



Oxidation of toluene by dielectric barrier discharge with photo-catalytic electrode



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HIGHLIGHTS

- A DBD reactor with photo-catalytic electrode was adopted to remove toluene.
- Photo-catalytic electrode improved toluene conversion dramatically.
- The reactor had good performance in mineralization rate and CO₂ selectivity.

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ABSTRACT

A specially designed photo-catalytic electrode-incorporated-dielectric barrier discharge (DBD) reactor has been used to investigate the interaction between photo-catalytic electrode and DBD. Toluene conversion by oxidation was enhanced dramatically due to the synergy between DBD and photo-catalytic electrode. The photo-catalytic electrode could be fully activated in the non-thermal plasma at high peak voltages without extra UV light source. Peak voltage, initial concentration, residence time and humidity were important factors that influenced the toluene conversion. 160 mg/m³ toluene conversion reached 89.9% at a peak voltage of 40 kV and with a residence time of 4 s in the reactor photo-catalytic electrode. Photo-catalytic electrode could improve DBD system's water-resistance by turning more water molecules into OH radicals. And it also made the reactor have better performance in mineralization rate and CO₂ selectivity.

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1. Introduction

Volatile organic compounds (VOCs) are considered to be primary toxic air pollutants due to their detrimental effects on both human health and environment [1]. Toluene is a typical toxic VOC. It is an important organic chemical material with a pungent odor [2,3]. It is also often used as ingredient for paints and diluents. The inhalation of toluene can cause human nerve system damage [4].

Traditional VOCs control methods, such as wet scrubbing, active carbon adsorption, ozone oxidation and bio-filtration, are limited technically and economically [5,6]. Non-thermal plasma (NTP) technique application in VOCs control has been receiving increasing attention because of good economy and easy operation [7,8]. Many studies on removal of VOCs by NTP have been performed under different conditions such as target molecules, gas composition, reactor type and geometry, type of power supply, etc. [9–20]. For the practical application, it is also important to control the product of the

VOCs after treatment by NTP. However, some studies reported poor mineralization efficiencies of VOCs decomposed by NTP alone. Wallis et al. [21] measured no CO₂ during the destruction of 500 ppm dichloromethane when using only plasma. Kim et al. [22] found the CO₂ selectivity was 26.7% when benzene was decomposed by pulsed corona. Combining NTP with other pollution control techniques to overcome the drawbacks was becoming a new focus of researches [23–29]. Karuppiah et al. [30] oxidized diluted benzene by a dielectric barrier discharge (DBD) reactor with sintered metal fiber (SMF) electrode modified with transition metal oxides. For 50 ppm of benzene, the reactor with TiO₂/MnO_x/SMF showed 100% conversion at 230 J/L. Thevenet et al. [31] thought the interaction between the plasma and the material porosity (silica fibers) clearly enhances the elimination of acetylene at atmospheric pressure. And the mineralization into CO and CO₂ could be improved by additional irradiation of the photo-catalyst. Subrahmanyam et al. [32,33] used a DBD reactor with SMF electrode modified with Ti, Mn and Co to oxidize VOCs (toluene, isopropanol and trichloroethylene). With CoO_x and MnO_x/SMF catalytic electrodes, 100% conversion of toluene was achieved at 235 J/L. However, catalyst

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was prone to being poisoned. And the research on stabilization of SMF electrode modified with catalyst was rare.

In this study, toluene was decomposed by a DBD reactor. SMF coated with TiO_2 was used as discharging electrode. The synergy between DBD and photo-catalytic electrode was explored. Special consideration was given to some important parameters that may affect toluene conversion, including peak voltage, initial concentration, residence time and humidity. The mineralization rate and CO_2 selectivity of toluene decomposed in various reactors were studied. A long-term experiment (25 d) was operated to investigate the stability of the photo-catalytic electrode.

2. Experimental

2.1. Experimental setup and reactor

Fig. 1 showed a schematic diagram of the experimental setup. Dry air from a gas cylinder was allowed into a buffer tank at a controlled flow rate through tubes. A fraction of the gas flow went through a toluene generator to carry out toluene with an appropriate level. For experiments with humid air, a portion of the air flow went through a bubbler filled with deionized water to an appropriate level. Then all the streams were mixed in the buffer tank, where humidity was measured by a hygrometer (Rotronic A1H). At last, the mixed stream was introduced to the DBD reactor. Samples of the treated gas for on-line chemical analyses were allowed into a gas chromatography (GC) from the sampling ports located at the inlet and the outlet of the reactor. The DBD reactor was a 24 mm (inner diameter) \times 200 mm quartz cylinder with a SMF wire of 1.5 mm in diameter fixed along its axis. The SMF wire served as the support of TiO_2 and also an energized electrode. The ground electrode was a 150 mm copper film which was embedded outside of quartz cylinder. An AC high voltage source with frequency of 20 kHz was applied in the experiment. The voltage and current waveforms were measured by using a four channel Tecktronix TDS 2014B 350 MHz digital storage oscilloscope capable of sampling 1 GS/s (Giga Sample per second), a Texas HVP-3020 high voltage passive probe and a CT4 TCP202 current probe.

