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VOC Emissions from a Rendering Plant and Evaluation for Removal of Pentanal by Oxidization Using Hydrogen Peroxide

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ABSTRACT

Rendering plants treat dead livestock and produce grease and bone meal. In a rendering plant, the cooking and drying processes are the main sources of odor emissions. Non-fresh dead livestock reduce the performance of odor control devices, and in Taiwan, the treatment facilities in a rendering plant mostly are operated in a batch feeding, which causes volatile organic compound (VOC) emissions in the exhausted gas, that always caused complaints from the nearby neighborhood. This study used respectively ozone and hydrogen peroxide to evaluate the removal efficiencies of pentanal, hexanal and toluene those are common VOCs in the rendering exhaustion. Experimental results indicated that ozone could not effectively reduce aldehydes and toluene, and the residual ozone remaining in the exhaust gas is a secondary air pollutant and irritate the human respiratory tracts. Oppositely, hydrogen peroxide effectively removed pentanal as a feasible VOC treatment oxidant by adding into a contact reactor. When the pentanal exhaustion concentration from the rendering process was around 36.23 ppm in the flue with the flow rates from 100 to 250 $Nm^3 min^{-1}$, the reaction rate constant of pentanal for the first-order reaction by aqueous hydrogen peroxide of 1,000 mg L^{-1} was obtained as 0.536 1 s^{-1}, and then the pentanal reduced to 0.68 to 2 ppm. Based on the simulation using the Gaussian dispersion model, the concentration ranges of pentanal in the exhausted stream resulted in the pentanal emission rate lower than 0.01 g s^{-1}, which no longer causes surrounding residents' complaints.

Keywords: Rendering, Ozone, Hydrogen peroxide, Pentanal, Volatile organic compounds, Oxidation

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1 INTRODUCTION

Rendering plants are built to treat dead livestock and poultry, and mainly produce grease and bone meal under high temperature ranging from 115 to 145°C for 40–90 min (Bhatti *et al.*, 2014; Meeker, 2021). Volatile organic compounds (VOCs) contained in the waste gases emitted from the rendering plants are very much complex, including organic acids, alcohols, aldehydes, amines, aliphatic hydrocarbons and aromatic hydrocarbons (Shareefdeen *et al.*, 2005, Tymczyna *et al.*, 2013; Bhatti *et al.*, 2014; NRA, 2016; Guerra *et al.*, 2017). At present, the rendering plants in Taiwan commonly use the batch feeding system to process the animal carcasses, resulting in unstable concentrations of VOCs in the exhaust gas are unstable, and the freshness of carcasses will also affect the VOC treatment efficiencies of the air pollution control devices (APCDs). Most of the rendering plants in Taiwan are located in agricultural counties where the animal husbandry is prosperous. Due to the distribution of dense population countryside, the stench generated by VOCs could easily affect the ambient air quality of neighboring communities, which often cause the neighboring residents to report to the local environmental agencies.

According to previous studies investigating on the composition of VOCs exhausted from the rendering plants, aldehydes were the most detected VOCs (Defoer *et al.*, 2002). The aldehydes emitted from the rendering processes are mainly produced during the processes of cooking and drying corpses (Defoer *et al.*, 2002; Lin, 2022). Notably, the odor thresholds of pentanal and hexanal are as low as 0.41 and 0.28 ppb, respectively. The odor of pentanal is characterized by a strongly pungent and ethereal smell, which is particularly unbearable (Nagata, 2003; Kultan *et al.*, 2022).

Rendering plants usually adopted bio-filters (Shareefdeen *et al.*, 2003, 2005; Tymczyna *et al.*, 2013) and wet scrubbers (Kastner and Das, 2002) for the abatement of VOCs. However, according to the earlier experiments in the field, bio-filters and wet scrubbers are suitable to treat the large flow rates of exhausted gas from rendering plants. For example, Shareefdeen *et al.* (2003, 2005) reported that the rendering plants in Ontario, Canada, designed a commercial-scaled bio-filter, which treated an exhausted air stream with an air flowrate of 250,000 ft^3 min^{-1} (147,000 m^3 h^{-1}). Kastner and Das (2002) also studied wet scrubbers with an air flowrate of 33,994 m^3 h^{-1}, which was operated by using chlorine dioxide as an oxidant to deodorize for a rendering plant in Georgia, United States. However, the overall abatement performance of aldehydes by the wet scrubber was unstable, widely ranging from 20% to 80%.

