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# Removal of dimethyl sulfide by the combination of non-thermal plasma and biological process ☆

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HIGHLIGHTS

- The system integrated with a NTP and a BTF unit achieved high efficiency to remove gaseous dimethyl sulfide.
- Bacterial communities in the BTF were assessed by PCR–DGGE.
- The addition of ozone from NTP made microbial community in BTF more complicated and active for DMS removal.
- Mechanism of DMS reaction in NTP–BTF was proposed.

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ABSTRACT

A bench scale system integrated with a non-thermal plasma (NTP) and a biotricking filtration (BTF) unit for the treatment of gases containing dimethyl sulfide (DMS) was investigated. DMS removal efficiency in the integrated system was up to 96%. Bacterial communities in the BTF were assessed by PCR–DGGE, which play the dominant role in the biological processes of metabolism, sulfur oxidation, sulfate-reducing and carbon oxidation. The addition of ozone from NTP made microbial community in BTF more complicated and active for DMS removal. The NTP oxidize DMS to simple compounds such as methanol and carbonyl sulfide; the intermediate organic products and DMS are further oxidized to sulfate, carbon dioxide, water vapors by biological degradation. These results show that NTP–BTF is achievable and open new possibilities for applying the integrated with NTP and BTF to odour gas treatment.



## 1. Introduction

Dimethyl sulfide (DMS) is an obnoxious odor pollutant owing to its offensive smells with a very low odor threshold value of 0.6–40 ppb and extreme negative hedonic characteristics, which is generated in the off-gas from paper mills, wastewater treatment, sewage sludge disposal, kraft process, organic synthesis, dyeing of acetate textiles, pharmaceuticals, insecticides and fungicides (Giri et al., 2010). Non-thermal plasma (NTP) is a promising technology for waste gas treatment (Durme et al., 2008). The non-thermal dielectric barrier discharge plasma reactor filled with ceramic raschig rings had the best $H_2S$ removal performance (Liang et al., 2011). Dimethyl sulfide was fully decomposed by two-electrode Ar micro-plasma, the $H_2$-, $CS_2$-, and $H_2S$-gaseous products were possibly recyclable and trapped (Chen et al., 2012). Breakdown voltage of DMS in Ar is lower than that of DMS in $N_2$, both of which are proportional to the gas pressures in a wire-cylinder pulse corona reactor. The conversion of DMS in Ar is more efficient than that in $N_2$ and air at a fixed peak voltage (Chen et al., 2009). When dimethyl sulfide and dimethylamine from pesticide factory were decomposed together in dielectric barrier discharges, synergistic actions exist in the processes, leading to higher conversion, higher energy yield and less byproducts formation (Chen et al., 2010). Increasing humidity inhibits the $O_3$ production in NTP and $MnO_x/Al_2O_3$ catalytic decomposition by the combination of NTP and its catalyst (Fan et al., 2010). The negative humidity effect on post-plasma catalytic toluene removal is mostly due to changing Van der Waals interactions (Durme et al., 2009).The efficient decomposition of $O_3$ on $MnO_2$ at ambient temperature leads to the formation of reactive oxygen species susceptible to react with residual pollutants in the effluent (Jarrige and Vervisch, 2009).

Biofiltration has been known as an efficient waste gas control technology for treatment odour at low cost of maintenance,

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and produces harmless by-products. Two *Hyphomicrobium* VS inoculation protocols were compared for start-up of a biotrickling filter (BTF) removing dimethyl sulfide (Sercu et al., 2005). Biological treatment of DMS was investigated in a bench-scale biofilter, packed with compost along with wood chips, and enriched with *Bacillus sphaericus* (Giri et al., 2010). Hydrogen sulfide, methanethiol, dimethyl sulfide and dimethyl disulfide was degradated by *Thiobacillus thioparus* DW44 isolated from peat biofilter (Cho et al., 1991). Dimethyl sulfide was conversed by *Methylophaga sulfidovoran* in a microbial mat (Zwart and Kuenen, 1997). The diversity of the bacterial community in the biofilter is dynamic and varies with inlet DMS loads, the addition of glucose, and fluctuating temperature (Chung et al., 2010).

