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Effects of electrode geometry on the performance of dielectric barrier/packed-bed discharge plasmas in benzene degradation

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HIGHLIGHTS

- Benzene was successfully degraded by dielectric barrier/packed-bed discharge plasmas.
- Different electrode geometry has distinct effect on plasmas oxidation performance.
- Benzene degradation and energy performance were enhanced when using the coil electrode.
- The reaction products were well determined by online FTIR analysis.

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ABSTRACT

In this study, the effects of electrode geometry on benzene degradation in a dielectric barrier/packed-bed discharge plasma reactor with different electrodes were systematically investigated. Three electrodes were employed in the experiments, these were coil, bolt, and rod geometries. The reactor using the coil electrode showed better performance in reducing the dielectric loss in the barrier compared to that using the bolt or rod electrodes. In the case of the coil electrode, both the benzene degradation efficiency and energy yield were higher than those for the other electrodes, which can be attributed to the increased role of surface mediated reactions. Irrespective of the electrode geometry, the packed-bed discharge plasma was superior to the dielectric barrier discharge plasma in benzene degradation at any specific applied voltage. The main gaseous products of benzene degradation were CO, CO_2, H_2O, and formic acid. Discharge products such as O_3, N_2O, N_2O_5, and HNO_3 were also detected in the outlet gas. Moreover, the presence of benzene inhibited the formation of ozone because of the competing reaction of oxygen atoms with benzene. This study is expected to offer an optimized approach combining dielectric barrier discharge and packed-bed discharge to improve the degradation of gaseous pollutants.

1. Introduction

Benzene is a toxic and stable volatile organic compound (VOC). It is used extensively as a reagent in the production of a wide variety of chemical substances such as dyes, detergents, plastics, pesticides, adhesives, and coatings [1,2]. When benzene diffuses into the environment, it poses serious hazards to ecological system and human health [3]. At ground level, benzene can react with other air pollutants which lead to the formation of potentially harmful concentrations of photochemical smog and secondary aerosol in troposphere [4].

Several conventional techniques such as adsorption [5], bio-filtration [6], and catalytic oxidation [7,8] have been used to degrade benzene in the gas phase. However, these techniques possess some inherent drawbacks. As for adsorption methods, disposal costs of the spent adsorbent must be taken into consideration. Bio-filtration methods are time-consuming and require keeping rigorous environmental conditions for optimum microbial growth. High temperature and energy consumption are required for catalytic oxidation methods. An alternative approach to these conventional methods is to use non-thermal plasma (NTP) at ambient temperature. In NTP, energetic electrons induce excitation, ionization, and dissociation of background gas molecules (N_2, O_2, and H_2O), producing energetic electrons, photons, ions, and radicals [9,10]. These reactive species are responsible for the destruction of VOC molecules.

As previously reported [11], an improved plasma reactor, which generates dielectric barrier/packed-bed discharge plasmas, was

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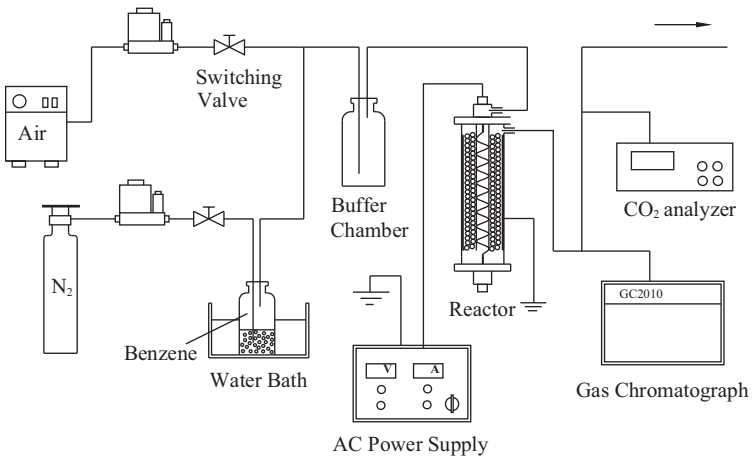

Fig. 1. Schematic diagram of the experimental system.

employed to enhance the degradation of benzene, and remarkably better benzene degradation and energy performance were obtained than in dielectric barrier or packed-bed discharge reactors alone. In this type of reactor, the energy deposited in the discharge is used preferentially to form energetic electrons and reactive species instead of species with low activity. However, the optimization of experimental parameters is still needed to further improve the benzene degradation and energy use, which mainly depend on gas properties, discharge conditions, and reactor configurations [12–14]. Hereinto, electrode geometry is one of the key structure parameters that remarkably affect the discharge characteristics and the energy distribution in a plasma reactor. Accordingly, the purpose of this study is to clarify the influences of electrode geometry on such a hybrid plasma reactor, with the goal of maximizing benzene degradation and energy use.