Pingtung County is an agricultural county in southern Taiwan, where a large number of poultry and livestock are raised. In the past decades, incineration and burial are two major methods commonly applied for disposing dead animal carcasses in Taiwan. However, the residual ashes still need to be treated separately after incineration. Burial sites are quite difficult to locate due to dense population in Taiwan. Moreover, the leachate and odors might also pollute the environment, causing serious secondary pollutant problems. Recently, Taiwan Environment Protection Administration (Taiwan EPA) promote the establishment of rendering plant to produce grease and bone meal which can be used for direct animal feeding and producing raw materials of food products in order to achieve the sustainable agriculture (NRA, 2016; Lin, 2022). In this study, the identification of odor characteristics and the feasibility for deodorization in a rendering plant with the exhaust gas of 6,000–15,000 m^3 h^{-1} was investigated. The target rendering plant uses a batch system to feed dead animals corpses to the rendering cooker. Thus, the composition and concentrations of VOCs in the exhaust gas are mostly unstable. Besides, the operating time of the target plant ranges from 12 h to 18 h per day, thus it is not suitable for using biological deodorization unit which requires a stable organic loading. Based on the stable treatment efficiencies of aldehydes by using chemical oxidation has been proved to be feasible in previous work (Kastner and Das, 2002), two low-cost oxidants, including ozone and hydrogen peroxide, were tested for abating the pentanal which is an irritating odorous VOC.

2 METHODS

2.1 Characteristics of Flue Gas from the Target Plant

In this study, the target rendering plant is located at Pingtung County in southern Taiwan. Its treatment capacity of animal corpses by a batch input was 18,000 ton y^{-1} in 2022. Due to its unsteady input of raw materials and varying dead animal corpses (different time of death and preservation conditions), the flow rates of the flue gas ranged from 100 to 250 Nm^3 min^{-1} at the site. The operation period starting from the noon to 6:00 am in the next morning for a batch and within this period both processes of cooking and drying corpses took nearly 11 h. The operating sequence of rendering process for the target plant is illustrated in Fig. 1. Additionally, the main odor emission sources and their photos at the site are also depicted in Fig. 1. As the earlier information, the cooker is the most odorous unit among the rendering processes in a typical rendering facility.

2.2 Equipment

The set-up of VOC contact reaction system is illustrated in Fig. 2. VOCs of various concentrations were prepared using a 50-L Tedlar sampling bag, or collected the exhaust gas of the rendering plant. The VOCs in the sampling bag was extracted by an air pump to be mixed with ozone and then injected into the oxidization reactor; or directly added aqueous hydrogen peroxide into the reactor. The oxidization reactor (No. 6 in Fig. 2) was adopted using the glass tube with a length of 45 cm and a diameter of 4 cm, which was packed with glass breeds (Ø = 1.5 cm) inside the reactor. An air pump (No. 1 in Fig. 2) was adjusted with its exhaust air for the air flowrates ranging

