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Combined radiolytic and catalytic oxidizing method to remove toluene in gas phase

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ABSTRACT

We enhanced the decomposition and the removal of toluene in gas phase using a combined method that consists of electron beam (EB) irradiation and catalytic oxidation. The catalytic oxidation system comprises oxidizing particles of 0.1 wt% Pt or 0.1 wt% Pd deposited to supporting materials, which were also used as adsorbents. These adsorbents—supporting materials are Cordierite, Y-zeolite, and γ-alumina. We demonstrated that 100% removal of toluene can be achieved using approximately 9 kGy of dose level in the presence of Pt or Pd deposited to 12 wt%. The G (-toluene) values were determined to be 0.498 and 0.829 μmol/J in the absence and presence of these catalysts with adsorbents, respectively. We have also demonstrated that the presence of the oxidation-catalyst prevents the toluene from undergoing radiolytic polymerization.

1. Introduction

Volatile organic compounds (VOCs) are emitted from ink coating, petrochemical and combustion processes into the lower atmosphere, play a pivotal role as ozone precursors, and are carcinogenic and mutagenic (Kim, 2002).

At present, conventional technologies such as adsorption, filtration, and combustion have some drawbacks like low efficiency, high maintenance cost; therefore, more cost effective technologies for VOCs control are needed in the world (Wu et al., 1997; Mätzing et al., 1994; Likci et al., 2003).

Over the last two decades, electron beam (EB) irradiation has been applied to remediate VOCs (Hirota et al., 1995, 2002; Kim, 2002). Most notably, Chmielewski's group in Poland has remarkably demonstrated the effectiveness of the use of electron beam in the remediation of VOC in flue gas (Ostapczuk et al., 2008, 1999; Chmielewski et al., 2007, 2004, 2003, 2002; Sun et al., 2007a, b, 2003; Sun and Chmielewski, 2004). Electron beam method can be applied to high flow rate and low concentration of emission facilities at ambient air temperature and requires relatively low dose. However, one of disadvantages associated with this technique is the formation of by-products (Kim et al., 2005; Jeon et al., 2008; Sun et al., 2009). Jeon et al. (2008), Kim et al. (2005), and Jun et al. (2004) have demonstrated that a combined method of electron beam and oxidation-catalyst with adsorbents can enhance the efficiency of the VOC remediation.

In this work, toluene was chosen as a typical compound in the VOC from coating process. Three catalyst support materials (Cordierite, Y-zeolite, and γ-alumina soaked with oxidizing catalysts platinum (0.1 wt% Pt) or palladium (0.1 wt% Pd)) are employed to compare combined effects. In addition, a dose range of 1.5–9 kGy is applied to the combined system in order to observe the removal efficiencies of toluene.

2. Experiment

2.1. Catalyst and support materials

For catalytic oxidation, we used Pt and Pd (0.1 wt%) because of their oxidation efficiencies of VOC at relatively low temperatures such as 170–250 °C (Grbic et al., 2004; Wu et al., 2000; Garetto et al., 2000; Burgos et al., 2002; Okumura et al., 2003; Dégé et al., 2000; Tidahy et al., 2007).

Ceramic honeycomb (Ceracomb, South Korea) was used as a catalyst support materials. It is composed of cordierite comprising 200 cells/in^2 with thickness of 0.33 mm and surface area of

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1818 m^2/m^3. The physical and chemical properties of cordierite and its chemical compositions are presented in Table 1.

We also used Y-zeolite (6 wt%) and γ-alumina (6 wt%) which have an excellent adsorption capacity. Adsorption, desorption, and catalytic reactions are carried out in a small pore of zeolite. Especially in case of ultra stable Y-zeolite, its pore inlet consists of 12 oxygen atoms, so relatively higher molecular weight VOC such as benzene, toluene, and their radiolytic products can be easily adsorbed. The physical properties of Y-zeolite are shown in Table 2.

2.2. Lab-scale VOC control system

The electron beam (EB) accelerator used in this study is a 1 MeV ELV-4 type with a maximum power of 40 kW. The average absorbed dose by the flowing sample gas was measured using cellulose triacetate (CTA) film dosimeter (FTR=125, Fuji, Japan). CTA films were located at the top and the bottom of the reactor.

Table 1
Physical properties of cordierite.

Composition (%)		Physical properties	
Al_2O_3	36	Compressive strength (kg/cm^2)	150
SiO_2	50	Volumetric specific gravity (g/cc)	1.7
MgO	14	Thermal expansion coefficient	$1.0 \times 10^{-6}\,°C^{-1}$
TiO_2	Max. 1.0	Specific heat (20–400 °C)	0.26 cal/g °C
Fe_2O_3	Max. 0.5	Heat shock resistant	Max. 400 °C
K_2O	Max. 0.5	Porosity	30%
Na_2O	Max. 0.5	Absorption rate	18%
CaO	Max. 0.5	Max. temperature	1300 °C

Table 2
Physical properties of zeolite (CBV 901).

