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Gas Dimethyl Sulfide Removal in Biotrickling Filtration

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Abstract

The biotrickling filter packed with ceramsite was set up to study the removal of dimethyl sulfide (DMS). The DMS removal efficiency in the biotrickling filter was up to 99% based on experimental results. The optimal spray density, empty bed residence time (EBRT) and pH are 100 mL.min^{-1}, 38 s and 6.0, separately. The microbial community composition taken from packing material samples in the biotrickling filter for removal of DMS developed, which were assessed by polymerase chain reaction-denaturing gradient gel electrophoresis (PCR-DGGE) of eubacterial 16S rDNA followed by clone library analysis, revealed four distinct bands. Phylogenetic analysis showed that the sequences of these bands were closest to sequences of species of the *Bacillus sp, Rhodobacteraceae bacterium, proteobacterium, delta proteobacterium.*

Keywords: Biotrickling filter; Odour; Dimethyl sulfide (DMS); PCR-DGGE; Microbial community; Biodegradation

Introduction

Odours emitting industrial activities, such as sewage treatment plants, waste treatment or disposal facilities, paint facilities, petroleum refineries, rendering plants, pulp mills, plastic and resin manufacturers and chemical industries, and that cause an odour nuisance problem, are often classified as contaminants and are subject to regulation [1]. Odours may cause a variety of undesirable reactions in people, ranging from annoyance to documented health effects [2]. Volatile organic sulfur compounds (VOSC) are main environmental odour contaminants, which includes methanethiol (CH3SH), dimethyl sulfide (CH_3SCH_3, DMS), dimethyl disulfide ($CH_3S_2CH_3$, DMDS). VOSC are characterized by their hightoxicity, potential corrosive effect, and very low odour threshold values (OTV), e.g. 0.6–40 ppbv for dimethyl sulfide (CH_3SCH_3, DMS) [3,4].

Biofiltration has been known as an efficient waste gas control technology for treatment VOSC at low cost of maintenance, and produces harmless by-products. Two *Hyphomicrobium* VS inoculation protocols were compared for start-up of a biotrickling filter removing dimethyl sulfide (DMS) [5]. A dynamic model was developed that described the removal of DMS in the presence of MeOH in inorganic biofilters under both steady and transient conditions [6]. Biological treatment of dimethyl sulphide (DMS) was investigated in a bench-scale biofilter, packed with compost along with wood chips, and enriched with DMS degrading microorganism *Bacillus sphaericus* [7]. Dimethyl sulfide was removal in a thermophilicbiotrickling filter operated at 52°C, using an enriched sludge inoculum [8]. The membrane bioreactor contained a polydimethylsiloxane/Zirfon composite membrane and inoculated with *Hyphomicrobium* VS, a methylotrophic micro-organism was used to remove dimethyl sulfide from waste air [9]. The biofilter process and bacterial community composition are key elements for biodegrading of dimethyl sulfide (DMS). Hydrogen sulfide, methanethiol, dimethyl sulfide and dimethyl disulfide was degradated by *Hyphomicrobium*DW44 isolated from peat biofilter [10]. Dimethyl sulfide was conversed by *Methylophagasulfidovoran* in a microbial mat [11]. A PCR-DGGE approach and constructed a dendrogram had been used to illustrate the diversity of the bacterial community in a biofilter at different operating conditions. The diversity of the bacterial community in the biofilter is dynamic and varies with inlet DMS loads, the addition of glucose, and fluctuating temperature [12].

In this study, experimental investigations were conducted to remove of the odor containing dimethyl sulfide (DMS) in biofilter filled with the ceramsite as a medium. The study analysis bacterial community composition in biofilters assessed by polymerase chain reaction-denaturing gradient gel electrophoresis (PCR-DGGE) followed by clone library analysis, and evaluates the factors such as the influence of inlet concentration, empty bed residence time (EBRT), inlet concentration, biological oxidation on the performance of the biofilter system.

Materials and Methods

Experimental procedure

The flow loop used in the study is shown schematically in figure 1. The dimethyl sulfide supplied from the gas cylinders, was first diluted with the compressed air, passed through an air mixture bottle, then flowed upwards the bottom of the biofilter. The biofilter column (internal diameter of 90 mm and 1200 mm long) was packed with ceramsite (external diameter of 8 to 15 mm) to a height of 510 mm, which was set up to study removal of dimethyl sulfide from stimulated waste gas. It was divided into three sections with the filter medium at each section was supported on a stainless steel sieve plate that ensured homogeneous distribution of gas flow over the entire cross section of the filter bed; biodegrading bacterials adhere to the surface of ceramsite to form the biofilm, the microbial inoculum culture was obtained by acclimating the activated sludge taken from the local wastewater treatment plant.