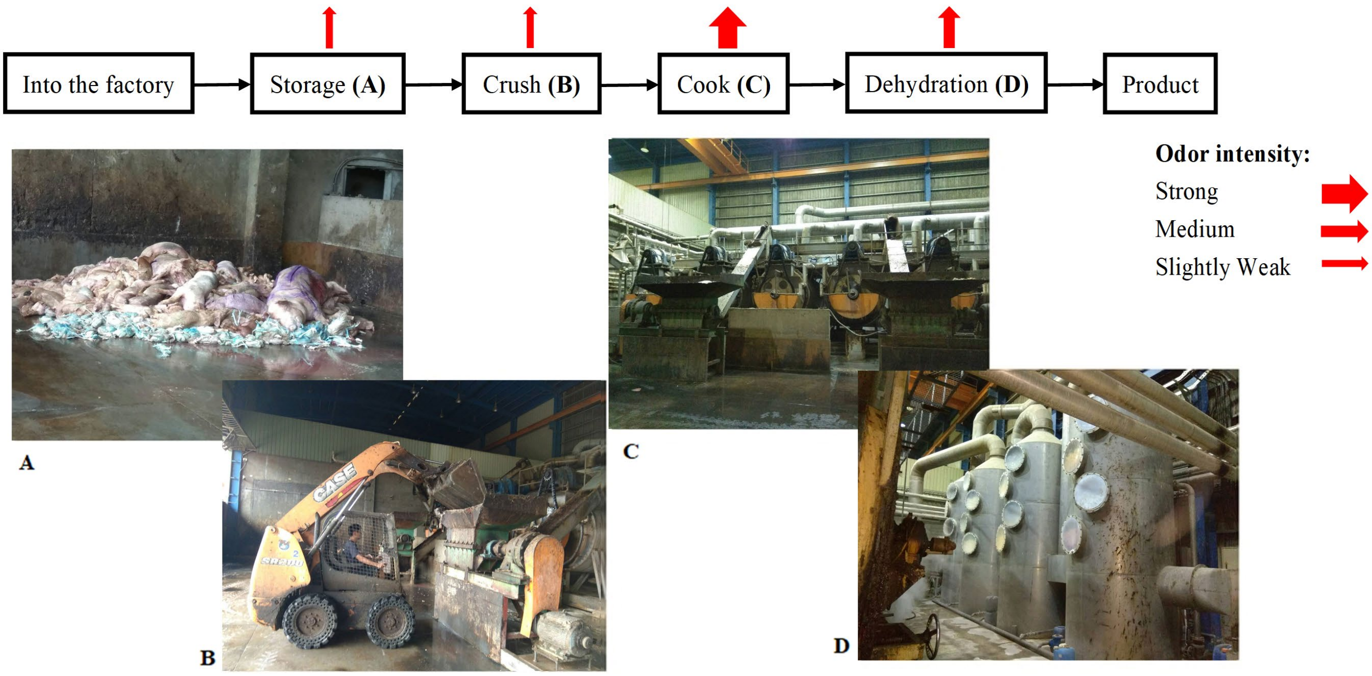

Fig. 1. Flow chart and photographs of the rendering process.

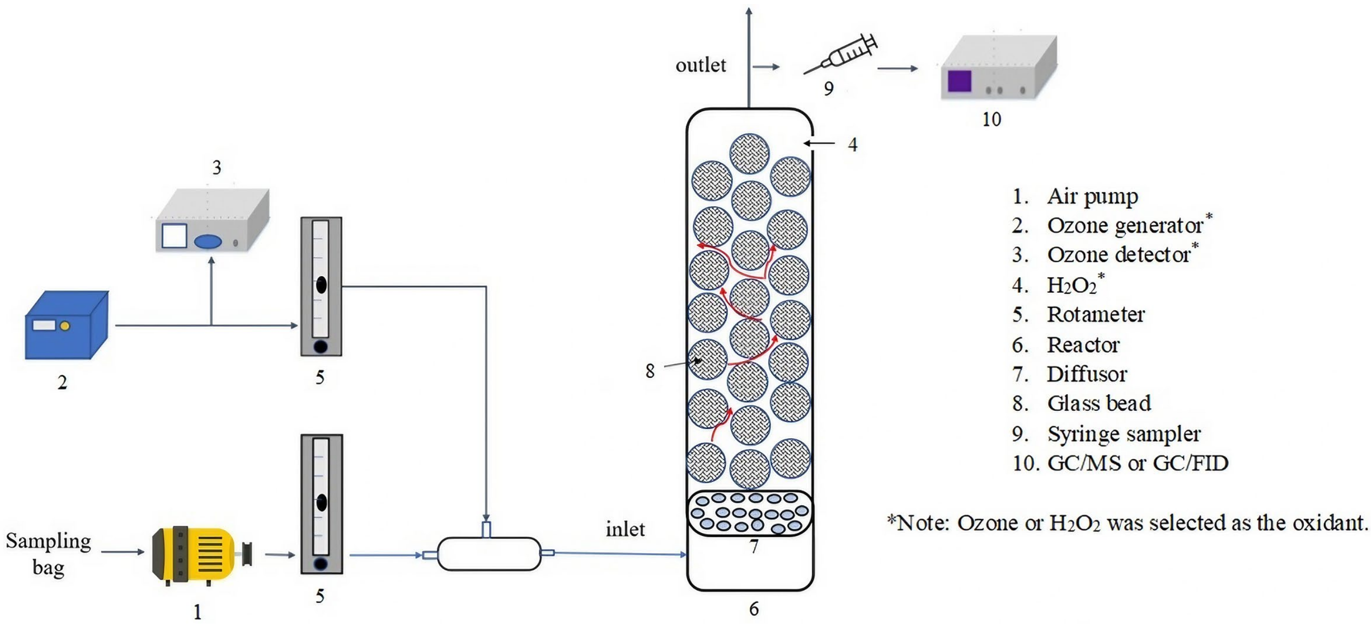

Fig. 2. Schematic of the laboratory-scale oxidation reactor.

from 100 to 400 mL min^{-1} through the reactor. The retention time of gas in the oxidization reactor excluding the volume of packed breeds were 60–240 s.

