

# Gas Dimethyl Sulfide Removal in Biotrickling Filtration

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V@^hãä[ciã&\jã] \*Á, |c^!Á] æ&\^ãÁ, ìc@Á&^!æ { •ìc^Á, æ•Á•^cÁ } ]Ác [Á•c~ã^Ác@^Á!^ { [çæ]Á[-hãä { ^c@^!Á•~ ], á^ÁçÖTÙDEÁV@^ÁÖTÙÁ ;^ { [çæ]Á^-, &ã^ } &^hã } Ác@^hãä[ciã&\jã] \*Á, |c^!Á, æ•Á~ ]Ác [ÁJJÁhãæ•^hã ] Á^ç ] Á^ä { ^ } çæ]Á!^•~ |c^ÉÁV@^Á [ ] cá { æ]Á• } ;æ^Áh^ } •ìc^ÉÁ empty bed residence time (EBRT) and pH are 100 mL.min<sup>-1</sup>, 38 s and 6.0, separately. The microbial community & [ { ] [ •ìcä [ ] Ácæ\^ } Á-! [ { Á ] æ&\jã } \*Á { æc^!æ]Á •æ { ] |^•Áã } Ác@^hãä[ciã&\jã] \*Á, |c^!Á-[!Á!^ { [çæ]Á [-ÁÖTÙÁ á^ç^ ] ] Á^ãÉÁ, @ì&@Á 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 (CH<sub>3</sub>SH), dimethyl sulfide (CH<sub>3</sub>SCH<sub>3</sub>, DMS), dimethyl disulfide (CH<sub>3</sub>S<sub>2</sub>CH<sub>3</sub>, DMDS). VOSC are characterized by their high toxicity, potential corrosive effect, and very low odour threshold values (OTV), e.g. 0.6–40 ppbv for dimethyl sulfide (CH<sub>3</sub>SCH<sub>3</sub>, DMS) [3,4].

Biotrickling has been known as an efficient waste gas control technology for treatment VOSC at low cost of maintenance, and produces harmless by-products. Two

*Biodegrading microorganism Bacillus sphaericus* [7]. Dimethyl sulfide was removed in a thermophilic biotrickling 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 microorganism was used to remove dimethyl sulfide from waste air [9].

The biotrickling process and bacterial community composition are key elements for biodegradation of dimethyl sulfide (DMS). Hydrogen sulfide, methanethiol, dimethyl sulfide and dimethyl disulfide was degraded by *Hyphomicrobium* DW44 isolated from peat biotrickling filter [10]. Dimethyl sulfide was converted by *Methylophaga sulfidovorana* 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 biotrickling filter at different operating conditions. The diversity of the bacterial community in the biotrickling filter 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 the odor containing dimethyl sulfide (DMS) in biotrickling filter with the ceramsite as a medium. The study analyzed bacterial community composition in biotrickling filters assessed by polymerase chain reaction-denaturing gradient gel electrophoresis (PCR-DGGE) followed by clone library analysis, and evaluates the factors such as the inlet concentration, empty bed residence time (EBRT), inlet concentration, biological oxidation on the performance of the biotrickling filter 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 biotrickling filter. The biotrickling filter 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 biotrickling 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 bacteria 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<sup>-1</sup> flow

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April 26, 2012;

May 22, 2013

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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 sulfoxide experiments, nutrient-containing aqueous solutions were sprayed downward at a rate of 3 ~ 18 L.h<sup>-1</sup> 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 sulfoxide-containing waste gas was supplied to the biofilter, at a flow rate of 100 to 600 L.h<sup>-1</sup> (EBRT, 19 to 114s).

### Bacterial community analysis by PCR-DGGE

Bacterial community compositions in the biotrickling 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 CCGGG CCGGG GCACG GGGGG CCTAC GGGAG GCAGC AG-3') and P2 (5'-ATTAC CGCGG CTGCT GG-3') were used to amplify the segment of eubacterial 16S 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 gel with 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, reamplified, and sequenced. The sequencing products were analyzed with an Applied Biosystems 377 DNA sequencer. The BLASTN program was used to search for nucleotide sequence similarity in the NCBI website. Sequences recovered from excised bands were analyzed for chimeric character using the Ribosomal Database Project II (RDP II) Chimera.