Dimethyl sulfide concentrations were monitored by the analysis device of MiniRAE PLUS PGM-7600 Photo-Ionization Detector, and gas flow rate was monitored by the rotameter and the mass flow controllers. Gas flow rates were measured using Model LZB^{-1} flow

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meters with units of 1 l/h. The pH values were measured by a Model pHB-3 pH Tester (Sanxin Instrument Company, Shanghai, China). In the process of the biodegradation of dimethyl sulfide experiments, nutrient-containing aqueous solutions was sprayed downward at a rate of 3 ~ 18 $L.h^{-1}$ with a peristaltic pump from the top of column to maintain the moisture of the biofilter and supply nutrients to the microbial population. The simulated dimethyl sulfide-containing waste gas was supplied to the biofilter, at a flow rate of 100 to 600 $L.h^{-1}$ (EBRT, 19 to 114s).

Bacterial community analysis by PCR-DGGE

Bacterial community compositions in the biotricking filter were assessed by polymerase chain reaction-denaturing gradient gel electrophoresis (PCR-DGGE). Following cell lysis, DNA extraction, and PCR amplification were as described by Ho et al. (2008). Two primers, P1(5'>CGCCC GCCGC GCGCG GCGGG CGGGG CGGGG GCACG GGGGG CCTAC GGGAG GCAGC AG<3') and P2 (5'>ATTAC CGCGG CTGCT GG<3') were used to amplify the segment of eubacterial16S rDNA. Samples (0.5g) of packing material were removed from the biotrickling filter, mixed with 10 ml distilled water, and vortexed for 20 min. The samples were run on an 8% acrylamide gelwith a 30-68% denaturant gradient using a Bio-Rad DGGE apparatus, at 60°C and a constant voltage of 180 V for 300 min. The DGGE bands chosen for cloning were excised, and then eluted, re-amplified, and sequenced. The sequencing products were analyzed with an Applied Biosystems 377 DNA sequencer. The BLASTN program was used to search for nucleotide sequence similarityin the NCBI website. Sequences recovered from excised bands were analyzed for chimeric character using the Ribosomal Database ProjectII (RDP II) Chimera.

Results and Discussion

Performance of the biofilter system

Figure 2 shows the removal performance of the biotricking filter for DMS gas removal during the 36-d continuous running test. The conversion of dimethyl sulfide biodegradation efficiency increases from 5.7% with one day to 98.8% at 36th d, showing good dimethyl sulfide degradation effect. Dimethyl sulphide biodegradation efficiencies were 97-99% with inlet concentrations of 12.8-63 $mg.m^{-3}$ from 24 to 36-day operating time. In the biofilter, dimethyl sulfide air stream is forced to pass through a ceramsite support material on which pollutant degrading cultures are immobilized. Dimethyl sulfide and oxygen diffuse from the gas phase to the wet layer of the biofilm and then are consumed by the microorganism communities. Under aerobic conditions in a biofilter, dimethyl sulfide is oxidized to carbon dioxide, sulfide (SO_4^{2-}), water vapors by biological oxidation; dimethyl sulfide solubility is small in water due to its low Henry's constants, mass transfer limitation may play an important role during biological treatment; gas-phase dimethyl sulfide should first diffuse through a thin aqueous layer surrounding the filter medium, and then dimethyl sulfide is directly adsorption to the surfaced of biofilm, biological oxidation is the process in which dimethyl sulfide is oxidized to CO_2 and H_2O.