An ozone generator (KI-10A, No. 2 in Fig. 2) is manufactured by Three Oxygen Enterprise Co., Ltd., Taiwan. An air pump (Buck LP-5, A.P. Buck, Inc., USA) was used for collecting air samples and extracting gas in the field. Tedlar bags (CEL Scientific Corporation, USA) of 1 L and 50 L were made of polyvinylfluoride (PVF). Two or three Rotameters (Models RMA-12-SSV and RMA-151-SSV, Dwyer, USA) illustrated in Fig. 2 were used for regulating the flow rates of gas streams.

2.3 Chemical Analysis and Instruments

A gas chromatograph (GC) (7890A, Agilent, USA) with a mass spectrum (MS) (5975C, Agilent, USA) was used to qualify and quantify VOCs emitted from the rendering facilities. The capillary column is DB-624 with a length of 60 m, an inner diameter of 0.32 mm, and a coating thickness 1.8 µm. Gas sample were carried by Helium (He) as the carrier gas. The analysis of VOCs was operated with the inlet gas temperature was of 180°C, the inlet pressure was of 4.96 psi, and the split ratio was of 50:1. The initial temperature in the oven of GC was 50°C for 1 min. The temperature of the oven was risen up at a heating rate of 35°C min^{-1} till 140°C for 6 min. An ozone detector (UVOZ-3000, ZHISO, China) was applied to detect ozone concentration in the ppm level. A photo-ionization gas detector (PID) (Tiger, Ion Science, United Kingdom) was applied to detect VOCs in the ppm level. Standard gases including toluene, pentanal and hexanal used in this study were all of analytical grades (Merck, Germany). The analysis and calibration of VOCs followed the procedures of the standard method developed by Taiwan EPA (2021).

3 RESULTS AND DISCUSSION

3.1 Characteristics of VOCs in Gas Streams Emitted from Rendering Plants

Four gas samples containing VOCs were respectively collected from the exhaust gas streams emitted from the cooking process of the target rendering plant in various operation days. The analytical results, which revealed the detected organic constituents, are summarized in Table 1. Typically, aldehydes, acetone and toluene are the most common VOCs, those were examined by Bhatti *et al.* (2014), and the VOCs were mainly emitted from the end process after the cooking process in a rendering plant. However, alcohols, especially ethanol, which was identified in the target plant for all gas samples, was not examined by earlier researchers (Anet *et al.*, 2013; Bhatti *et al.*, 2014; Sazakli and Leotsinidis, 2021). Notably, butanol was only detected during the sterilization process, but not during the cooking process Bhatti *et al.* (2014).

Table 1. Emitted organic compounds of four samplings in the target plant and comparisons to the results studied by Bhatti *et al.* (2014).

Compounds	To be detected or not for the samples taken at the site[a]				Concentrations analyzed by Bhatti *et al.* (2014)[b]
	#1	#2	#3	#4	
Aldehydes					
Propanal				✓	
Butanal				✓	
2-Methyl propanal		✓			270 mg m^{-3}
2-Methyl butanal		✓			210 mg m^{-3}
3-Methyl butanal	✓	✓		✓	61 mg m^{-3}
Pentanal	✓	✓	✓	✓	40 mg m^{-3}
Hexanal	✓	✓	✓	✓	142 mg m^{-3}
Alcohols					
Ethanol	✓	✓	✓	✓	
Propanol	✓	✓			
Butanol					34 mg m^{-3}
Pentanol		✓			
Ketones					
Acetone		✓	✓		116 mg m^{-3}
Aliphatic HCs					
Octane	✓				20 mg m^{-3}
Aromatic HCs					
Toluene	✓	✓	✓	✓	400 mg m^{-3}
Nitrogenous compounds					
Propionamide	✓			✓	
Dimethylamine	✓				

[a] Symbol ✓ indicates the compound was detected in the air samples, which were exhausted from the cooker of the target plant.
[b] The total concentrations emitted from the cooker, sterilizer and condenser in a rendering plant in Ichtegem, Belgium (Bhatti *et al.*, 2014).