Biological treatment of dimethyl sulphide was investigated in a bench-scale biofilter *B. sphaericus*. Evaluation of microbiological status of the biofilter indicated the presence of other bacterial cultures viz. *Paenibacillus polymyxa*, and *Bacillus megaterium*, besides *B. sphaericus* (Giri et al., 2010). Biofilter filled with a mature compost containing sewage sludge and yard waste as filter medium was designed to treat an average ammonia and volatile organic sulfur compounds (Hort et al., 2009). Methanol (MeOH) addition could enhance DMS treatment by up to 35% through a MeOH pulse feeding strategy (Zhang et al., 2007), but too much MeOH is detrimental (Kumar et al., 2010). A dominant DMS-oxidizing strain Au7 was isolated and identified as *Chemolithotrophic thiobacilli* (Wang et al., 2011). The biofilter containing the yard waste compost filter medium is the more efficient to remove a mixture of gaseous reduced sulfur compounds containing hydrogen sulphide, methyl mercaptan and dimethyl sulphide (Hort et al., 2013). Reduced sulphur compounds such as hydrogen sulphide, methyl mercaptan, dimethyl sulphide and dimethyl disulphide were removal simultaneously by a two-phase biotrickling filter in series – the first one inoculated with A. thiooxidans and the second one with *T. thioparus* (Ramírez et al., 2011). Hydrogen sulphide and methyl mercaptan have an inhibitory effect on the bio-oxidation of DMS and dimethyl disulphide. The inhibitory effect of $H_2S$ over the oxidation of DMS and dimethyl disulphide is greater than methyl mercaptan (Cáceres et al., 2012).

Non-thermal plasma may cause secondary pollution. Biofiltration, its disadvantages, include large volume of bioreactor and slow adaptation to fluctuating concentrations in waste gas. This requires combination of non-thermal plasma and biological process to meet the more strict standards.

The objective of this work is to study the dimethyl sulfide removal by integrating reactor consisting of a NTP and a BTF, to further improve removal efficiency, to eliminate secondary pollution from NTP and reduce the volume of bioreactor and run stability. The study analysis bacterial community composition in BTF assessed by polymerase chain reaction-denaturing gradient gel electrophoresis (PCR–DGGE), and evaluates residence time (RT) on the integrating reactor. The mechanistic for NTP-biological oxidation of odor were elicited, which is believed to promote the application of the NTP–BTF.

## 2. Methods

### 2.1. Experimental procedure

The schematic of the bench-scale integrated reactor includes a NTP and a BTF unit is presented in Fig. 1. The non-thermal plasma discharge is produced in NTP reactor consisting of two electrodes. The outer electrode is a multiaperture aluminum piece (140 mm length and 100 mm width) and the inner electrode is a copper rod (diameter = 10 mm). The outside wall of the reactor is wrapped over by a glass. The non-thermal plasmas were generated by applying high voltage power into the reactor. The biotrickling filter (internal diameter of 90 mm and 1200 mm long) was packed with ceramsite (external diameter of 8–15 mm) to a height of 510 mm. 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 dimethyl sulfide supplied from the gas cylinders, was first diluted with the compressed air, passed through an air mixture bottle, then flowed through the NTP reactor and flowed upwards the bottom of the BTF. DMS concentrations were monitored by the analysis device of Photo-Ionization Detector, and gas flow rate was monitored by the rotameter and the mass flow controllers. In the process of the biodegradation of DMS experiments, nutrient-containing aqueous solutions was sprayed downward at a rate of 0.94 $m^3\ m^{-2}\ h^{-1}$ with a peristaltic pump from the top of column to maintain the moisture of the BTF and supply nutrients to the microbial population.

### 2.2. Analytical methods

Bacterial community compositions in the biotricking filter of dimethyl sulfide were assessed by PCR–DGGE, and identify the colonies of the predominant microorganisms by the procedures of total DNA extraction, polymerase chain reaction (PCR) amplification of 16S rDNA, and sequencing and comparing results with those in the GenBank database by using the BLAST server of nucleotide sequence similarity in the NCBI website. MiniRAE PLUS PGM-7320 Photo-Ionization Detector analysis device was used for analysis of DMS concentration, which was made in USA (RAE systems Company, USA). Gas flow rates were measured using Model LZB-1 flow meters with units of 0.1 $m^3\ h^{-1}$. The pH values were measured by a

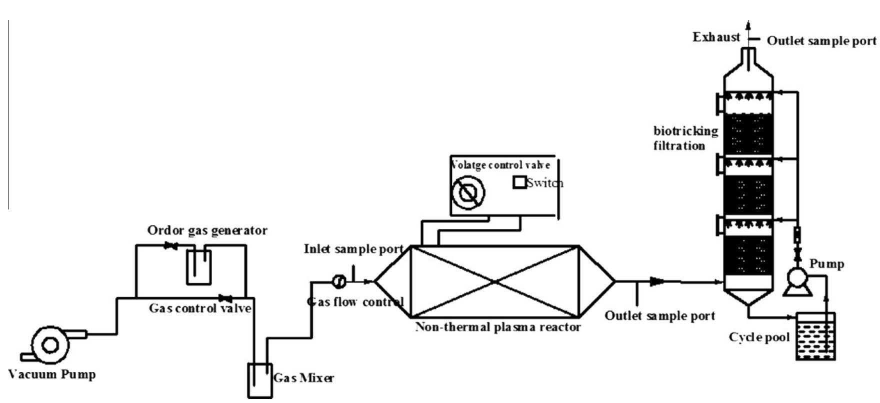


**Fig. 1.** Schematic diagram of an integrated reactor includes a non-thermal plasma (NTP) and a biotricking filtration (BTF) unit for treatment of waste gas containing DMS.