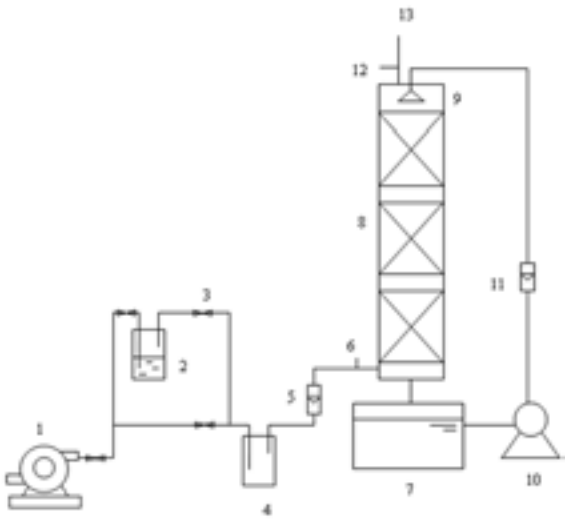

Figure 1: Experimental flow loop of biodegradation of gas-phase dimethyl sulfide. (1) dimethyl sulfidegas cylinder; (2) air compressor; (3) the bottle of gas mixture; (4) flow mete; (5) biofilter column; (7) nutrient tank; (8) peristaltic pump; (9) outlet port; (G) sampling port.

The influence of dimethyl sulfide concentration

Keeping EBRT of 36 s, and sprinkling amount (6.0 $L.h^{-1}$), pH of 6.0 fixed, the influence of dimethyl sulfide concentration in inleton removal of dimethyl sulfide with the biofilter are presented in figure 3. The conversion of dimethyl sulfide removal efficiency reduces from 100% with 5.5 $mg.m^{-3}$ to 54% with 249 $mg.m^{-3}$ dimethyl sulfide. More

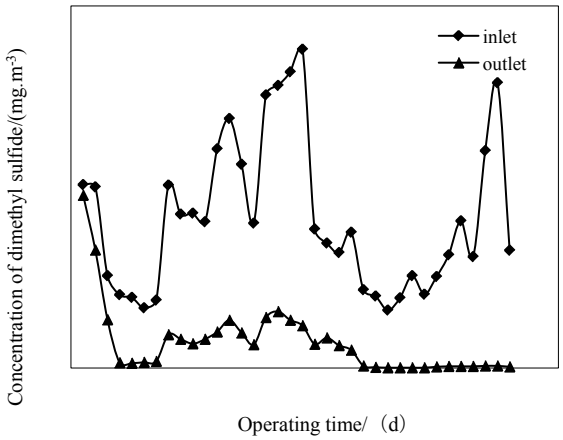

Figure 2: Performance of the biotricking filter of dimethyl sulfide during the 36-d continuous running test.

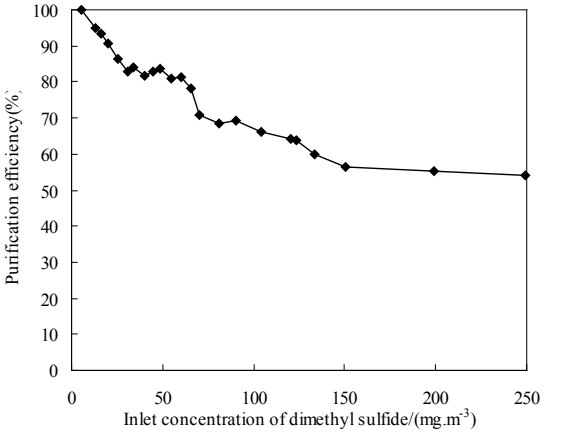

Figure 3: Influence of concentration of dimethyl sulfide in inlet on dimethyl sulfide removal.

than 80% dimethyl sulfide is biological oxidized for less than the initial concentration of 60 mg.m^{-3} dimethyl sulfide. This illustrates that the biological reactor is efficient in purifying the waste gas whose dimethyl sulfide concentration is between 5.5 mg.m^{-3} and 249 mg.m^{-3}. The biofilter to photocatalytic reactor eliminates gas-phase dimethyl sulfideto produce CO_2, H_2O.

The influence of empty bed residence time (EBRT)

The effect of EBRT on removal of dimethyl sulfide is presented in figure 4, under the conditions of pH of 6.0, inlet concentration of 20 mg.m^{-3}dimethyl sulfide andsprinkling amount at 6.0 L.h^{-1} in the biotreatment system. Dimethyl sulfide biodegradation efficiency increases from 63.1 to 100% with EBRT increasing (Figure 3). This indicates the longer residence time is a benefit on DMS removal, in the case where the EBRT is too short to biological oxidized dimethyl sulfide to SO_4^{2-}, CO_2, H_2O before release. The degradation DMS microorganisms and volume of biofilter with the microorganisms are the key elements. From figure 4, in our experimental conditions, we can assume the optimum EBRTis 36 s in the system, and about 90% dimethyl sulfide in the gas stream is converted.