2.2. Materials

Air (Jingong Co., Ltd.) was a synthetic mixture (80% nitrogen and 20% oxygen) from pure liquid nitrogen and pure liquid oxygen with

specified impurities of H_2O . The purity of toluene (Huadong Medicine Limited CO. LTD) was 99.9%. The concentration of anatase nano- TiO_2 solution (VK-TG02, Wangjing Material Co. Ltd., China) was 10%. SMF (average pore radius of 5 μm , porosity of 65% and diameter of 100 μm) was supplied by Xi'an Filter Metal Materials Co. Ltd., China. And the $\text{TiO}_2/\text{SMF}-n$ electrode was prepared by a modified anodic oxidation method [24]. UV light was applied by a 125 W high-pressure mercury lamp (Shanghai Yamingdianqi Zhaoming Dianqi Limited CO. LTD) with wave length of 365 nm.

2.3. Experimental procedure

The following standard procedure was developed to achieve stable and reproducible conditions for the gas flow rate and composition. The reactor was first flushed with air at 1 L/min for a few minutes. Toluene-containing gas was then allowed in at an appropriate flow rate for 30 min, at which point the toluene concentration measured at the reactor outlet was the same as that at its entry. The desired flow rate for the specific experiment was then set and the high voltage was applied. After a stabilization time of about 5 residence times to obtain a steady concentration of the residual odor at the reactor outlet, two consecutive samples of the treated gas were allowed to GC analysis. After a group of experiments was completed, the gas flow was stopped and the high voltage source was switched off. The apparatus was finally cleaned by flushing air at 1 L/min for 15 min. In experiments with humidified gas, preconditioning of the reactor was achieved by flushing humidified air through it for 30 min. Then, the toluene-containing gas, humidified as described above, was introduced into the system. All the experiments above were achieved at ambient temperature.

2.4. Chemical analyses

The concentration of toluene was measured by the gas chromatograph GC7890II (Tianmei corporation) which was equipped with 6-port gas sample valve. Ozone formed in the DBD reactor was measured by an UV absorption ozone monitor (AP1-450 NEMA). The formation of CO and CO_2 was monitored with an infrared gas analyzer (Siemens ultramat 23). A Fourier transform infrared (FTIR) (Ncolet6700) was used for gas qualitative analysis.

OH radical in the DBD reactor was detected by salicylic acid method [34]. A film, which was impregnated with salicylic acid,

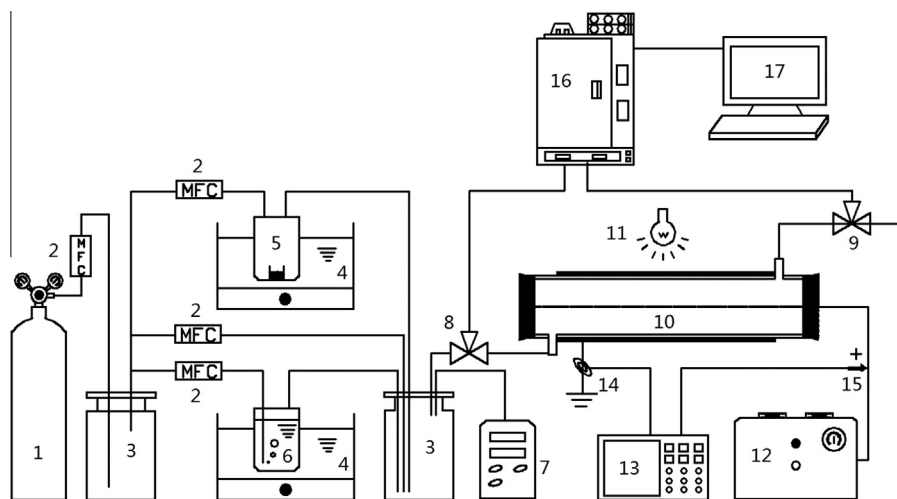


Fig. 1. Schematic diagram of the experimental setup. (1) Gas cylinder; (2) mass flow control; (3) buffer tank; (4) water bath tank; (5) toluene generator; (6) water vapor generator; (7) hygrometer; (8) sampling of inlet gas; (9) sampling of outlet gas; (10) DBD reactor; (11) UV lamp; (12) high voltage source; (13) digital storage oscilloscope; (14) current probe; (15) high voltage probe; (16) gas chromatograph; (17) computer.

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