

Technical Note

# Co-treatment of hydrogen sulfide and methanol in a single-stage biotrickling filter under acidic conditions

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Received 12 October 2006; received in revised form 19 January 2007; accepted 22 January 2007

Available online 8 March 2007

## Abstract

Biofiltration of waste gases is cost-effective and environment-friendly compared to the conventional techniques for treating large flow rates of gas streams with low concentrations of pollutants. Pulp and paper industry off-gases usually contain reduced sulfur compounds, such as hydrogen sulfide and a wide range of volatile organic compounds (VOCs), e.g., methanol. It is desirable to eliminate both of these groups of compounds. Since the co-treatment of inorganic sulfur compounds and VOCs in biotrickling filters is a relatively unexplored area, the simultaneous biotreatment of H<sub>2</sub>S and methanol as the model VOC was investigated. The results showed that, after adaptation, the elimination capacity of methanol could reach around 236 g m<sup>-3</sup> h<sup>-1</sup> with the simultaneous complete removal (100%) of 12 ppm H<sub>2</sub>S when the empty bed residence time is 24 s. The pH of the system was around 2. Methanol removal was hardly affected by the presence of hydrogen sulfide, despite the low pH. Conversely, the presence of the VOC in the waste gas reduced the efficiency of H<sub>2</sub>S biodegradation. The maximal methanol removal decreased somewhat when increasing the gas flow rate. This is the first report on the degradation of methanol at such low pH in a biotrickling filter and on the co-treatment of H<sub>2</sub>S and VOCs under such conditions.

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*Keywords:* Acidophiles; pH; Fungi; Substrate competition; Oxygen limitation

## 1. Introduction

Biofiltration is an established waste gas treatment technology that may provide a cost-effective solution for many industries facing emission problems (Kennes and Veiga, 2001). A biofilter consists of a packed bed of organic or synthetic material on which a microbial film is attached. When a contaminated air stream passes through the reactor, the pollutants are transferred to the biofilm, where they are biodegraded to simple end products, such as water and carbon dioxide in the case of non-halogenated volatile organic compounds (VOCs). Instead of using large amounts of thermal energy to destroy pollutants, or removing pollutants by transfer from one phase to another, biofilter systems harness the natural ability of microorganisms to degrade

organic and inorganic contaminants biochemically into environmentally harmless end products at ambient temperature. Under adequate conditions, gas-phase bioreactors are more efficient than non-biological treatment processes for VOC removal (Kastner and Das, 2005). A biotrickling filter works in a similar manner to a biofilter, except that an aqueous phase is continuously trickled over the packing, and the filter bed is made of some synthetic or inert material, like plastic rings, open pore foam, or lava rock. The trickling solution contains essential inorganic nutrients such as nitrogen, phosphorous, and potassium, and is usually recycled.

To date, most lab-scale biofiltration studies address the removal of single pollutants under constant operating conditions. Such conditions are highly unusual at wastewater treatment plants and some other facilities. For instance, the malodorous emissions into the atmosphere from a pulp mill originate mainly from cooking, pulp washing, recovery boiler, evaporation, bark boiler, white liquor preparation,

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lime kiln, and pulp drying. These emissions tend to be a complex mixture of  $\text{H}_2\text{S}$  and other reduced sulfur compounds (such as dimethyl sulfide, dimethyl disulfide, and methyl mercaptan), as well as VOCs (such as methanol, terpenes, alcohols, phenol, ketones, and formaldehyde). The actual composition and individual concentrations often vary over time. In addition to being malodorous and toxic, some of these gases also undergo photochemical reactions in the atmosphere, contributing to the formation of photochemical oxidants, principally ozone.

Relatively little is known on the treatment of off-gases that contain both  $\text{H}_2\text{S}$  and VOCs. One problem encountered is that, as the sulfur in hydrogen sulfide is converted to sulfuric acid, the pH of the biofilm decreases. If the pH declines rapidly, biological activity in the biofilter may be inhibited, thereby reducing treatment effectiveness for organic compounds and sulfide, although  $\text{H}_2\text{S}$  is generally oxidized by *Thiobacillus* species that exhibit optimum activity at relatively acidic pH. However, most *Thiobacillus* species are autotrophic organisms and, therefore, many of them do not use VOCs as a carbon source for growth. Most known heterotrophic bacteria capable of consuming VOCs prefer neutral pH, and heterotrophic VOC-degraders are often more inhibited by acidification than  $\text{H}_2\text{S}$ -degraders. In addition, acid conditions may hasten the deterioration of organic support media. These apparently conflicting pH optima for microbial activity are a challenge for developing bioreactors for the simultaneous removal of both  $\text{H}_2\text{S}$  and VOCs.