Pentanal, hexanal and toluene were the mostly frequently detected VOC species in the VOC gas samples which have been also identified by Bhatti *et al.* (2014). Hence, pentanal, hexanal and toluene were the major VOC species simulated as the mixture fed as the influent VOCs for the abatement of odors. Ethanol was not selected as the target compound in this study owing to its higher odor threshold value of 10 ppm (Nagata, 2003).

3.2 Degradation of VOCs in the Gas Streams by Ozone and Hydrogen Peroxide

VOC samples containing toluene, pentanal, and hexanal with the concentrations of 46.9, 9.77, and 15.6 ppm, respectively, were collected and stored in the Tedler sampling bags. The VOC gases were fed into the oxidization reactor in a flow rate of 400 mL min^{-1}, and meanwhile ozone of concentration of 250–300 ppm was also injected into the reactor in a flow rate of 20 mL min^{-1} (i.e., the retention time of 68 s). After the oxidation of ozone in the reactor, the concentrations of toluene, pentanal, and hexanal were decreased to 39.6, 8.19, and 12.31 ppm, respectively. As illustrated in Fig. 3, the removal efficiencies of toluene, pentanal, and hexanal were 3.4, 4.5 and 4.68%, respectively. Fig. 3 depicts that the removal efficiencies of VOCs gradually increased with the increase of ozone concentrations fed into the reactor. When the ozone concentration was 1,700 ppm and the ozone retention time was 125 s, the removal efficiencies of pentanal and hexanal were 64% and 68%, respectively. When the ozone retention time was increased to 200 s, the removal efficiencies of pentanal and hexanal increased to 96 and 89%, respectively. However, the removal efficiencies of toluene were always lower than 40%. According to the results of the experimental tests, in addition to the poor efficiencies of ozone in removing toluene, if the efficiencies of ozone for abating aldehydes exceed 90%, the concentration of ozone injected into the oxidation reactor needs to be as high as 1,700 ppm. That dosage of ozone lacks economic feasibility and the residual ozone is much irritating stink, as well as causing secondary pollution problems.

The mixture gas of toluene, pentanal, and hexanal in the Tedler sampling bag was further tested for VOCs removal with hydrogen peroxide as an alternative oxidant. The initial concentration of total volatile organic compound (TVOC) in the mixture gas of toluene, pentanal, and hexanal in the Tedler sampling bag was 307 ppm, which was fed into the oxidation reactor in the air flowrate of 200 mL min^{-1}, and 30% hydrogen peroxide was also injected through the top inlet port of the reactor using a batch feeding mode as illustrated in Fig. 2. It was found that the TVOC concentration with the consumption time of hydrogen peroxide gradually rose to 151 ppm, and the removal efficiencies of TVOC decreased gradually to 51% (Fig. 4). Notably, when the dosage time was 30 min by adding 30% hydrogen peroxide, the removal efficiencies of TVOC still remained approximately 70%, i.e., the effluent TVOC concentration of about 95 ppm. Therefore, the batch addition of hydrogen peroxide revealed a dominant performance on the abatement of the mixture of toluene, pentanal, and hexanal. Pentanal is the compound with a lower odor threshold of three target VOCs, and it is often complained by residents around the rendering plant as its strongly irritating smell. Therefore, more hydrogen peroxide addition tests were conducted using pentanal

