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Experimental study of acetone removal by packed-bed dielectric barrier discharge reactor

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ABSTRACT

In this study, a packed-bed dielectric barrier discharge (DBD) reactor was developed for acetone removal. The effects of packing materials were investigated in terms of discharge characteristics, removal efficiency and byproducts formation. The identification of organic byproducts was conducted. The results indicated the presence of packing materials enhanced the energy density and exhibited a positive effect on acetone removal, while high oxygen concentration inhibited the removal of acetone. The formations of byproducts were significantly reduced in the presence of packing materials. The degradation pathways and mechanisms were discussed, showing CH_3 radicals played a crucial role in the acetone degradation process.

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1. Introduction

In recent years, abatement of volatile organic compounds (VOCs) has attracted great interest since VOC emissions have an severe impact on global environment and may cause people's health problems even at low concentration [1,2]. Great efforts have been devoted to the abatement of VOCs by means of adsorption [3], catalytic combustion [4], condensation [5], photo-catalytic process [6] and biodegradation [7]. However, these conventional methods may not be able to deal with low-concentration VOCs cost-effectively and meet the restrict emission standards. Non-thermal plasma (NTP) has been demonstrated as an alternative technology for removal of various kinds of air pollutants such as SO_2, NO_x and particulate matters (PM) under ambient conditions over last two decades [8–10]. In NTP, the energy dissipated to the system is mainly used to generate energetic electrons, while the background gas remains at ambient temperature. These energetic electrons collide, excite, dissociate and ionize background gas molecules as well as pollutants, producing active species such as secondary electrons, ions and radicals.

The common methods for NTP generation include dielectric barrier discharge (DBD), corona discharge, surface discharge and microwave discharge, etc. Among these methods, DBD has been widely investigated in VOC control due to its high electron density within the discharge channels which is beneficial for VOC treatment [11,12]. VOC removal is generally attributed to the electron impact and radical reactions between reactive species and pollutant molecules [13].

However, the main challenges of NTP technology for the removal of gas pollutants in waste gas streams are the formation of unwanted byproducts and low energy efficiency [14,15]. Researchers have been attempting to combine NTP technology with catalysis to overcome these problems. Up to now, studies on plasma-catalysis system mainly focused on the synergy effect, energy efficiency improvement and byproducts formation inhibition [16–19]. Only few reports demonstrated the mechanisms and degradation pathways of VOC abatement in plasma-catalysis systems. Investigation on the contribution of dominant reactive species in the NTP discharge region may greatly contribute to understanding the underlying mechanisms and lead to a better industrial application of NTP technology.

Among the various VOC molecules, acetone was selected as the model VOC since it's one of the most abundant oxygenated VOC in the air. The major sources of acetone emission include chemical production processes, vehicle emissions, and it is also widely used as solvent in cleaning and painting. A few researchers reported acetone removal with NTP based technologies. Lyulyukin et al. [20,21] investigated acetone decomposition in negative corona discharge and proposed simplified decomposition pathways with products, including HCHO, CO and CO_2. Great efforts have been

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devoted to acetone decomposition in packed DBD reactors. The introduction of packing materials modified the electron energy distribution and resulted in a better removal of acetone and CO_2 selectivity [22,23].

In the present study, a DBD plasma system was designed and built for the removal of low concentration acetone in gas stream. Three kinds of packing materials were employed. The characteristics of acetone removal with and without packing materials were compared in the aspects of discharge characteristic, removal rate, energy efficiency, oxygen concentration and byproducts formations. Identifications of decomposition products and organic byproducts were conducted. Finally, reaction mechanisms and pathways in the DBD plasma reactor were proposed based on the identified decomposition products.

2. Experimental description

2.1. Experimental setup

The schematic diagram of the experimental setup is illustrated in Fig. 1. It consisted of a reaction gas feeding system, a DBD reactor with high-voltage AC power supply (0–40 kV, 50 Hz, sine wave) and a chemical analysis instrumentation. For all experiments, gaseous acetone was acquired by feeding a dry nitrogen stream through a bubbler kept in a water bath (0 °C). Carrier gases (N_2 and O_2) were mixed with gaseous acetone in a mixing chamber before introduced into the DBD reactor. Each flow of different gases was regulated by a set of mass flow controllers (DB-07, Sevenstars). Total flow rate for all experiments was regulated at 200 mL/min, while the initial concentration of acetone was kept at 100 ppm unless otherwise mentioned.

