Chemical Engineering Journal 274 (2015) 9–16

Contents lists available at ScienceDirect

Chemical Engineering Journal

journal homepage: www.elsevier.com/locate/cej

Decomposition of sulfur compounds by a radiolysis: III. A hybrid system and field application

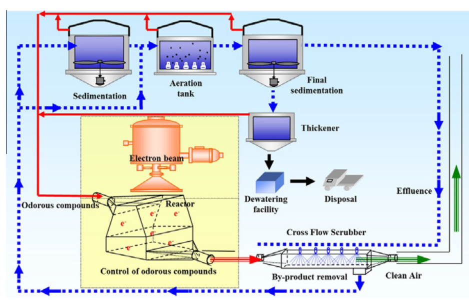
Youn-Suk Son [a,*], In-Ha Jung [a], Sung-Joo Lee [a], Jo-Chun Kim [b]

[a] Research Division for Industry & Environment, Korea Atomic Energy Research Institute, Jeongeup-si, Jeollabuk-do 580-185, Republic of Korea
[b] Department of Environmental Engineering, Konkuk University, Seoul 143-701, Republic of Korea

HIGHLIGHTS

- This hybrid system was developed to control odorous compounds.
- REs by an EB were $H_2S > CH_3SH > (CH_3)_2S > (CH_3)_2S_2$ at 5–10 kGy.
- The CF scrubber was sufficient to remove residual compounds and its by-products.
- REs of all compounds were about 100% at 2.5 kGy in the pilot-scale hybrid system.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 22 January 2015
Received in revised form 23 March 2015
Accepted 25 March 2015
Available online 1 April 2015

Keywords:
Electron beam
Cross flow scrubber
Sulfur compound
Hydrogen sulfide
Methyl mercaptan
Hybrid system

ABSTRACT

In this study, a hybrid system combining two different techniques (electron beam and cross flow scrubber) has been developed and applied to control odorous sulfur compounds (hydrogen sulfide, methyl mercaptan, dimethyl sulfide and dimethyl disulfide) in a wastewater treatment facility. Removal efficiencies of sulfur compounds using an electron beam were high in the order of hydrogen sulfide > methyl mercaptan > dimethyl sulfide > dimethyl disulfide when the range of absorbed dose was 5–10 kGy. We also found that the liquid/gas ratio at 4.7 L/m^3 was sufficient to remove residual odorous sulfur compounds and its by-products. Based on these results, a pilot study was carried out. As a result, the removal efficiencies of all target compounds were approximately 100% at 2.5 kGy in the pilot-scale hybrid system. It was also found that the levels of NO_2 and CO formed by electron beam irradiation were lowered down to the ambient level by the hybrid system.

1. Introduction

There have been increased concerns regarding odorous sulfur compounds such as hydrogen sulfide (H_2S), methyl mercaptan (CH_3SH), dimethyl sulfide ($(CH_3)_2S$) and dimethyl disulfide ($(CH_3)_2S_2$) due to industrialization and urbanization [1–3]. These compounds are emitted from numerous industrial facilities (metal smelting and leather tanning) and environmental pollution control facilities (landfill and wastewater treatment) [4–6]. Most of these compounds have a very low threshold value (e. g. CH_3SH: 0.07 ppb), high toxicity, and potential corrosive effect. Thus, these compounds induce adverse effects on human health and materials [7,8]. Additionally, it has been reported that a part of these compounds, such as $(CH_3)_2S$, affect climate change [2,3].

In order to treat odorous sulfur compounds, diverse techniques have been studied during the last few decades, including absorption, adsorption, incineration and biofiltration [8,9]. However, these methods have certain disadvantages such as high operating and maintenance costs [10,11]. To overcome these problems, advanced oxidation technologies such as plasma, photocatalyst, and electron beam (EB) have been studied [12–14]. Among these methods, EB is well known as an economical and energy efficient technology because it is operated at ambient air temperature and

* Corresponding author. Tel.: +82 63 570 3345; fax: +82 63 570 3347.
E-mail address: sonys@kaeri.re.kr (Y.-S. Son).

http://dx.doi.org/10.1016/j.cej.2015.03.122
1385-8947/

a short reaction time [15–19]. Accordingly, EB has been studied to removal various odor compounds (ammonia, acetaldehyde, styrene and trimethylamine) [14,20–22]. Furthermore, many studies regarding the removal of odorous sulfur compounds using EB have been carried out [23,24]. Notwithstanding the development of EB technology, various limitations such as a by-product (aerosol, ozone, and etc.) and low removal efficiency with some compounds have been reported. Thus, a few hybrid systems such as an EB/catalyst were introduced to resolve these problems [19,25–28]. However, the waste gases emitted from diverse environmental treatment processes, e.g., a wastewater treatment facility (WTF) include various levels of humidity that can induce a reduction of the removal efficiency.

In this study, to overcome these problems, a novel hybrid system was developed to control odorous compounds emitted from a WTF. An EB and cross flow scrubber (CFS) were combined to minimize the formation of by-products as well as a reduction of the removal efficiency by humidity. To this end, we used a small scale reactor to identify the removal characteristics and efficiency on mixed odorous compounds. The reactor was scaled up to the pilot scale to apply a field test.