Items		Value	Unit
XRF	Na_2O	< 0.05	wt%
	SiO_2/Al_2O_3	83	molar ratio
XRD	Unit cell size	24.26	Å
	Crystallinity	110	%
Surface area	Multi point	782	m^2/g
Particle size	D50	5.6	μm
	D90	20.2	μm

The irradiated films were analyzed by UV–visible spectrophotometer (UV-160 A, Shimadzu, Japan) at 280 nm within 2 h.

The lab-scale VOC control system shown in Fig. 1 consists of air flow control, VOC generation, reaction and sampling sections, and the electron beam accelerator.

Air flow was introduced into the VOC control system using a 250 L/min of air compressor (AM TECH, NCP01, Korea), and zero air was then generated through a series of scrubbers, including silica gel, activated carbon, purafil, and charcoal traps as, shown in Fig. 1. All the experiments were carried out using the flow system, and the flow rates were controlled by mass flow controllers (MFC, Bronkhorst, USA; Brooks Instrument, 5850E, USA). In addition, a diffusion technique was adopted to generate constant VOC concentrations. The temperature inside a diffusion vial was controlled by a Band Heater with a temperature controller, and a constant level of 214 ppm (8.76 μmol/L) toluene was introduced into the VOC control system at a flow rate of 15 L/min.

The reactor shape used in this work was cylindrical with the internal size 7 cm in diameter and 5.5 cm in height. The top of the reactor was covered with titanium film (50 μm). Analytical instruments were set up to measure gaseous concentrations before and after EB irradiation.

2.3. Sampling and chemical analysis

Gaseous samples were collected before and after irradiation in order to investigate the removal efficiency of toluene. A Tedlar bag (5 L, SKC, USA) was used for sampling. A lung sampler was devised to take the samples directly into the bag instead of going through the air pumps in order to prevent them from deteriorating (Kim, 2002). A GC/FID (HP5890) was used to make quantitative analyses of the samples. For the qualitative analysis, a GC (HP6890)/MSD (HP5973) was also employed. Duplicate samples were collected and analyzed, the precision of which was found to be 5%. The replicate analysis was carried out and RSD was less than 6.7%.

A CO_2 analyzer (PAQ, Gas Data, UK) was adopted to measure the amount of CO_2. The Aerotrap 6000 (Tekmar) was used to pretreat the samples as they were collected through the adsorbent trap. An ozone analyzer (Model 49C O_3 Analyzer, Thermo Electron Corporation, USA) was also used to measure ozone concentrations formed during the toluene removal process. Membrane filters (Membrane filters cellulose nitrate, MFS®, USA; 0.45 μm, 47 mm)

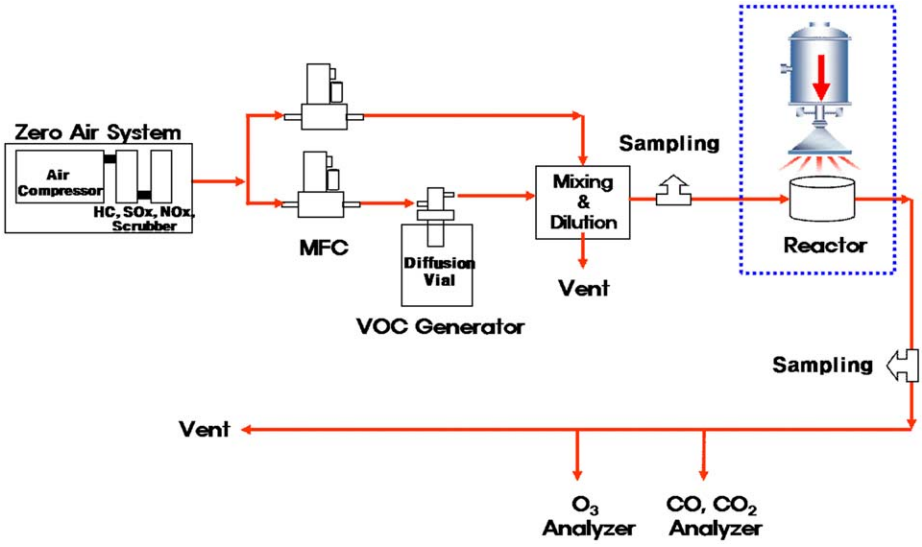

Fig. 1. Schematic diagram of lab-scale VOC remediation system.