## Results and Discussion

### Performance of the biofilter system

Figure 2 shows the removal performance of the biotrickling filter for DMS gas removal during the 36-d continuous running test. The conversion of dimethyl sulfoxide biodegradation efficiency increases from 5.7% with one day to 98.8% at 36<sup>th</sup> d, showing good dimethyl sulfoxide

degradation effect. Dimethyl sulphide biodegradation efficiencies were 97-99% with inlet concentrations of 12.8-63 mg.m<sup>-3</sup> from 24 to 36-day operating time. In the biofilter, dimethyl sulfoxide air stream is forced to pass through a ceramsite support material on which pollutant degrading cultures are immobilized. Dimethyl sulfoxide 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 sulfoxide is oxidized to carbon dioxide, sulphate (SO<sub>4</sub><sup>2-</sup>), water vapors by biological oxidation; dimethyl sulfoxide 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 sulfoxide should first diffuse through a thin aqueous layer surrounding the filter medium, and then dimethyl sulfoxide is directly adsorption to the surface of biofilm, biological oxidation is the process in which dimethyl sulfoxide is oxidized to CO<sub>2</sub> and H<sub>2</sub>O.

### Effect of dimethyl sulfoxide concentration

Keeping EBRT of 36 s, and sprinkling amount (6.0 L.h<sup>-1</sup>), pH of 6.0 fixed, the effect of dimethyl sulfoxide concentration in inlet on removal of dimethyl sulfoxide with the biofilter are presented in Figure 3.

than 80% dimethyl sulfoxide is biological oxidized for less than the initial concentration of 60 mg.m<sup>-3</sup> dimethyl sulfoxide. This illustrates that the biological reactor is efficient in purifying the waste gas whose dimethyl sulfoxide concentration is between 5.5 mg.m<sup>-3</sup> and 249 mg.m<sup>-3</sup>.

The biofilter to photocatalytic reactor eliminates gas-phase dimethyl sulfoxide to produce CO<sub>2</sub>, H<sub>2</sub>O.

#### **The influence of empty bed residence time (EBRT)**

The effect of EBRT on removal of dimethyl sulfoxide is presented in Figure 4, under the conditions of pH of 6.0, inlet concentration of

*Rhodobacteraceae bacterium, Bacillus* sp. e strains identi ed are potential candidates for purifying waste gas containing DMS.

Foundation of Guangdong Zhuhai science & technology (PC20082029).

- Sironi S, Capelli L, Céntola P, Rosso RD, Pierucci S (2010) Odour impact assessment by means of dynamic olfactometry, dispersion modelling and ...
- Nicell JA (2009) Assessment and regulation of odour impacts. *Atmos Environ* 43: 196-206.
- Öhler A, Scharf S, Rühl Y, Öhler S, ... (2010) Mediated heterogeneous photocatalytic degradation of gaseous dimethyl ...
- Vogelbein B, ... (2010) Various biological matrices. *J Chromatogr B* 877: 3366-3377.
- Zhang Y, ... (2010) ...
- Öhler A, ... (2010) ...
- Öhler A, ... (2010) ...
- Öhler A, ... (2010) ...
- Öhler A, ... (2010) ...

structures in the biotrickling filter for dimethyl sul de 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 bacteria for degradation of DMS are *Bacillus* sp., *Rhodobacteraceae bacterium*, *proteobacterium*, *delta proteobacterium*. The dominant bacteria, *Bacillus* sp., takes up 68.6%; while *Rhodobacteraceae bacterium*, *proteobacterium* and *delta Proteobacterium* 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 bio filter. *Bacillus* sp., sulfur-oxidizing bacteria, was able to degrade H<sub>2</sub>S [13,14]. *Proteobacterium* [15], oxidizing inorganic sul de and mercaptans, and *Rhodococcus*, deodorizing domestic animal feces [16] have been described as sul de oxidizers. Since DMS can be metabolized to dimethyl sulfoxide, methyl mercaptan, hydrogen sul de, 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 bio filter, dimethyl sul de is oxidized to carbon dioxide, sul de (SO<sub>4</sub><sup>2-</sup>). 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 for removal of dimethyl sul de 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<sup>-1</sup>, 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*,