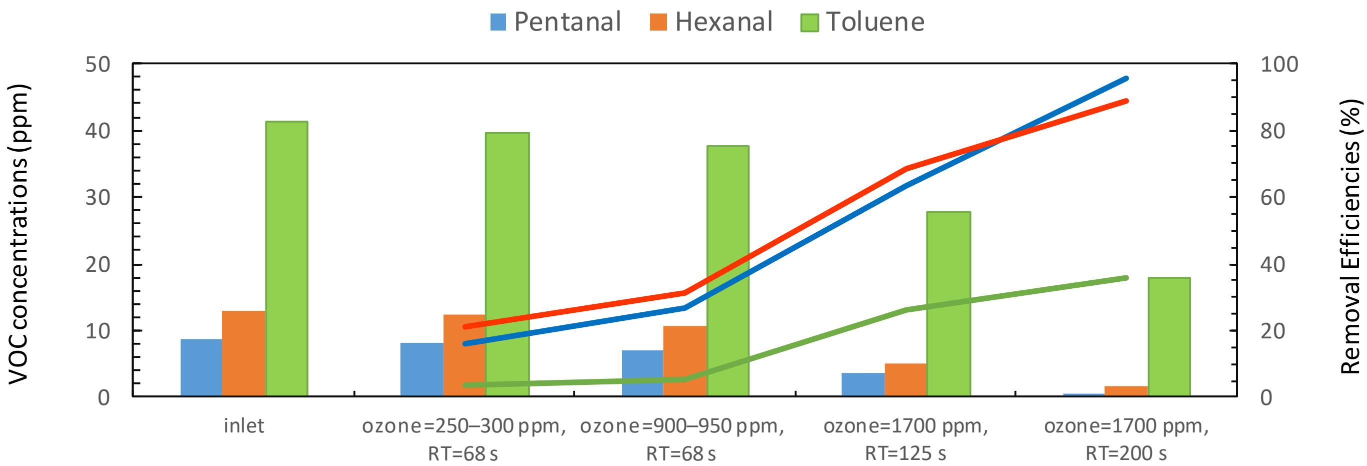

Fig. 3. VOC concentrations and removal efficiencies at different ozone reaction conditions.

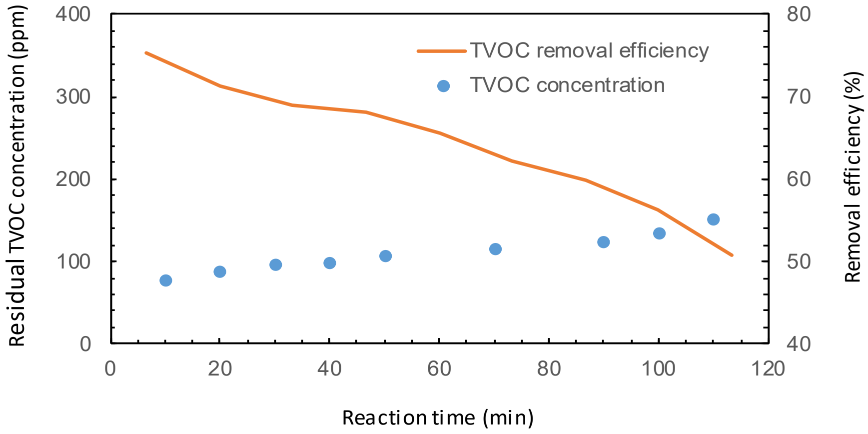

Fig. 4. Variation of removal efficiencies of the mixture of pentanal, hexanal and toluene (an initial TVOC concentration 307 ppm @ 200 mL min^{-1}), oxidized by 30% hydrogen peroxide solution.

as a single component to explore the detailed reaction mechanisms for the chemical oxidation of pentanal by hydrogen peroxide.

3.3 Removal of Pentanal Using Hydrogen Peroxide as an Oxidant in the Reactor

Different volumes (100, 80, 60, 40, and 20 mL) of 1,000 mg L^{-1} hydrogen peroxide were added into the oxidation reactor in a batch feeding. The concentration of pentanal in the Tedlar sampling bag was 36.23 ppm which was continuously added into the reactor in a flow rate of 500 mL min^{-1}. As illustrated in Fig. 5, the smaller the volume of hydrogen peroxide added, the faster the residual concentration of pentanal left in the exhaust gases at the outlet of the reactor. Based on the removal efficiencies of pentanal which are calculated as illustrated in Fig. 6, adding at least 80 mL of hydrogen peroxide keeps the removal efficiencies of pentanal higher than 80% within 10 min contact after the oxidation of pentanal by hydrogen peroxide.

