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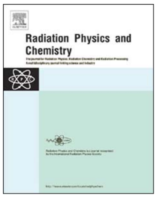

Treatment of toluene and its by-products using an electron beam/ultra-fine bubble hybrid system

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ABSTRACT

Although, until quite recently, many technologies (electron beam (EB), plasma, and ultraviolet) have been studied to overcome disadvantages of conventional methods (such as absorption, adsorption, biofiltration and incineration) for treatment of volatile organic compounds (VOCs), their techniques still have some problems such as formation of a by-product. Generally, it is reported that various by-products are generated from the EB irradiation process to remove VOCs. Therefore, we developed an electron beam/ultra-fine bubble (EB/UB) hybrid system to enhance removal efficiency of a VOC (toluene) and to reduce its by-products formed by electron beam irradiation. As a result, the removal efficiency of toluene (30 ppm) by only EB (10 kGy) was 80.1%. However, the removal efficiency of toluene using the hybrid system (water temperature: 5 °C) was increased up to 17% when compared to only EB (10 kGy). Additionally, the 65.2% of ozone formed from the EB process was removed in UB reactor. In case of other trace by-products such as undesired VOCs and aldehydes, the levels were lowered down to the below detection limit by the subsequent UB reactor. We also found that the amount of toluene collected and solubilized into water is affected by the water temperature in the UB reactor.

1. Introduction

Volatile organic compounds (VOCs) emitted from various sources such as combustion processes, painting shops, and petrochemical industries contribute to the formation of photochemical oxidants (O_3 and PAN), produce foul odors, and cause cancers and other harmful effects on human health and the environment (Chang et al., 2005; Duan et al., 2008; Finlayson-Pitts and Pitts, 1997; Khan and Ghoshal, 2000; Ling et al., 2011; Shareefdeen and Singh, 2005).

A lot of physical, chemical, and biological techniques (such as carbon adsorption, absorption, catalytic oxidation, thermal incineration, and biofiltration) have been researched to reduce the amount of VOCs emitted from industrial processes (Burgess et al., 2001; Smet et al., 1998; Wani et al., 1997). It is reported that their techniques have both strengths and weaknesses (Khan and Ghoshal, 2000; Kim et al., 2004, 2005; Son et al., 2011). In the case of absorption, product recovery can countervail the annual operating costs, but requires strict maintenance and pretreatment of the VOCs. For biofiltration, it requires less initial investment, less non-harmful, and non-hazardous by-products. However, it is slow and requires a mixed culture of microbes because selective microbes decompose selective organics (Khan and Ghoshal, 2000).

To settle these demerits, some researchers have developed new technologies using an electron beam (EB), plasma, and ultraviolet (UV) (Boulamanti et al., 2008; Chaichanawong et al., 2005; Chmielewski et al., 2002; Kim, 2002; Tanthapanichakoon et al., 2004). Since it is operated at ambient temperature and a short reaction time (10^{-8}–10^{-1} s), the EB among these techniques is acknowledged as an economical and energy efficient method (Hirota et al., 1995; Kim et al., 2005; Kim, 2002; Son et al., 2010a, 2010b, 2015a; Sun and Chmielewski, 2012). Therefore, a study using EB has been lively carried out to treat VOCs (acetaldehyde, benzene, butylacetate, ethylbenzene, styrene, toluene and xylene) (Kim, 2002, 2007; Son et al., 2010a, 2012, 2014, 2015a). In results of these studies, maximum removal efficiency (RE) of benzene was reported approximately 85% at 2 ppm (initial concentration) and 16 kGy (absorbed dose) (Kim, 2002). In case of styrene of 50 ppm, the RE was reached to 98% at 2.5 kGy (Son et al., 2012). For acetaldehyde, Son et al. reported that the RE was almost

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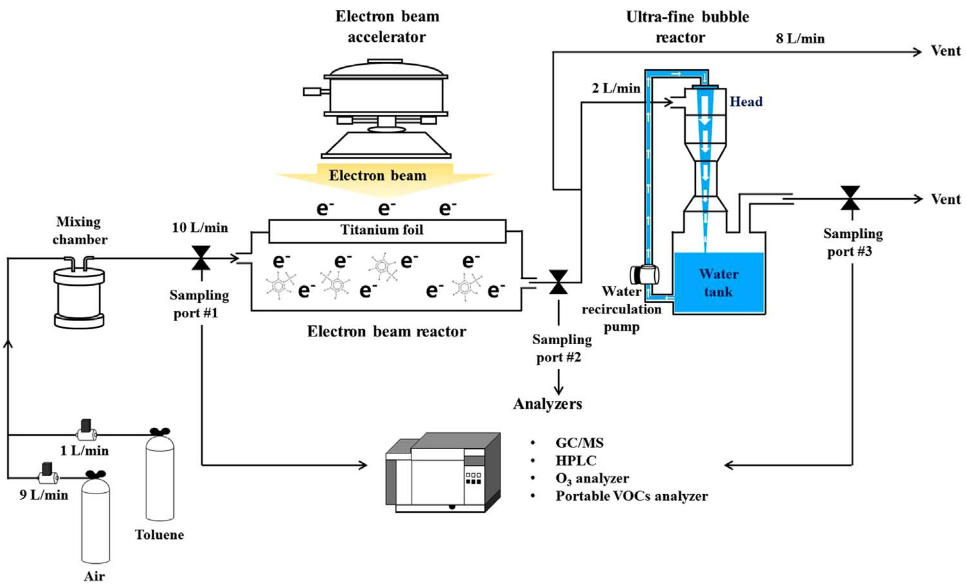

Fig. 1. Schematic diagram of an EB/UB hybrid system for the treatment of toluene and its by-products.

