



Toluene gas treatment by combination of ionic liquid absorption and photocatalytic oxidation



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ABSTRACT

Toluene exhaust gas is a nuisance to the environment and human beings. In this study, 1-dodecyl-3-methylimidazolium chloride (DDMIM Cl) was selected as the absorbent solution and was combined with photocatalytic oxidation (PCO) for the treatment of toluene gas. The effects of toluene concentration, UV lamp power, catalyst dosage, coexisting ions and pH on the toluene removal ratio by PCO were investigated. Changes to the absorbent structure after four reuses were compared according to the UV–vis absorption spectrum, and the anti-oxidation ability of the absorbent was evaluated. The results showed that the absorbent concentration was an important factor in the absorption of toluene. At the absorbent concentration of 5%, the initial absorptivity reached 96.79%, and the saturated absorption capacity was 43.8 mg/L. With a toluene concentration of 13.1 mg/L, an 18-W UV lamp, a photo catalyst dosage of 400 mg/L, and a reaction time of 80 min, the removal ratio of toluene reached 91.3%. The PCO of toluene followed pseudo-first-order kinetics. The main intermediates of toluene oxidation were benzoic acid and benzaldehyde, while traces of cresol and benzyl alcohol were also found. After four reuses, the absorption capacity of the absorbent was not weakened, and the molecular structure of DDMIM Cl remained stable, reflecting its oxidation resistance. Therefore, the use of an ionic liquid as an absorption solution combined with PCO for the treatment of toluene waste gas is theoretically feasible.

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1. Introduction

Toluene exhaust gas emitted during production processes in the chemical, ceramic, paint, electroplating, and other industries greatly harms employee health [1–3]. Currently, combustion and adsorption methods are used to treat toluene gas [4]. Of these, the combustion method has a greater temperature requirement for the exhausted gas, with a direct combustion that should occur at temperatures above 1100 °C. The investment and operation costs of this method are also great [5]. The adsorption method has a better removal ratio of toluene exhaust gas at low concentrations, although it has some problems, such as adsorbent regeneration and the inability to reduce contaminants to harmless levels [6]. Photocatalytic oxidation (PCO) is considered a promising method for the treatment of toluene exhaust gas owing to its powerful catalytic activity, mild working conditions, and safe environment [7–9]. However, the removal ratio of the exhaust gas is restricted by

the contact reaction time and the toluene concentration on the surface area of the photo catalyst [10]. By adding absorbents that enhance the solubilization of toluene in the solution, absorption can make up for the shortcomings of PCO [11]. Thus, absorption combined with PCO should be feasible for the treatment of toluene exhaust gas.

Ionic liquids (ILs), composed of an organic cation and an organic or inorganic anion, are molten salts at room temperature, and imidazoles are the most common cations [12]. They have many unusual properties, such as high thermal and chemical stability and excellent solvability for inorganic and organic compounds [13]. In recent years, many researchers have reported treating inorganic and organic gases using ILs [14–18], but rarely treating toluene exhaust gas. By analyzing Henry's constant of toluene over 272 types of ILs using the COSMO-RS method, Bedia et al. [19] found that imidazole ionic liquid with long-chain alkanes in the imidazolium cations had better toluene absorption capabilities. Stepnowski et al. [20] noted that elongating the substituent length in the imidazolium cations could significantly decrease degradability. Siedlecka et al. [21] observed that Cl[−] was able to compete

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for hydroxyl radicals with BMIM⁺, and the degradation of ionic liquids by PCO could be inhibited.

Therefore, in this study, 3-methyl, 1-dodecyl imidazole chloride (DDMIM Cl) was selected as an absorbent to treat simulated toluene gas, and the influence of its concentration on absorption was assessed. Further, the effects of toluene concentration, UV lamp power, photo catalyst amount, system pH, and coexisting ions on the PCO process were investigated. Oxidation mechanisms of toluene were determined based on the identification of intermediates. The oxidation resistance of the absorbent was also investigated.

2. Experimental design and methods

2.1. Materials

Toluene (purity: 99.8%) was purchased from Sigma-Aldrich, DDMIM Cl (purity: 97–98%), was purchased from Shanghai Chengjie Chemical Co. without further purification, and the photo catalyst (80% anatase TiO₂, particle diameter ≈ 15–20 nm, surface area ≈ 60–100 m²/g) was purchased from Hangzhou Wan Jing New Material Company.

2.2. Experimental device

A schematic diagram of the absorption and PCO processes is depicted in Fig. 1. The inner diameter of the reaction vessel was 4.5 cm; its height, 30 cm; and working volume, 300 mL. An 18-W ultraviolet (UV) lamp with a radiation peak at 254 nm (Fujitsu, Japan) was placed in the middle of the vessel for illumination. The reaction vessel was shielded with tinfoil for protection against UV rays. The reaction temperature was controlled at 25 ± 1 °C via a water bath.