The DBD reactor was a concentric cylinder, which had a cylindrical quartz tube with an inner diameter of 20 mm and wall thickness of 2.5 mm. The quartz tube was wrapped by a 200-mm long copper mesh which acted as a ground electrode. Both ends of the quartz tube were sealed with insulators made of PTFE. Through hole was made on the center of each insulator to hold the inner discharge electrode. The inner discharge electrode was a stainless steel rod (16 mm in diameter). It was placed on the axis of the reactor and connected to the high-voltage output of the AC power supply. The discharge was limited in the space between the inner electrode and quartz tube with a discharge gap of 2 mm width. The discharge volume was 22.6 cm^3 with a corresponding gas residence time of 6.8 s. The packing materials employed in this study were glass pellets, α-Al_2O_3 and γ-Al_2O_3 with the same diameter of 1 mm. The relative permittivity was 3.9, 9.5 and 12.6 for glass pellets, α-Al_2O_3 and γ-Al_2O_3, respectively, while the specific surface area for the three packing materials was 48, 53 and 174 m^2/g. The packing materials were placed in the discharge zone and held by glass pellets in the regions between the edges of discharge zone and the insulators. The outlet acetone concentrations in blank tube reactor and glass packed reactor were the same at a given inlet acetone concentration. The packing method was acceptable since no significant adsorption effects were observed for the glass pellets packed in the regions between insulators and discharge zone.

2.2. Electrical measurements

All the experiments were carried out at room temperature and ambient pressure. The discharge was generated after the outlet concentration of acetone reached a steady state. The discharge voltage and current waveforms were monitored by an oscilloscope (Tektronix 3034B, USA). High voltage applied to the reactor was measured with a 1000:1 voltage probe (Tektronix, P6015A). A 1000:1 capacitor was connected in series between the reactor and the ground electrode. The total charge was calculated by measuring the voltage across the capacitor with a probe (Tektronix P5100). V-Q Lissajous method was used to determine the power consumption. Total power consumption of the reactor is proportional to the area of the Lissajous diagram and can be calculated as:

$$P = f \cdot C_m \cdot A \tag{1}$$

where C_m is the 1 μF measuring capacitance, f is the AC frequency and A is the area of the Lissajous diagram.

The specific input energy (SED) defined as energy dissipated to per unit volume of the gas stream is expressed as follows:

$$\mathrm{SED\ (J/L)} = \frac{P\ (\mathrm{W})}{Q\ (\mathrm{L/min})} \times 60 \tag{2}$$

where Q denotes the total flow rate (L/min).

2.3. Chemical analysis

The inlet and outlet concentrations of acetone were analyzed on-line by a gas chromatograph (Agilent 7890A, USA) equipped with a flame ionization detector (FID) and a capillary column of HP-Innowax (Agilent, USA) operated at 60 °C. The acetone removal efficiency ($\eta_{actetone}$) is calculated as follows:

$$\eta_{acerone}\ (\%) = \frac{c_{in} - c_{out}}{c_{in}} \times 100\% \tag{3}$$

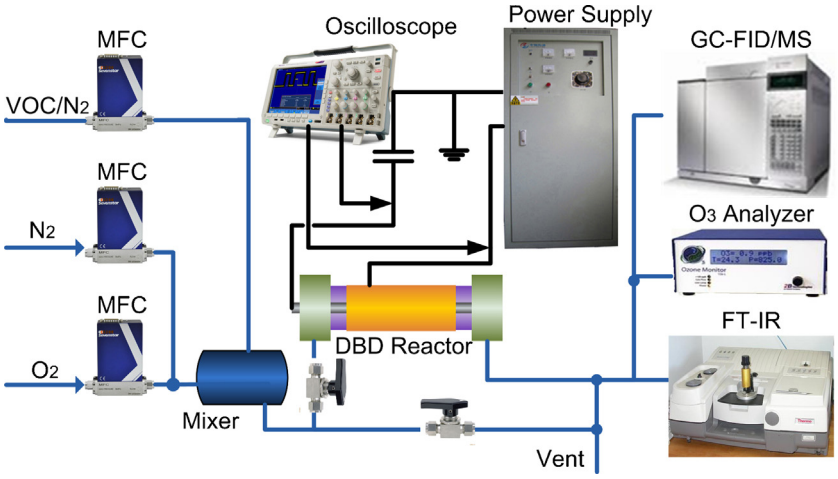

Fig. 1. Schematic diagram of the experimental setup.