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Coupling ozone and hollow fibers membrane bioreactor for enhanced treatment of gaseous xylene mixture

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HIGHLIGHTS

- ▶ This paper reports coupling ozone and hollow fibers membrane bioreactor (HFMBR) to treat VOCs.
- ▶ Ozone helps to enhance the max elimination capacity of HFMBR.
- ▶ The presence of ozone can avoid the formation of EPS and help stable long-term operation.

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ABSTRACT

Two hollow fiber membrane bioreactors (HFMBRs) inoculated with activated sludge were used in series to biodegrade continuously mixed xylene. The influence of gas residence time (τ) and mass loading rate (*LR*) on elimination capacity (*EC*) of the mixed xylene was investigated. A maximum elimination capacity ($EC_{max,v}$) of 466 g m^{-3} h^{-1} was achieved at τ = 10 s and LR_v = 728 g m^{-3} h^{-1}. Thereafter, ozone was introduced into inlet gas and the influence of ozone was investigated. Results showed that the maximum xylene elimination capacity increased from 524 g m^{-3} h^{-1} to 568 g m^{-3} h^{-1} and 616 g m^{-3} h^{-1} at τ = 10 s, respectively when the inlet ozone concentration rose from 200 mg m^{-3} to 400 mg m^{-3} and 600 mg m^{-3}, respectively. HFMBR coupled with O_3 has higher performance and stability for the long-term operation at the same condition.

1. Introduction

Volatile organic compounds (VOCs) are of great concern due to their toxicity, odor pollution and contribution to photochemical oxidants. Some of VOCs such as aromatic hydrocarbon and reactive alkenes have been identified to form ozone through combining with NO_x in the condition of exposure to UV light. Some of VOCs are air toxics and could bring about acute and chronic toxicity to human health and other biological impact through cumulative emission and accidental leakage, particularly in the local area. VOCs have been listed as one of the priority gaseous organic pollutants of chemical, petrochemical and allied industries (Khan and Ghoshal, 2000). Aromatic hydrocarbons, such as benzene, xylene, toluene and ethylbenzene, were the most crucial composition of VOCs because of their higher reactivity and ozone production potential. For instance, in the process of vent and storage tank of paint manufacturer, miscellaneous coating manufacturing, miscellaneous organic chemical production together with processes as well as toluene and xylene have been frequently detected.

The feature of waste gas stream including higher volume, miscellaneous pollutants and low concentration brings more difficulties for cost-effective purification technology. Consequently, biological treatment technique for VOC and odor control has gained tremendous popularity in view of its several advantages in comparison to the traditional physical and chemical methodology, such as incineration and adsorption (Khan and Ghoshal, 2000; Mudliar et al., 2010). The bioreactors applied commonly in industries for odor and VOC removal have three main types including biofilters, biotrickling filters and bioscrubbers (Shareefdeen and Singh, 2005). Although the three types of bioreactors have been extensively investigated and applied in the real waste gas treatment, the conventional biofiltration was also found limitations for VOCs removal. This is because that its operation condition is not stable and the phenomenon of clogging of the medium occurs frequently. Moreover, a large land area is needed for its setup, and so this is often a restriction where a land is costly or not available (Mudliar et al., 2010). Compared to the conventional

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biofiltration, membrane bioreactor (MBR) system has been proved to meet the above requirements and be capable of achieving high VOC removal efficiency in bench studies and pilot studies (Kennes and Veiga, 2001; Shareefdeen and Singh, 2005). Unlike the conventional MBR for waste water treatment widely applied (Robles et al., 2012), the MBR for waste air treatment still is scarce. Several reactor configurations such as spiral-wound (Luisa et al., 1995), tubular (Attaway et al., 2001, 2002), flat MBRs (De Bo et al., 2002; Kumar et al., 2008) and hollow fiber membrane (HFMBRs) were investigated as well, among which, hollow fiber has been studied extensively (Reij et al., 1998; Reij and Hartmans, 1996) attributing to its large specific gas membrane contact area (σ).

Xylene is an important chemical raw material. Organic synthesis, synthetic rubber, paints and dyes, synthetic fiber, petroleum processing, pharmaceuticals, cellulose production plant is the main source of xylene in the environment. Moreover, in order to improve the removal efficiency, particularly for recalcitrant volatile organic compounds with lower biodegradability, the pretreatment process via advanced oxidation has been investigated as the focus of this study. In addition, ozone oxidation was introduced into HFMBRs in this study to improve the mixed xylene removal performance.

2. Methods

2.1. Nutrient

The nutrient 1 used for the cultivation of microorganism and recirculation solution of HFMBRs contained in per liter of distilled water: 5.00 g of KNO_3, 3.00 g of NH_4Cl, 3.00 g of K_2HPO_4, 3.00 g of KH_2PO_4, 0.50 g of $MgSO_4{\cdot}7H_2O$ and 0.01 g of $FeSO_4{\cdot}7H_2O$ (Smet and Chasaya, 1996). The final pH of the medium was maintained in the range of 7.0–7.5. After Run 2, this nutrient was adjusted by replacing NH_4Cl with 5.66 g KNO_3 L^{-1}, resulting in a total concentration of 10.72 g KNO_3 L^{-1} (the nutrient 2) (Jacobs et al., 2004).