3.4 Feasibility Assessment of Pentanal Removal for the Real Plant

Assuming that the pentanal-hydrogen peroxide redox follows the first-order reaction (Benefield *et al.*, 1982), the reaction equation can be expressed by Eq. (1)

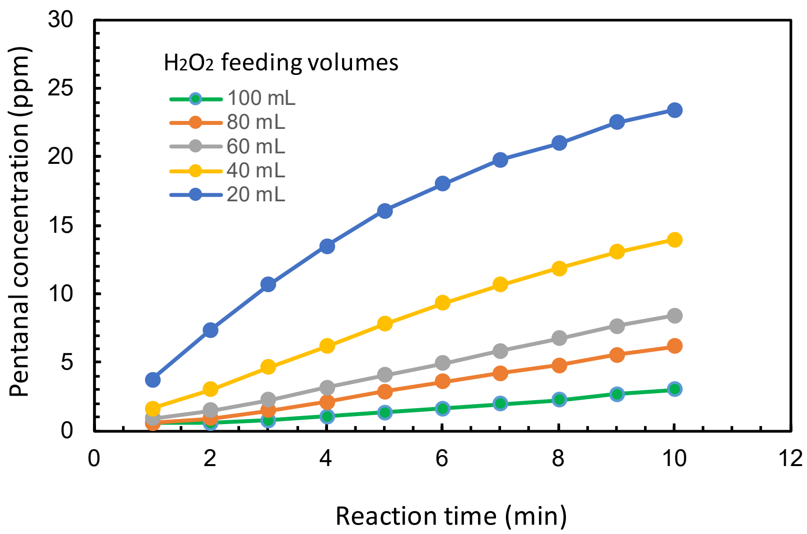

Fig. 5. Variation of residual pentanal (the initial concentration of 36.23 ppm) with reaction time for adding various volumes of 1,000 mg L^{-1} hydrogen peroxide.

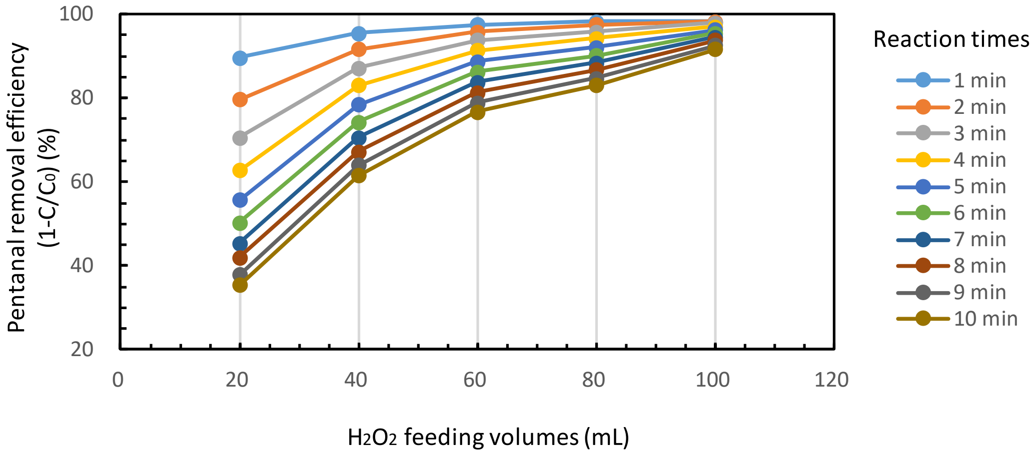

Fig. 6. Pentanal removal efficiencies varying with reaction time for adding various volumes of 1,000 mg L^{-1} hydrogen peroxide.

$$C = C_0 e^{-kt} \quad (1)$$

where C is the final concentration (ppm) of pentanal during the reaction; C_0 is the initial concentration (ppm) of pentanal before the reaction, (i.e., 36.23 ppm); k is the reaction rate constant of pentanal-hydrogen peroxide reaction rate (1 s^{-1}); and t is the contact time for the reaction of pentanal and hydrogen peroxide.

Taking the logarithm of Eq. (1), Eq. (2) can be obtained as follows,

$$\ln C = \ln C_0 - kt \quad (2)$$

The experimental data of 1–10 min contact oxidation reaction are plotted with the values of ($\ln C$) and (t) in the Eq. (2). The linear regression coefficients (R^2) of the reaction formula for t = 3–10 min exceeded 0.988, except for t = 1 min, R^2 was less than 0.90, and t = 2 min, R^2 was less than 0.98. It confirmed that the oxidation of pentanal by hydrogen peroxide follows the first-order reaction at the high contact reaction time, ranging from 3 to10 min.

