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Oxidation of gaseous styrene by electron beam irradiation

Youn-Suk Son^a, Yeong-Sik Son^a, Jun-Hyung Park^b, Pillheon Kim^c, Jo-Chun Kim^{a,c,*}^a Department of Advanced Technology Fusion, Konkuk University, 1 Hwayang-Dong, Gwangjin-Gu, Seoul 143-701, Republic of Korea^b Department of Eco friendly Research, Korea Dyeing Technology Center, 404-7 Pyongri-6dong, Seo-gu, Daegu 703-834, Republic of Korea^c Department of Environmental Engineering, Konkuk University, 1 Hwayang-Dong, Gwangjin-Gu, Seoul 143-701, Republic of Korea

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ABSTRACT

In this research, the absorbed dose, background gases, relative humidity, and initial concentrations were selected as control factors to discover decomposition characteristics of styrene using electron beam irradiation. It was confirmed that a considerable amount of styrene was removed by primary electrons as well as radicals and ions, which were produced after the electron beam irradiation. Moreover, the removal efficiencies of styrene were observed to rise when the initial concentration was lower, moisture content was higher, and the absorbed dose increased. For instance, 50 ppmv styrene showed extremely high removal efficiency (over 98%) at a condition of 2.5 kGy. A small amount of styrene oxide, benzaldehyde, aerosol, CO, and CO₂ were produced as by-products after EB irradiation.

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1. Introduction

Recently, problems of malodor have emerged as a social issue. It is known that even traditional control techniques with high removal efficiencies have low decomposition efficiencies for low ambient concentrations of malodor in terms of economic points. Therefore, it is difficult to regulate and manage foul smell, compared to other air pollutants (Willing et al., 2004).

Moreover, some VOCs are foul odor-causing substances as well as carcinogens, and mutagens which are harmful to the human body and natural environment. They can also cause photochemical smog, global warming, and ozone destruction, and therefore countries around the world are making greater endeavors to reduce these VOCs (Licki et al., 2003; Vineet and Verma, 2002).

Of these VOCs, Styrene (C₆H₅CH=CH₂) has been classified as a hazardous air pollutant in the 1990 Clean Air Act. Styrene is emitted into the atmosphere from polymer and rubber production processes as well as from solvents and adhesives (Ostapczuk et al., 1999; Tuazon et al., 1993).

In general, removal techniques for odorous compounds include physical, chemical, and biological treatments (Burgess et al., 2001). However, in the case of a high flow rate and a low concentration of styrene, its decomposition by conventional treatment methods (e.g., catalytic burning) is not very effective (Mesyats et al., 2001). Therefore, recent plasma, photocatalyst,

and electron beam (EB) methods have been applied to these odorous compounds (Anderson et al., 1999; Jintawat et al., 2005; Kim, 2002; Kim et al., 2005; Son et al., 2010a, 2010b, 2010c; Ostapczuk et al., 1999).

EB technique, one of the novel methods to remove odorous compounds, can be operated at room temperature, and therefore its energy consumption is extremely low and it can be economically applied to facilities producing a diverse and greater amount of VOCs (Jeon et al., 2008; Kim, 2002; Kim et al., 2010; Son et al., 2010a). This technology is also considered as economical for industries that discharge sundry VOCs and odorous compounds with low concentrations since chemical reaction is exceptionally fast (Kim et al., 2005).

This research aims at identifying the styrene removal efficiency and final by-products depending on the process parameters including absorbed dose, background gases, relative humidity, and initial concentrations as well as the types of reactors.

2. Experiment

2.1. Batch and flow systems

In this research, a 1 MeV EB accelerator (maximum power 40 kW, ELV-4 type, Korea Dyeing Technology Center, Korea) was used. In order to study styrene decomposition characteristics using EB, reactors with batch and continuous flow systems were used.

A cellulose triacetate (CTA) film dosimeter (FTR=125, Fuji, Japan) was used to measure the absorbed dose. A UV/VIS

* Corresponding author at: Department of Advanced Technology Fusion, Konkuk University, 1 Hwayang-Dong, Gwangjin-Gu, Seoul 143-701, Republic of Korea. Tel.: +82 2 450 4009; fax: +82 2 450 2994.

E-mail address: jckim@konkuk.ac.kr (J.-C. Kim).

Spectrophotometer (UVIKONxs, SECOMAM, France) was used to measure the CTA film absorbance at 280 nm wavelength. Based on the results of these measurements, ranges of the absorbed dose were adjusted between 0 and 10 kGy to conduct styrene (50–200 ppmv) decomposition research by EB.

The reactor used in the batch system was a 1 L Tedlar bag (SKC, USA), which is relatively stable to an EB irradiation. This reactor was passed through an irradiation window of the EB accelerator at a velocity of 10 m/min via a conveyor system.

In a continuous flow system, a round-shaped reactor made of stainless steel material of 70 mm diameter, 55 mm height, and 0.212 L volume was used. A thin film (Titanium foil, 50 μm) was installed on the upper part of the reactor, so the EB, which was irradiated from the accelerator, could penetrate inside the reactor.

