fig. 1. The system consists of a link tooth wheel-cylinder plasma reactor with a 50 kV/10 mA positive DC high-voltage power supply, mist-formation apparatus, and equipment for gaseous components analysis. A stainless steel cylinder with the inner diameter of

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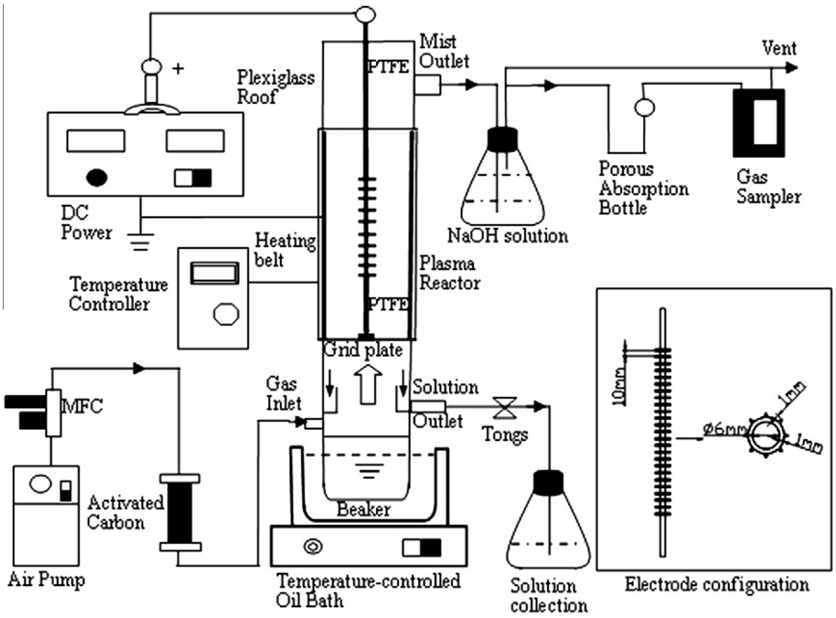

Fig. 1. Schematic diagram of the experimental setup.

39 mm and length of 490 mm was used as the ground electrode, and a stainless steel rod (od = 6 mm), through which 25 discharge tooth wheels (Fig. 1) were linked at intervals of 10 mm, was used as the high-voltage electrode.

Vapor was produced by evaporating 100 mL of phenol solution (100 mg L^{-1}) or ultrapure water in an oil bath controlled at 150 °C. The average evaporation rate of phenol solution or ultrapure water was around 0.25 mL min^{-1}. The generated vapor was diluted by up-flowing air with a flow rate of 2 L min^{-1}, producing a rising mist. To decrease mist condensation, the exterior of the plasma reactor was wrapped in a heating band maintained by a temperature controller at 60 °C.

When the phenol mist flowed into the plasma discharge zone, some of the mist droplets were trapped by the reactor and gathered by the collecting tank as a trapped solution. The outlet mist was absorbed using NaOH solution (0.1 M, 100 mL), and then exhausted. The mist passed once through the discharge zone, the residence time being 9.3 s, and the discharge duration of each experiment was 3 h. Ultrapure water was used to wash the inner surface of the reactor and the washing solution was collected. Phenol, the TOC concentration in the initial phenol solution before an experiment, and the residual phenol solution in the beaker, trapped solution, absorbed solution, and washing solution after the experiment were determined.

Phenol and gaseous ozone concentrations were respectively measured by 4-aminoantipyrine spectrophotometry (MEPPRC, 1987, 1999) and sodium indigotin disulfonate spectrophotometry (MEPPRC, 1995). A TOC-V_{CPH} TOC analyzer (TC-IC mode, Shimadzu, Kyoto, Japan) was used to determine the TOC concentration. The pH values, conductivities, and colors of the trapped solutions were monitored by an ion analyzer (PXSJ-216, Shanghai Precision & Scientific Instrument, China), a conductivity meter (DDB-303A, Shanghai Precision & Scientific Instrument, China), and a spectrophotometer (722S, Shanghai Precision & Scientific Instrument, China) at 455 nm (APHA, 1995), respectively. A GC-MS (QP2010; Shimadzu) was used to identify the organic byproducts in the trapped solution, esterified by *n*-butyl alcohol. The GC-MS conditions are as follows: a DB-5 column (60.0 m × 0.25 mm × 0.25 μm) was used; the programmed temperature was kept at 70 °C for 1 min and then raised at a rate of 20 °C min^{-1} to 230 °C and held for 2 min; the flow rate of the carrier gas (He) was 1 mL min^{-1}, and the sample was injected in a split mode (the split ratio was 50), with each injection being 1 μL. Quantification of the organic acids in the trapped solutions was carried out by ion chromatography (IC, ICS200, Dionex, Sunnyvale, CA, USA).

The phenol decomposition (η_{PH}) and TOC removal (η_{TOC}) are calculated as follows:

$$\eta_{PH} = \frac{m_0 - m_R - m_C - m_W - m_A}{m_0 - m_R}(100\%) \qquad (1)$$

$$\eta_{TOC} = \frac{m_{TOC0} - m_{TOCR} - m_{TOCC} - m_{TOCW} - m_{TOCA}}{m_{TOC0} - m_{TOCR}}(100\%) \qquad (2)$$

where m_0, m_R, m_C, m_W, and m_A are the masses of phenol in the initial phenol solution, in the residual phenol solution, in the trapped solution, washing solution, and absorbed solution; m_{TOC0}, m_{TOCR}, m_{TOCC}, m_{TOCW}, and m_{TOCA} are the corresponding TOC masses.

Energy consumption is related with the life span cost which is an important parameter in assessing the capability of non-thermal plasma reactor for practical applications. In this investigation, input power (W) was selected to value the power deposited into the reaction mist. Voltage and current can be automatically detected by the power supply equipment.

3. Results and discussion

3.1. Discharge characteristics and ozone formation

To check the discharge reliability of the non-thermal plasma and the effects of the gaseous conditions on discharge, the discharge voltage–current characteristics of the reactor, and ozone formation, were investigated under conditions of airflow only, mist produced by ultrapure water, and mist produced by a phenol solution. The results are shown in Fig. 2.

As the figure shows, when discharge occurred in the airflow condition, the corona starting voltage was 8.6 kV, and spark discharge occurred when the applied voltage reached 12.2 kV. However, when mist was introduced into the reactor, the corona starting voltage increased to 10.0 kV and the spark discharge voltages in the mist with and without phenol were 15.9 and 15.1 kV,