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Long-term MIBK removal in a tubular biofilter: Effects of organic loading rates and gas empty bed residence times

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ABSTRACT

A novel gas biofilter, namely the tubular biofilter (TBF) was developed and successfully used for toluene removal from waste gas streams. To furtherly explore its performance for methyl isobutyl ketone (MIBK) removal, the TBF was continuously operated for over 284 days under various organic loading rates (OLRs) (ranging from 20 to 340 g m^{-3} h^{-1}) and gas empty residence times (EBRTs) (ranging from 15 to 5 s) in this paper. The results show that the TBF could achieve a high MIBK removal efficiency (RE) at a short gas EBRT of 5 s. The mean maximum REs of MIBK were 98.67%, 88.95%, 77.93%, 71.74%, and 55.92% under the OLRs of 19.89, 39.33, 78.96, 160.17, 338.02 g m^{-3} h^{-1}, respectively. The maximum elimination capacity for MIBK was as high as 200 g m^{-3} h^{-1}, while the critical load was about 20 - 30 g m^{-3} h^{-1}. Based on the Michaelis-Menten kinetic model, the biokinetic constants K_s was found to be 8.56 g m^{-3} and r_{max} was 0.14 g m^{-3} h^{-1}. The biomass clogging appeared at the EC of over 80 g m^{-3} h^{-1}. The TBF showed a high efficient performance for MIBK removal due to the enhancement of highly porous polyurethane sponge and new tubular configurations.

1. Introduction

Methyl isobutyl ketone (MIBK) is commonly used for manufacturing paints, rubbers, pharmaceuticals, chemicals, and machinery, and can be harmful to humans (irritation of mucous membrane and weakness of central nervous system) if exposed to a concentration of 50–100 ppm (Kawai et al., 2003; Raghuvanshi and Babu, 2010; Datta and Philip, 2014). Therefore, stringent requirements are enforced for controlling the discharge of waste gases containing MIBK. As a cost-effective and environment-friendly technology, biofiltration has been widely used for removing odors and volatile organic compounds (VOCs) from waste gas streams (Amin et al., 2017; Chang et al., 2015; Wang et al., 2012; Zhang et al., 2017), which also includes the removal of MIBK (Lee et al., 2006; Cai and Sorial, 2009; Farnazo et al., 2012). Deshusses et al. (1995, 1996) and Deshusses and Johnson (2000) investigated the transient behavior of biofilters for MIBK removal from waste gases, and extended the methods to evaluate their performances. A laboratory-scale trickle-bed air biofilter packed with 6 mm Celite pellets (R-635) was evaluated for removing MIBK from a waste gas stream (Cai et al., 2005). *Rhodococcus globerulus*, *Gordonia terrae*, *Gordonia bronchialis* and *Bacillus subtilis* were identified as the active strains in a biofilter, whereas *Gordonia terrae* exhibited the highest biodegradation rate in a suspension for removing MIBK (Przybulewska and Wieczorek, 2008). However, conventional biofilters and trickling biofilters (or biotrickling filters) are still the most commonly implemented bioreactors in both the laboratory and industrial applications for MIBK removal from waste gases (Deshusses et al., 1996; Cai et al., 2005).

In fact, large footprints and performance deterioration during long-term operation of both conventional biofilters and biotrickling filters (Ryu et al., 2010; Rene et al., 2011; Cheng et al., 2016; Skerman et al., 2017) hinder their large-scale applications. To overcome such problems and enhance the reactor performance, especially in maximizing the pollutant removal capacity, a wide range of new biofilter configurations were developed (Yang et al., 2010b). Some of these configurations included rotating drum biofilter (RDB) (Yang et al., 2008a, 2008b), rotating biological contactor

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Nomenclature and units

b	Shear/decay coefficient, d^{-1}
b_d	Decay rate of biomass, 0.432 d^{-1}
b_{s0}	Default shear rate coefficient, 0.005 d^{-1}
B	mass of biomass (dry weight), gVSS
$C_{gas,in}$	Inlet isobutanol concentration, g m^{-3}
$C_{gas,out}$	Outlet isobutanol concentration, g m^{-3}
C_{ln}	the log mean concentration
ε_0	Clean bed porosity with water film, 90%
$\varepsilon(t)$	Porosity in bed with biofilm at time t, %
EBRT	Empty bed residence time, s
EC	Elimination capacity, g m^{-3} h^{-1}
OLR	Organic loading rate, g m^{-3} h^{-1}
Q	Inlet gas flow rate (volumetric), m^3 h^{-1}
γ	Conversion factor of actual space volume, dimensionless
r_{max}	maximum degradation rate per unit filter volume, g m^{-3} h^{-1}
ρ_b	Average dry density of the biomass, gVSS L^{-1}
K_s	Saturation constant, g m^{-3}
RE/η	Removal efficiency, %
t	Elapsed time, d
TBF	Tubular biofilter
V_f	Apparent volume of the filter media, m^3
Y	Biomass yield, g biomass g^{-1} MIBK

(Datta and Philip, 2014), foamed emulsion bioreactors (Kan and Deshusses, 2005), stirred tank bioreactors (Bailón et al., 2009), air diffusion bioreactor (Lebrero et al., 2010), rotating rope biofilter (Mudliar et al., 2010), modified rotating biological contactor (Ravi et al., 2010), and tubular biofilter (TBF) (Chen et al., 2012). Among these new technologies, TBF seems to be the most promising, and is composed of a closed chamber, a module polyurethane sponge tube and a nutrient solution distributor. The TBF has shown good long-term (391 days) performance for toluene removal with no excessive biomass accumulation in the filter media (Chen et al., 2012). For isobutanol removal from waste gases using TBF, the elimination capacity (EC) was found to be 60.42 g m^{-3} h^{-1} at the inlet OLR of 66.45 g m^{-3} h^{-1}, whereas the critical EC was around 30 g m^{-3} h^{-1} (Chen et al., 2018).