In the present study, the effects of electrode geometry on benzene degradation were investigated in a dielectric barrier/packed–bed discharge plasma reactor with three electrode geometries: rod, bolt, and coil. The results were evaluated in terms of discharge characteristics, benzene degradation efficiency, energy yield, CO_x selectivity, and the formation of degradation products and discharge byproducts. The detailed degradation process in different discharge regions of the plasma reactor was also investigated.

2. Experimental

2.1. Experimental setup

The schematic diagram of the experimental apparatus is shown in Fig. 1. It consisted primarily of a dielectric barrier/packed-bed discharge plasma reactor, a benzene feeding system, a high-voltage alternating current (AC) power supply (0–35 kV, 50 Hz), and a series of analytical instruments. Fig. 2 shows the schematic diagram of the plasma reactor with various electrode configurations. The reactor was made of a Plexiglas cylinder (35 mm inner diameter × 280 mm) with a quartz tube (14 mm inner diameter × 250 mm) fixed along its axis as the dielectric barrier. A stainless-steel mesh (190 mm) that was wrapped around the inner surface of the reactor served as the ground electrode. A stainless-steel rod (6 mm diameter), a stainless-steel bolt (6 mm diameter), and a roll of stainless-steel wire (1 mm diameter) were employed as the discharge electrode. The open area between the quartz tube and the ground electrode was filled with glass beads (3 ± 0.3 mm). In the case of the rod/bolt electrode, when the AC power is applied, a large number of filaments are formed between the discharge electrode and the quartz tube. Simultaneously, a mass of microdischarges are generated at the contact points between the glass beads. These hybrid plasmas are named “volume/packed-bed plasmas”. In the case of the coil electrode, a stable surface discharge starts from the peripheral edges of the coil electrode and stretches out along the surface of the quartz tube. The packed-bed discharge is also formed when using the rod/bolt electrode. These hybrid plasmas are named “surface/packed-bed plasmas”. In this reactor, the discharge region inside the quartz tube is defined as the dielectric barrier discharge region (part I), and the region between the quartz tube and the ground electrode is defined as the packed-bed discharge region (part II) (Fig. 2). The stimulated gas enters the reactor via the gas inlet 1, traverses part I and II in turn, and then exits via the gas outlet 2.

2.2. Experimental conditions

The benzene vapor was evaporated by bubbling it with nitrogen in a water bath (293 K) and diluted by a stream of compressed air to give the desired initial concentration and flow rate (400 ppm of benzene, 0.5 L min^{-1}). The benzene concentration was measured on a gas chromatograph (Shimadzu GC-2010), equipped with a flame ionization detector (FID). Qualitative analysis of the outlet gas was performed using a Fourier transform infrared spectroscopy (FTIR, Nicolet 6700), equipped with a DTGS detector and a 2.4 m gas cell with KBr windows. The O_3 concentration was performed by the iodometric method [15]. CO and CO_2 were quantified using a gas chromatograph (SRI-8610C), equipped with a FID and a methane conversion furnace (GS-101A). The CO, CO_2 and CO_x selectivities are calculated as followed:

$$S_{CO}(\%) = \frac{[CO]}{6([benzene]_{in} - [benzene]_{out})} \times 100 \quad (1)$$

$$S_{CO_2}(\%) = \frac{[CO_2]}{6([benzene]_{in} - [benzene]_{out})} \times 100 \quad (2)$$

$$S_{CO_x}(\%) = S_{CO} + S_{CO_2} \quad (3)$$

where S_{CO}, S_{CO_2}, and S_{CO_x} represent the CO, CO_2, and CO_x selectivities, respectively. [CO] and [CO_2] are the CO and CO_2 outlet concentrations, respectively.

2.3. Electrical measurements

The applied voltage and current were monitored with a digital oscilloscope (Tektronix TDS2024), a voltage probe (EP-50K 5054),