The inlet gas feed was provided via bubbling a bottle of compressed dry air through liquid toluene in the glass cylinder. The air flux containing toluene was mixed with air, forming a simulated gas with the desired toluene concentration. It was then fed into the reaction vessel, which was filled with 200 mL of absorption solution, through a gas disperser. In this study, the total flow ratio of gas measured and controlled with rotary flow meters was fixed to 100 mL/min, and the toluene concentration of the inlet gas was approximately 1000 mg/m³.

2.3. Toluene absorption experiments

The absorption solutions were prepared according to varying weight ratios of DDMIM Cl to water (0, 0.01%, 0.02%, 0.05%, 0.1%,

0.2%, 0.5%, 1%, 2%, and 5%). During the experiment, the UV lamp was turned off. The toluene concentrations of the inlet and outlet gas were measured once every 10 min by a ppbRAE monitor (PGM 7340, RAE Systems, USA) [22,23]. When the toluene concentration of outlet gas was equal to that of the inlet gas, the absorption solution reaches its saturation toluene absorption, its toluene concentration was determined from the absorbance of the peak maximum at 261 nm on a Lambda 850 UV–vis spectrophotometer [24]. Repetition experiments were performed 3 times.

Toluene absorptivity (A, %) at a certain absorption time was calculated as follows:

$$A = \frac{Y_0 - Y_g}{Y_0} \times 100\% \quad (1)$$

where Y_0 (mg/m³) and Y_g (mg/m³) are toluene concentrations of the inlet and outlet gas, respectively.

2.4. PCO of toluene in absorption solution

The two rotary flow meters were turned off, and a certain amount of photo catalyst was added to the saturated toluene absorption solution. Prior to the PCO experiments, the suspension was stirred for more than 1 h in the dark, ensuring full adsorption of toluene on the photo catalyst [25]. When the UV lamp was switched on, air was bubbled continuously into the suspension at low speed (20 mL/min) by opening rotary flow meter 1 during the runs. Aliquots of 10 mL were collected every 20 min and were centrifuged before being analyzed by the ultraviolet absorption method. Each analyzed sample was returned to the reaction vessel after the test [26]. Blank tests were performed in the absence of the photo catalyst and illumination by the UV lamp.

After their extraction by dichloromethane, the intermediate oxidation products of toluene were analyzed by GC–MS (Agilent 7890A) equipped with a DB-WAX column (30 m × 0.25 μm × 0.25 mm) and a flame ionization detector. The primary temperature of 40 °C was maintained for 1 min, after which the temperature was increased by 10 °C/min to 200 °C. The temperature was again raised by 8 °C/min to a final temperature of 240 °C. The sample was held at the final temperature for 5 min. The MS operating conditions included an ion source at 200 °C, full scan mode, and the MS mass range of m/z 35–450 with an acquisition ratio of 200 spectra per second. The electron impact (EI) ionization source at –70 eV was used after a 4-min solvent delay. Intermediates were identified using a program of the NIST Library.

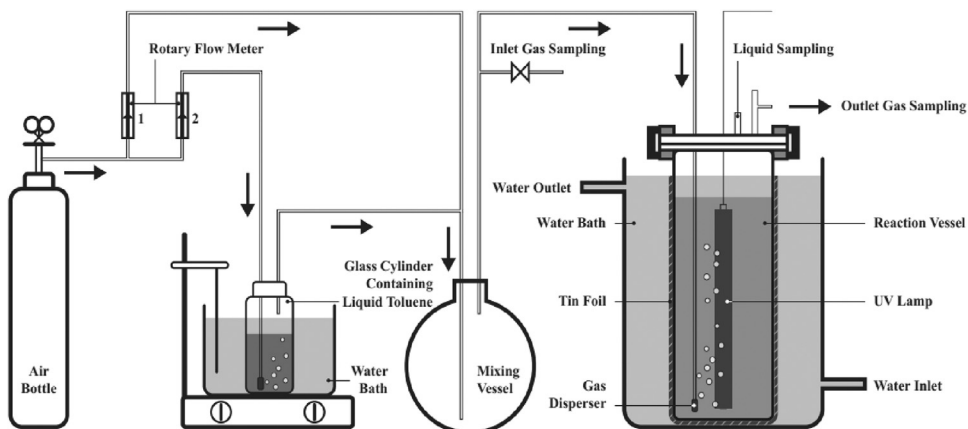


Fig. 1. A sketch map showing the lab-scale set-up.

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