Despite the development of dozens of new bioreactor configurations, the removal of MIBK using new biofilters under long-term operation has not yet been reported in literature. Some key operational parameters, especially the organic loading rate (OLR) and gas empty residence time (EBRT), can have great impact on the pollutant removal performance for biofiltration process (Yang et al., 2008b; Chen et al., 2012; Tu et al., 2015). Therefore, the treatment of waste gases for MIBK removal using TBF was urgently needed. Such a study can expand the application scope of new biofilters, and can also improve the removal capacity of biofilters for MIBK. In this study, a pilot-scale TBF was studied, which used MIBK as the model VOC. Experiments were conducted to investigate the effects of OLR (ranging from 20 to 340 g m^{-3} h^{-1}) and gas EBRT (ranging from 15 to 5 s) on the performance of TBF for continuous operation for 284 days.

2. Materials and methods

2.1. Experimental apparatus

The TBF that operated in continuous mode was developed in our previous study (Chen et al., 2012, 2018), which consisted of a closed cylindrical chamber, a module polyurethane sponge tube and a nutrient solution distributor, was used to assemble the experimental set up for the current work. The closed chamber had the height and inner diameter of 15 cm and 16 cm, respectively, and consisted of a plexiglas pipe with two perforated flange cover plates at both ends. A module tube of polyurethane sponges (Shenzhen Jiechun Filter Material Co., Ltd., Guangdong, China) having a porosity of 98% and a pore size of approximately 0.8 mm (30 PPI) was used. The tubular media was perpendicularly mounted in the center of the closed cylindrical chamber, which was located between the nutrient solution distributor and the bottom of the chamber, and had an outer diameter of 14 cm, radial thickness of 3 cm, and height of 10 cm. The schematic of the experimental apparatus with the tubular filter media is shown in Fig. 1.

The simulated waste gases entered the TBF through gas inlet located at the center of the top surface of the chamber, flowed over the nutrient solution distributor and passed through the module polyurethane sponge tube. The biofilm attached onto the filter media, and biodegraded contaminants in the waste gas stream. The purified gas was finally discharged out of the chamber through the gas outlet that was located at the center of the bottom. Meanwhile, the nutrient solutions were pumped into the TBF at a rate of 6.0 L d^{-1}, which was controlled using both a rotameter and a repeat cycle timer (1-min on/30-min off). A nutrient solution distributor, which was one of the key components of the TBF, was assembled for uniformly distributing the nutrients on the module sponge tube. A nutrient solution tank for the regeneration and circulation of nutrients collected the effluents at the bottom of the chamber. More details about the experimental apparatus and nutrient solutions can be founded in Chen et al. (2012).

2.2. Chemical and biological materials

Analytical methyl isobutyl ketone (MIBK; $C_6H_{12}O$; 99%; Damao Chemical Industry Co., Ltd., Tianjin, China) was used as the target contaminant while preparing the simulated waste gas, which was synthesized using clean air and MIBK vapors. The flow rates of both streams could be adjusted based upon the operational needs. Some commercial analytical reagents, including sodium bicarbonate, sodium nitrate, potassium dihydrogen phosphate, potassium hydrogen phosphate, calcium chloride, magnesium sulfate, ferric chloride, sodium chloride, cobalt chloride, copper chloride, manganese chloride, vitamin B_2, nicotinic acid, and folic acid were used for preparing the nutrient solutions. The nutrient solution was kept at a pH of 7.0 ± 0.2. The fresh activated sludge was taken from a secondary sedimentation tank of a municipal wastewater treatment plant (Changsha Guozhen Wastewater Treatment Co., Ltd., Hunan, China) and used for seeding the TBF.

2.3. Analytical methods

When the nutrient feeding was underway, a U-type water manometer (Ruijun Electronic Technology Co., Ltd., Nanjing, China) was employed for determining the pressure drop. Gas-phase measurements included MIBK concentrations in the influent and effluent gas streams, and were determined everyday using a gas chromatograph (GC) (HP 6890 N; Series II; Hewlett-Packard, Palo Alto, California, USA). The GC was equipped with a flame ionization detector and with a HP-VOC capillary column (60 m × 320 μm ID × 1.8 μm; Agilent, USA). The temperatures of 120, 120, and 250 °C were maintained for the GC injector, the oven, and the detector, respectively. Highly pure nitrogen gas (99.9%) with the flow rate of 30 mL min^{-1} was used as the carrier gas. The sample measurements were triplicated for calculating the average values, and their corresponding standard deviations were determined using OriginPro2015 (OriginLab Co.). To observe and photograph the biological phases based on the removal performance and the pressure drop,