The influence of pH

Maintaining constant pH in the biofilter system is an important operating factor. To estimate the biofilter response to pH variations, the inlet concentration of dimethyl sulfide was controlled at 20 mg.m^{-3}, EBRT of 36s and the average removal efficiencies were calculated over a 36-d experiment for every pH change (4.0-7.5) as shown in figure 5. Approximately 72.9–90.4% removal efficiencies for dimethyl sulfide are achieved in the range of 4.0–7.5. When the pH rises from 5.5 to 6.0, dimethyl sulfide removal efficiency increases from 88.7 to 90.4%. In contrast, dimethyl sulfide removal efficiency decreases to 77.9% when the pH is raised to 7.5. Although pH changes do not result in significant decreases in cell numbers, the dimethyl sulfide $_x$biodegradation is inefficient at low and high pH, and a decrease in purifying efficiency was observed. The optimal pH in the biofilter for dimethyl sulfide removal ranges from 5.5 to 6.5, with especially good results at pH 6.0, it is suspected that a neutral pH resulted in maximal enzyme activities for dimethyl sulfide degradation. During long term operation, sulfide (SO_4^{2-}) concentration in circulation liquid should increase; pH may be adjusted to maintain bacterial activities by means of adding alkali to recycled liquid. Therefore, periodic replacement of circulation liquid is required in order to control sulfide (SO_4^{2-}) concentrations.

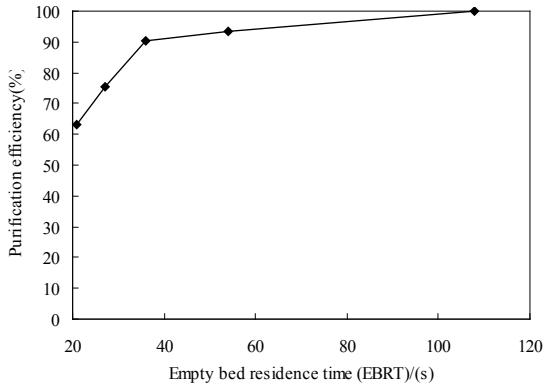

Figure 4: Influence of EBRT on dimethyl sulfide removal.

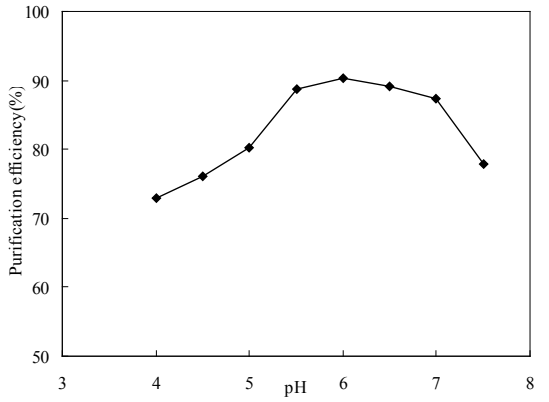

Figure 5: Influence of pH change on dimethyl sulfide removal.

The influence of sprinkling amount

Figure 6 shows influence of sprinkling amount on DMS removal. With sprinkling amount increasing, DMS removal efficiency increases from 89 to 90.4% with sprinkling amount increasing from 3 to 6l.h^{-1}, decreases from 90.4% with 6l.h^{-1} to 73.6% with 18l.h^{-1}. This indicates that dimethyl sulfide may be not more absorbed with increasing sprinkling amount. Because dimethyl sulfide solubility is small in water, the scrubbing effect of water could hardly play a role in dimethyl sulfide removal, dimethyl sulfide is oxidized to carbon dioxide, sulfide (SO_4^{2-}),water vapors by biological oxidation at the steady state in biofiltration process, dimethyl sulfide biodegradation efficiency remain attained 92% maintaining adequate moisture in the filter bed without supply of water from the top of bioreactor. From figure 6, in our experimental conditions, we can assume the optimum sprinkling amountis 6l.h^{-1}, and about 90.4 % dimethyl sulfidein the gas stream is converted.