2. Experimental

2.1. Lab scale study

The continuous flow system was consisted of a zero-air supply system, mixing chamber, EB reactor, standard gas cylinders (H_2S, CH_3SH, 1% $(CH_3)_2S)/N_2$ and 400 ppm $(CH_3)_2S_2/N_2$, Rigas, Korea) and sampling ports. To introduce 15 L/min clean air into a reactor, a zero-air system was used and it was composed of a compressor (NCP01, AMTECH, Korea) with a 250 L/min flow capacity and various scrubbers such as silica gel, purafil, and activated charcoal. The clean air generated from the zero-air system and standard gases were introduced and adjusted to the appropriate concentration by rotameters, mass flow controllers (5850E, Brooks Instrument, USA), and mixing chambers. The initial concentrations of H_2S, CH_3SH, $(CH_3)_2S$ and $(CH_3)_2S_2$ adjusted in this study were approximately 80, 63, 109 and 35 ppm, respectively. At that time, relative humidity of the inflow gas was about 4%.

A lab scale EB reactor was used to figure out the removal efficiency and characteristics of mixed odorous compounds. To do this, a round-shaped reactor made of stainless steel (70 mm (D) × 55 mm (H) × 0.212 L (V)) was used, and the residence time was 0.85 s. To penetrate the EB, a thin titanium foil (50 μm) was installed on the upper part of the reactor. A thermocouple was used to measure the temperature inside the reactor. The concentrations of sulfur compounds before and after the EB reactor were measured to assess the removal efficiencies. Detailed information regarding the reactor was previously published [20–22,24].

To treat by-products such as ozone and aerosol formed during the EB irradiation process, a CFS was developed, as shown in Fig. S1 of Supporting information. The size of the CFS was 640 mm (L) × 85 mm (W) × 100 mm (H), and the residence time was 20 s. To improve the air liquid contact time, the inside of the CFS was divided as a zigzag shape by three barriers and four nozzles (HM-FF, orifice diameter (0.9 mm), droplet diameter (500–550 μm), and spray angle (58°), Hanmi Nozzle, Korea) were installed in parallel with the upper part of the CFS. The liquid/gas ratios (L/G ratios) were adjusted to 4.7–100 L/m^3.

2.2. Pilot scale study

The pilot scale control system for odorous compounds was roughly composed of a flow control system, odor generation system, EB reactor, CFS and advanced ozone treatment reactor. The air sampling was simultaneously carried out to assess the removal efficiencies at four sampling ports (stage 1, 2, 3 and 4), as shown in Fig. 1. The values measured at stage 1, 2, 3 and 4 were the initial concentration emitted from an odor generation reactor, the concentration after EB irradiation, the concentration after CFS, and the concentration after advanced ozone treatment, respectively. Each sampling port was equipped with a manifold to concurrently collect and measure various odorous compounds. To supply an appropriate air flow, two fans equipped with an inverter (N100-1HP, Hyundae, Korea) were used and the flow rate of the flow control system was adjusted to 100 m^3/h. Additionally, a flowmeter (MP200, KIMO, France) was used to check the relationship between the flow rate and fan frequency. The coefficient of determination (r^2) was above 0.999 (Fig. S2 in Supporting information). A self-developed odor generation reactor (V: 600 L), which was filled with sludge generated from a dyeing WTF, was used to make similar smell to WTF. Air that was generated from an aeration tank of the dyeing WTF was introduced to the odor generation reactor. The air was then passed through an EB reactor. Based on the lab scale EB reactor, a pilot scale EB reactor was made to conduct a field test. The size of this reactor was 650 mm (L) × 175 mm (W) × 340 mm (H) and the volume was 37 L. The residence time of the reactor was 1.39 s. In order to minimize the bias of the flow stream, the inlet section was designed in the shape of a quadrangular pyramid. Four thermocouples were installed to monitor the temperature inside the reactor. In addition, the distance between an EB window and reactor was 170 mm.

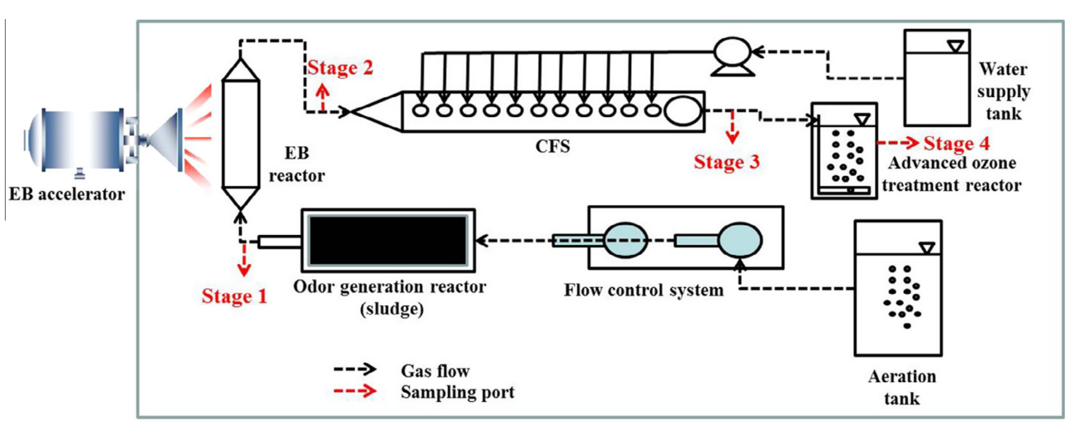


Fig. 1. Schematic diagram of pilot-scale control system for odorous compounds.