The reaction formula at t = 8 min is as follows

$$\ln C = 3.816 - 0.536t \quad (3)$$

The intercept of Eq. (3) is very near to the logarithm of the initial pentanal concentration, 36.23 ppm, at the real emission source of the rendering facility, that is, $\ln C_0 = \ln(36.23) = 3.59$. As a result, the reaction rate constant of pentanal-hydrogen peroxide redox for the flue gas of this rendering plant is reasonably used as 0.536 1 s^{-1}.

After the oxidation of hydrogen peroxide in the oxidation reactor, the residual pentanal in the exhaust gas ranged from 0.68 to 2 ppm. The dispersion of residual pentanal was further simulated by Gaussian dispersion model (Wark *et al.*, 1997). Simulation conditions were set that the pentanal concentration in the area, 4.5 km × 3.5 km, with the chimney of the rendering plant in the center, should be below 0.41 ppb, which is the odor threshold concentration of pentanal. The grid spacing for the simulation was set as 100 m. The 2020 meteorological data of Kaohsiung Meteorology Station, which is the nearest station to the rendering plant in the southern Taiwan, including hourly average wind speed, wind direction, and atmospheric stability, were used for the neighboring air quality simulation. Additionally, the parameters included the height of the chimney (18.84 m), the inside diameter (60 cm) at the exit of the chimney, and the discharge gas temperature 80°C (353 K).

The atmospheric dispersion results of pentanal are summarized in Table 2. When the pentanal emission rate in the exhaust gas was set as 1 g s^{-1}, the daily and hourly average maximum values would be higher than the odor threshold. When the pentanal emission rate was decreased to 0.1 g s^{-1}, only the hourly average maximum value exceeded the odor threshold. When the pentanal

Table 2. Summary of pentanal mass emission rates simulated by Gaussian dispersion model (Wark *et al.*, 1997).

Pentanal emission rates (g s^{-1})	Maximum average concentration (ppb)		
	Annual	Daily	Hourly
1	0.2	0.56[a]	4.56[a]
0.1	0.02	0.056	0.456[a]
0.01	0.002	0.0056	0.0456

[a] The concentrations are higher than 0.41 ppb.

emission rate was decreased to 0.01 g s^{-1}, all average maximum values of the annual, daily, and hourly would be lower than the odor threshold of 0.41 ppb. Therefore, when the pentanal emission rate was controlled below 0.01 g s^{-1}, there should be no smell of pentanal within the neighboring area (4.5 km × 3.5 km) around the rendering plant.

4 CONCLUSIONS

From the viewpoints of public health and air quality, the exhausts from rendering plants must be treated using a novel technology of high treatment efficiencies for reducing the long-standing complaints from community residents for the odors (Vieira *et al.*, 2016; Sazakli and Leotsinidis, 2021; Sazakli *et al.*, 2022). Chemical oxidation technology has been developed and widely applied for abating industrial odors over 30 years, especially for the odor emissions from various biological sources (Cheremisinoff, 1992; Capelli *et al.*, 2019; Chou, 2009, 2019; Wysocka *et al.*, 2019; Zarra *et al.*, 2019; Senatore, 2021). However, aldehydes emitted from the rendering processes have been seldom treated by a chemical oxidation system using hydrogen peroxide as an oxidant. This study verified when the concentration of pentanal emitted from the rendering process was around 36.23 ppm in the gas streams with the flow rates from 100 to 250 Nm^3 min^{-1}, the reaction rate constant of pentanal for the first-order reaction by aqueous hydrogen peroxide of 1,000 mg L^{-1} was 0.536 1 s^{-1}, and then the pentanal concentrations after the oxidation by hydrogen peroxide were as low as 0.68-2 ppm. Actually, the removal efficiencies of pentanal exceed 80% for the batch feeding of hydrogen peroxide for a 10-min contact oxidation. This concentration ranges of pentanal in the exhaust gas stream resulted in the pentanal emission rate lower than 0.01 g s^{-1}, which would not cause complaints of the surrounding residents.

According to Anet *et al.*'s (2013) and Sazakli and Leotsinidis' (2021) reports, aldehydes are one of the most malodorous compounds those were emitted from the rendering plants. However, the current technologies, such as the bio-filtration, which have been applied to abate the mixture of VOCs cannot perform stable treatment efficiencies for all compounds. Based on the success of abatement of pentanal using hydrogen peroxide in this work, the development for treating air exhausts containing matrix of VOCs and odors is an key point to be implemented in the future.

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DISCLAIMER

The authors declare no conflict of interest.

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