Bacterial community composition by PCR-DGGE

DMS is directly adsorption to external surfaced biofilm of ceramsite, it is attached, degraded by microorganism in the biofilm. Biological processes are currently run under mesophilic conditions, at temperatures below 35°C. The biomass concentration in the biofilter, which is also an important factor for biofilter performance, does not appear to be influenced in lowering the purification efficiency as can be seen from the nearly uniform concentration of biomass (1.8×10^6 to 2.2×10^8 CFU g^{-1} of compost) throughout the operation of the biofilter. Electron micrographs of DMS -degrading strain is shown in figure 7 (SEM photographs). The microbial community

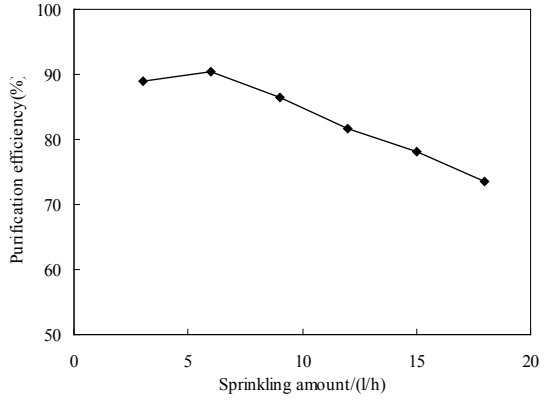

Figure 6: Influence of sprinkling amount on dimethyl sulfide removal.

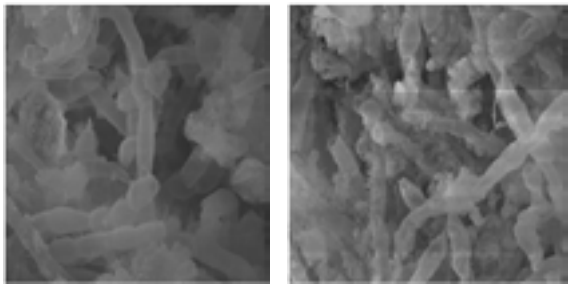

Figure 7: SEM images of DMS -degrading strain.

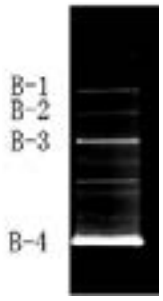

Figure 8: DGGE pattern of different strains (B-1, B-2, B-3, B-4).

structures in the biotrickling filter for dimethyl sulfide removal were assessed by polymerase chain reaction-denaturing gradient gel electrophoresis (PCR-DGGE). Denaturing gradient gel electrophoresis of eubacterial 16S rDNA samples taken from packing material revealed four distinct bands (Figure 8). Based on 16SrDNA sequence data, results show that the predominant bacterias for degradation of DMSare Bacillussp, *Rhodobacteraceae bacterium, proteobacterium, delta proteobacterium*. The dominant bacteria, *Bacillus* sp., takes up 68.6%; while *Rhodobacteraceae bacterium*, *proteobacterium* and *delta Proteobacterium* are take up 14.8%, 2%, 2.8%, respectively. *Bacillus* sp. was very predominant in its role of DMS-degrader, enhancing the metabolism of DMS in the biofilter. *Bacillus* sp., sulfur-oxidizing bacteria, was able to degrade H_2S [13,14]. *Proteobacterium* [15], oxidizing inorganic sulfide and mercaptans, and *Rhodococcus*, deodorizing domestic animal feces [16] have been described as sulfide oxidizers. Since DMS can be metabolized to dimethyl sulfoxide, methyl mercaptan, hydrogen sulfide, and sulfate [17], this predominant bacteria may be attributable to the potential for sulfur oxidation and carbon oxidation processes to occur simultaneously in the biotrickling filter system. Under aerobic conditions in a biofilter, dimethyl sulfide is oxidized to carbon dioxide, sulfide (SO_4^{2-}). Biooxidation of sulphide and intermediary sulphur compounds carried out by sulphide oxidizing bacteria are crucial in biotreatment of acidmine drainage and in the bioleaching of refractory minerals.

Conclusions

The paper revealed that the biotrickling filter packed with ceramsite could be used forremoval of dimethyl sulfide from waste gas. DMS removal could be achieved with high efficiency in the biotrickling filter. The optimal spray density, empty bed residence time (EBRT) and pH are 100 $mL.min^{-1}$, 38 s and 6.0, separately. PCR-DGGE was performed to study the 16S rRNA gene fragment profiles of microbial community composition taken from packing material samples in the biotrickling filter for removal of DMS. The research showed that this bacteria of purifying DMS is *delta proteobacterium, proteobacterium, Rhodobacteraceae bacterium, Bacillus* sp. The strains identified are potential candidates for purifying waste gas containing DMS.

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