100% at condition of 34 ppm (initial concentration) at 10 kGy (Son et al., 2014). Kim (2002) also reported that absorbed dose of over 16 kGy would be required to obtain toluene RE of 90%. Nonetheless, there are some problems such as formation of by-products (aerosol, ozone and trace other VOCs and aldehydes) and low removal efficiency up to the present. To overcome these issues, diverse hybrid systems (EB/catalyst, EB/microwave/catalyst, and EB/scrubber) have been designed and researched. To increase the RE of VOCs, Various experiments on an EB/catalyst system have been conducted during the last 10 years (Kim, 2007; Kim et al., 2004, 2005, 2010; Son et al., 2010a, 2015a). On the other hand, Calinescu et al. (2009) developed an EB hybrid technology combined with a microwave and/or catalyst to reduce input energy. Son et al. (2015a) reported that the RE is increased and the input energy is decreased by the EB/scrubber process. However, their technologies have still some limitations from the viewpoint of a by-product reduction (trace gaseous compounds) because most of these researches are concentrated to improve the removal efficiency of an original target compound (excluded by-products) and energy efficiency of processes.

Accordingly, in this study, an EB/ultra-fine bubble (UB, which was designed to enhance the collection and dissolution of gaseous pollutants with water) hybrid system was developed to efficiently improve the RE of toluene and reduce the by-products formed from the EB irradiation process. To assess performance of the EB/UB hybrid system, we investigated the RE of toluene and its by-products (ozone and trace VOCs and aldehydes formed by EB irradiation) using the EB/UB hybrid system. Additionally, variations on the efficiency of the UB system with respect to the water temperature were explored.

2. Experimental methods

2.1. EB accelerator and reactor

In this study, a mobile EB accelerator of 0.6 MeV and maximum power 20 kW (ELV 0.6-33, Korea Atomic Energy Research Institute, Jeongeup-si, Korea) was used.

A cellulose triacetate (CTA) film dosimeter (FTR = 125, Fuji, Japan) was utilized to measure the absorbed dose of EB in a reactor, which was a round reactor (70 mm (D) × 55 mm (H), Volume: 0.212 L and retention time: 1.27 s) made of stainless steel. Absorbed dose means that the average energy imparted to matter per unit mass by ionizing radiation. The unit of absorbed dose is Gy (1 Gy = 1 J/kg) (Son and Kim, 2015). Installing a thin film (Titanium foil, 50 mm) on the upper part of the reactor allowed the EB to penetrate through the film. The absorbance of CTA films irradiated by EB was measured by an AUV/VIS spectrophotometer (UVIKONxs, SECOMAM, France) at a 280 nm wavelength within 2 h. More detailed information regarding the method for measurement of absorbed dose can be found in previous publications (Son et al., 2010b, 2012; Son and Kim, 2015). The mean (n = 6) and standard deviation of absorbance based on the location in the reactor were 1.1088 and 0.0257, respectively. At this time, the mean (n = 3) and standard deviation of absorbance by blank CTA films were 0.0930 and 0.0038, respectively. Based on the results of these measurements, the ranges of the absorbed dose were adjusted to be 2.5–10 kGy.

2.2. UB reactor

The UB reactor was developed to collect by-products generated from the decomposition process of toluene using EB. An UB reactor, a so-called ultra-fine bubble (diameter smaller than 1 μm) generator, was used to dissolve and collect residual toluene and by-products in a solution (water) in the form of ultra-fine bubbles. This device was designed based on two principles. The first one is that the dissolution and collection efficiency of gaseous compounds in water will be increased by increasing the bubble count and contact surface of gas and liquid. The second one is that the ultra-fine bubbles in water remain more stable for a longer period of time than the fine bubbles (diameter smaller than 100 μm) because the bubble rise velocity reduces as the bubble size decreases.

The UB reactor consisted of a gas inlet, head, water recirculation pump and water tank, as shown in Fig. 1. The flow rate of gas flowed into UB reactor was 2 L/min. The shape inside head was spiral to promote collision between the gas and liquid. The water in the reactor was recirculated by a pump (MHi202M, Wilo Pumps Ltd., Korea) for 20 min. The maximum flow rate of liquid recirculated in the UB reactor was 70 L/min. The gas and liquid was mixed in the cross direction at the top of head and dropped into a water tank. The size of water tank was 20 L. A rotameter and pressure gauge were equipped to control the gas and liquid flow and pressure in the reactor.

2.3. EB/UB hybrid system

In this study, toluene was selected as representative VOC to assess decomposition characteristics of VOCs by EB irradiation because it is an artificial VOC emitted most abundantly from industrial facilities (Son