A compressor (NCP01, AM TECH, Korea) of a 250 L/min volume was used to supply air flow in the continuous flow control system, and a scrubber of silica gel, purafil, and activated charcoal was used to remove moisture and air pollutants (VOCs, SO_x, NO_x, etc.) so that zero air might flow inside the system. During this period, the retention time inside the reactor was 0.85 s.

A VOC generator comprised of a diffusion chamber (diffusion vial), a band heater, and a temperature control apparatus was used in order to produce a consistent level of concentration of styrene. An MFC (mass flow controller, GMATE, Germany) was used to control the concentrations of styrene depending on the flow of dilution air, and a mixing chamber was used so that the styrene with high concentrations would be sufficiently diluted with background gases. A total hydrocarbon measuring device (Total Hydrocarbon, THC; MicroFID, Photovac, Inc., USA) was used to check the instantaneous concentration of styrene, which flowed into the reactor prior to EB irradiation (Fig. 1).

2.2. Sampling and analysis

GC/FID (HP 5890 series II GAS Chromatograph, Hewlett Packard, USA) was used in order to conduct a quantitative analysis before and after EB irradiation, and HP-1 (25 m \times 0.32 mm \times 0.52 μm) was used as a column. GC/MS (Agilent 5975, USA), which was equipped with a thermal desorber (Aerotrapp 6000, Tekmar, USA), was used to make a qualitative analysis of by-products after the EB irradiation.

A NDIR CO/CO₂ measuring device (Gas Data PAQ, UK) was used to measure carbon monoxide and carbon dioxide concentrations. Moreover, an ozone analyzer (Model 49C O₃ Analyzer, Thermo Electron Corporation, USA) was used to measure ozone

concentrations, while a 0.45 μm \times 47 mm membrane filter (Membrane filters cellulose nitrate, MFS[®], USA) was used to measure the aerosol produced after irradiating EB. In this process, the aerosol sampling was conducted at a flow rate of 1 L/min for 10 min.

3. Results and discussion

3.1. Batch system

3.1.1. Background gases

In this research, air, oxygen, and nitrogen of high-purities (99.999%) were selected as background gases in order to understand the removal characteristics of the styrene EB depending on background gases. Moreover, the EB was irradiated on the 200 ppmv styrene while selecting helium (99.999%) as a background gas in order to confirm the decomposition characteristics of the EB while only excluding a radical reaction. 2.5–10 kGy of the EB was irradiated based on absorbed dose, and Fig. 2 shows the removal efficiencies of styrene depending on the background gases.

The residual ratios of styrene (C/C_0 ratio; C: the styrene concentration after EB irradiation, C_0 : the initial concentration of styrene) concentrations on 4 background gases commonly declined when the absorbed dose increased.

The decomposition efficiency of styrene, when helium was a background gas, was lower (45%, 5 kGy) compared to the efficiencies when other background gases were selected, and such results were confirmed to be similar to the removal characteristics of toluene, which belongs to the category of aromatic VOC malodorous substances as well as butane, which belongs to aliphatic VOC (Kim, 2002; Son et al., 2010a). It can be assumed that only primary electrons, which were directly produced by the EB accelerator, influence styrene's decomposition when relatively stable helium is the background gas.

On the other hand, other background gases (air, oxygen, and nitrogen), excluding helium, showed removal efficiencies of 76–86% at 5 kGy. Removal efficiencies when air and oxygen were background gases were 91% and 89% at 10 kGy, respectively, which are higher than those under nitrogen atmosphere. It is assumed that removal efficiencies, when background gases were air and oxygen, were higher than those under helium background since the primary electrons produced by EB as well as major air constituents of air such as oxygen and nitrogen react with electrons, which were produced when EB was irradiated, and

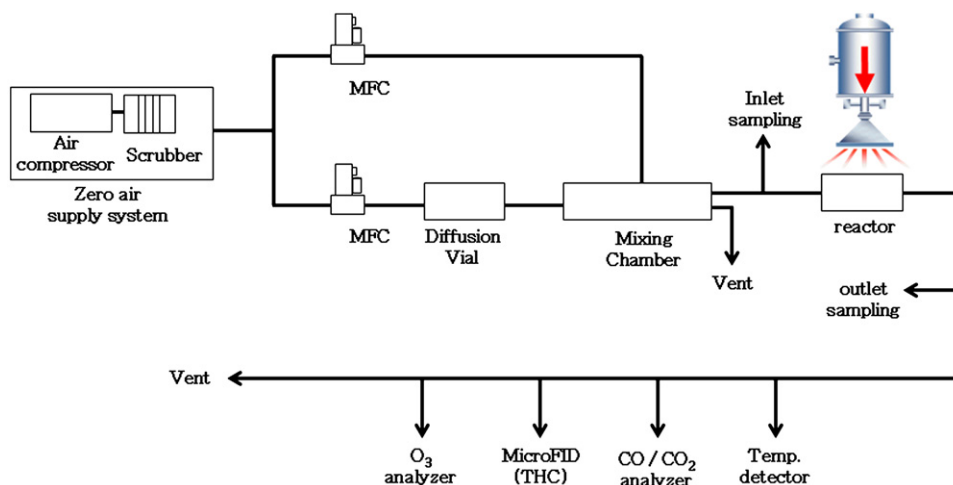


Fig. 1. Schematic diagram of continuous flow system.

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