2.2. Lab-scale membrane bioreactor

Two HFMBs were used in series. Each HFMB consisted of 84 hydrophobic, microporous membrane polyvinylidenefluoride fiber bundles with the inner diameter of 1 mm, outer diameter of 1.5 m and length of 235 mm. The overall surface area of two HFMBs was 1.476 cm^2. This membrane was plotted into epoxy resin fittings at both ends, and immersed in water in a plexiglass cylinder which was 23 cm long and 5.0 cm internal diameter. The membrane bioreactor was then placed in a lab kept at the room temperature (20–24 °C). The scheme of the experimental system is shown in Fig. 1. For the generation of the synthetic waste gas stream, a metered stream of oil-free compressed air was passed through a flask in which liquid VOCs was allowed to evaporate as needed. This concentrated contaminant vapor was diluted to the expected concentration with the oil-free compressed air (Zhao et al., 2011). The waste gas stream was then delivered to the lumen of HFMBs and after that, it was degraded with and without ozone, respectively. Ozone mixed with VOCs and reacted to it in lab-scale membrane bioreactor where was after buffer bottle. Table 1 describes the operating conditions of various operational runs carried out in this investigation.

2.3. Analytical methodology

Gas phase xylene concentration was measured using a gas chromatograph (Agilent 6890II, Wilmington, US) equipped with a flame ionization detector (FID), a 30 m HP-5 capillary column (J&W Scientific, Folsom, CA) with the inner diameter of 320 μm and a film with the thickness of 0.25 μm. The temperature of the injector, detector and column was set at 120 °C, 180 °C and 250 °C, respectively. Highly purity nitrogen at a flow rate of 5 mL min^{-1} was adopted as the carrier gas, and the FID was fed with air at 400 mL min^{-1} and H_2 at 40 mL min^{-1}. A 0.1 mL air stream sample was taken from the injector and injected into the gas chromatograph using a gas-tight syringe. The cross and surface structure of PVDF were obtained by membrane scanning electron microscope (SEM) (S-4800, Hitachi, Japan). An optical microscope (Eclipse 80i Nikon, Tokyo, Japan) was used for bacterial observation. TOC value in circulating liquid was analyzed by TOC analyzer (liquil TOC, Elementar, Germany).

2.4. Calculation

All terms were used in conjunction to describe the performance of the system. They are defined as:

$$LR_v = C_{in}/\tau \tag{1}$$

$$EC_v = (C_{in} - C_{out})/\tau \tag{2}$$

$$\eta = 100\%\ (C_{in} - C_{out})/C_{in} \tag{3}$$

in which LR_v is the volumetric loading rate (g m^{-3} h^{-1}). C_{in} is the inlet concentration (mg m^{-3}). τ is the gas residence time based on the lumen volume. EC_v is the volumetric elimination capacity (g m^{-3} h^{-1}). C_{out} is the outlet pollutant concentration (mg m^{-3}) and η is the degradation efficiency (%). In this study, the loading rate and elimination capacity per unit membrane area (LR_m, EC_m, mg m^{-3} h^{-1}) was correlated with LR_v and EC_v, respectively, by the specific membrane area of 4000 m^2 m^{-3} in the hollow fiber module.

3. Result and discussion

3.1. Membranes

Two types of membrane were used and compared in membrane bioreactor: dense phase and microporous. Dense phase membranes generally features a lower permeability but allowing for the selectivity in permeation. On the contrary, microporous membranes offer high rates of flux without selectivity (Reij et al., 1998).

The size of the membrane pore was in the range about between 0.1 μm and 0.2 μm determined by the liquid permeation, whose osmotic pressure and porosity was 15 kPa and 40%, respectively. The SEMs shows the cross section of PVDF membrane presents successive trend without stratified structure. Its surface was rough which was beneficial for zooglea to adhere to it. Internal structure of PVDF membrane was loose and pore distribution was sufficient, therefore the membrane offered extremely high rates of flux for both oxygen and VOCs.

3.2. Inoculation and start-up of the HFMBR

The membrane bioreactor was inoculated by the aerobic activated sludge derived from a municipal wastewater treatment plant which locates in the Xuhui district in Shanghai with the purpose of utilizing as the inoculum. The initial pH value, SVI of activated sludge was around 7.0 and 120, respectively. Its mixed liquid suspended solids (MLSS) and volatile suspended solids (VSS) was 5 g L^{-1} and 3.5 g L^{-1}, respectively. The sludge was then bubbled with the synthetic waste air lasting for several weeks in the nutrient 1. The inlet xylene concentration was set within a specific range. The gaseous mixed concentration was measured daily at both inlet and outlet. The enriched consortium was added into the aqua storage tank.