One solution is the treatment in a two-stage process as proposed by Chitwood et al. (1999). In the first stage,  $\text{H}_2\text{S}$  is oxidized in a biotrickling filter in which the pH drops as a result of  $\text{H}_2\text{SO}_4$  accumulation. The  $\text{H}_2\text{S}$ -free off-gas is then passed through a neutral-pH biofilter for the removal of VOCs. Considerable savings could possibly be made if  $\text{H}_2\text{S}$  and VOC removal was combined in a single bioreactor. The same authors also investigated another solution for the treatment of mixed hydrogen sulfide and organic vapors in a rock packed pilot-scale biofilter (Chitwood and Devanny, 2001). The removal efficiencies of  $\text{H}_2\text{S}$  and VOC reached at pH 4 were comparable to the values expected for a neutral-pH biofilter. However, lava rock is susceptible to dissolution at low-pH conditions (around 4). Recently, Cox and Deshusses (2002) reported that  $\text{H}_2\text{S}$  and toluene can be effectively treated simultaneously in a single-stage biotrickling filter. The pH of operation (4.5 or 7.0) did not greatly affect the performance of  $\text{H}_2\text{S}$  and toluene removal, except that at pH 4.5, the startup phase of toluene degradation was relatively long. Recent research at publicly owned treatment works has shown that  $\text{H}_2\text{S}$  and low concentrations of VOCs can be co-treated in biofilters without pH control.

In our previous research, the autotrophic biodegradation of hydrogen sulfide as single pollutant was studied in a low-pH biotrickling filter (Jin et al., 2005a,b). However, hydrogen sulfide and VOCs such as methanol, formaldehyde, methyl ethyl ketone, are often found together in

waste air streams. Since the co-treatment of  $\text{H}_2\text{S}$  and VOCs in biotrickling filters is a relatively unexplored area, and autotrophic microorganisms do not use methanol as a carbon source for growth, the objective of this research was to investigate the feasibility of using a single-stage low-pH biotrickling filter to treat a mixture of methanol and  $\text{H}_2\text{S}$ . Methanol was added to an originally autotrophic low-pH  $\text{H}_2\text{S}$ -degrading reactor to determine to what extent the organic compound could be removed.

## 2. Material and methods

### 2.1. Experimental setup

The schematic of the biotrickling filter used in this study is shown in Fig. 1 and has been described previously in detail (Jin et al., 2005a). It is a cylindrical packed bed reactor made of glass, 75 mm in diameter and 700 mm in height, filled with polypropylene Pall rings of a nominal height of 15 mm. The total height of the packed bed was 640 mm. The Pall ring bed had an initial porosity of 91% and a specific surface area of  $350 \text{ m}^2 \text{ m}^{-3}$ . This inert synthetic packing material was used to exclude any carbon source other than methanol and prevent complications arising from bed compaction and aging over the experimental period. The glass column contained four equidistant sampling ports. All fittings, connections and tubings were made of Teflon.  $\text{H}_2\text{S}$  was introduced by passing the gas stream over a  $\text{H}_2\text{SO}_4$  solution into which a solution of  $\text{Na}_2\text{S}$  was dripped. Gas phase  $\text{H}_2\text{S}$  concentrations were regulated by changing the  $\text{Na}_2\text{S}$  concentration and/or dripping rate. Methanol was introduced into the gas by saturating a side air stream sparged into a bottle filled with pure methanol. The resulting waste gas was introduced through the bottom of the column for counter-current flow operation (Fig. 1). The gas flow rate was maintained constant at  $71 \text{ min}^{-1}$ , corresponding to an empty bed residence time (EBRT) of 24 s. The aqueous mineral medium was continuously recirculated over the packed bed using a peristaltic pump (model 323E/D, Watson-Marlow Limited, Fal-

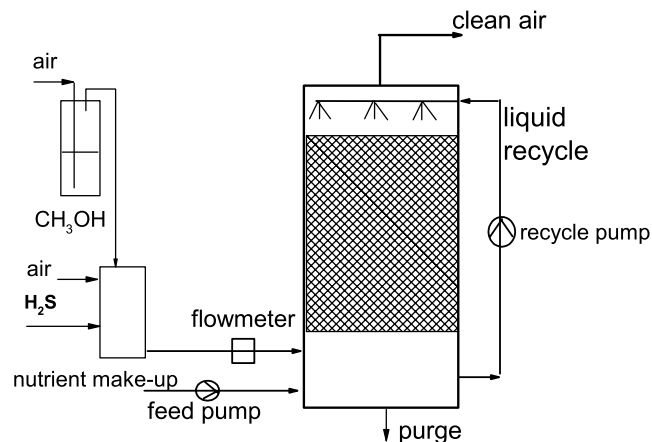


Fig. 1. Schematic of the laboratory scale biotrickling filter.

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