One-stage biotrickling filter for the simultaneous removal of hydrogen sulphide, methanol and α -pinene

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A one – stage biotrickling filter was evaluated for its potential to treat a gas – phase pollutant mixture comprising hydrogen sulfide, methanol and α – pinene, both under steady – and transient – state conditions. Experiments were carried out at different gas – flow rates, corresponding to empty bed residence times (EBRTs) of 38 and 26 s, and at different inlet concentrations of the individual pollutants. The results obtained from this study, in terms of the elimination capacity (EC), were then compared to a two – stage bioreactor (BTF + BF) that was designed to remove these pollutants. Under the tested experimental conditions, the maximum EC values, in the one – stage BTF, were 191, 307 and 123 g m $^{-3}$ h $^{-1}$, for H₂S, methanol and α -pinene, respectively.

Keywords: Two – stage and one-stage bioreactors, biotrickling filter, biofilter, hydrogen sulphide, VOC, shock loadings.

1. Introduction

The acclivitious rise of malodorous emissions from the pulp and paper manufacturing industry has led to the development of newer techniques for its control amongst practising environmental researchers. Hydrogen sulphide (H_2S), methanol and α – pinene are representative inorganic, as well as organic, hydrophilic and hydrophobic pollutants present in emissions from pulp and paper industry. Biological treatment systems such as biotrickling filters (BTF) and biofilters (BF) have shown promising results for handling waste gases at relatively low concentrations and at moderately high flow rates (Kennes and Veiga, 2001). A typical bioprocess depends on the activity of microorganisms to decontaminate polluted air, through a series of complex comprising absorption, adsorption, diffusion phenomenological steps biodegradation. Bioprocesses can be used for a wide range of odours and volatile pollutants, including both organic and inorganic compounds, either hydrophobic or hydrophilic (Kennes et al., 2009).

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2. Materials and methods

2.1 Inoculum

2.1.1 Two – stage bioreactor

The first – stage BTF was inoculated with a mixture of an autotrophic H_2S – degrading – culture and an acid – tolerant methanol degrading yeast (*Candida boidinii*) that was obtained from our previous work on the co – treatment of H_2S and methanol in a low – pH BTF. *Ophiostoma stenoceras*, a well known sap – wood colonizing fungus, isolated from a biofilter efficiently degrading α – pinene was used to inoculate the 2^{nd} stage BF (Rene et al., 2010).

2.1.2 One – stage bioreactor

The one – stage BTF was inoculated with a mixture of the above mentioned consortium, together with a *Rhodococcus* strain, that possess the ability to efficiently degrade gas – phase α – pinene.

2.2 Media composition

The composition of the mineral salt medium used in the BTF, for both experimental studies (two – stage bioreactor and one – stage bioreactor), was (in g L $^{-1}$ of de – ionized water); KH₂PO₄: 2; K₂HPO₄: 2; NH₄Cl: 0.4; MgCl₂·6H₂O: 0.2; FeSO₄·7H₂O: 0.01. The medium used in the fungal BF had the following composition (g L $^{-1}$); K₂HPO₄: 0.5, MgSO₄·7H₂O: 0.1, KH₂PO₄: 4.5, NH₄Cl: 2, and 2 mL trace elements and vitamin solutions. The final pH of this medium was 5.9.

2.3 Experimental

During the first phase of the experimental work, a BTF and a BF arranged in series was used as the two – stage system (Fig 1). The first stage is a 2.78 L BTF, filled with polypropylene pall rings, while the second stage BF had a working volume of approximately 4.88 L and was packed with irregular grains of perlite mixed with 50 % (weight) of the same polypropylene pall rings as used in the former BTF. In this experimental study, one – stage system, a BTF filled with pall rings and having an operational volume of 4.55 L was used. The bioreactors were provided with equidistantly placed gas sampling ports and two filter material sampling ports uniformly distributed on the other side of the column. All fittings, connections and tubings were made of either glass or teflon.

2.4 Operational procedure

A compressed air stream was split into three flows. H_2S was generated by passing the major portion of the air stream over a H_2SO_4 solution into which a solution of Na_2S was dripped. Different gas phase H_2S concentrations were obtained by changing the Na_2S concentration and/or dripping rate. The other two minor air streams were bubbled through liquid methanol and α – pinene, separately in flasks. The three streams were combined in a mixing chamber, and fed to the bottom of the BTF column in a counter – current flow mode. The aqueous mineral medium described above was continuously recirculated over the packed bed using a peristaltic pump (323E/D, Watson – Marlow Ltd, Falmouth Cornwall, England) at a constant volumetric flow rate of 2.77 L h $^{-1}$.

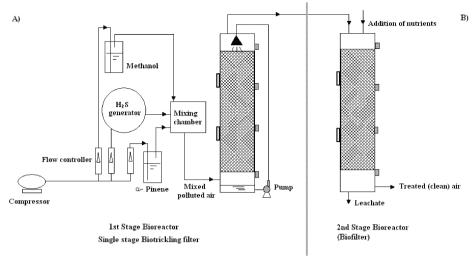


Fig. 1. Schematic of A) the first – stage BTF, (same configuration as the one – stage), and second stage – BF.

The exit air from the top of the BTF was later fed to the fungal BF in a downflow mode. For experiments with the one – stage BTF, the same operational procedure was followed, and the exit air from the BTF was vented out.

2.5 Analytical methods

 H_2S concentration was determined using a hand held sensor (Dräger Sensor XSEC H2S HC6809180). Inlet and outlet gas – phase concentrations of methanol and α – pinene were measured via gas chromatographic analysis using a Hewlett – Packard 6890 series II GC, equipped with a flame ionization detector (FID). The GC was equipped with a 50 m TRACER column (TR – WAX, ID: 0.32 mm, film thickness: 1.2 μm) and helium was used as the carrier gas (flow rate: 2.0 mL min – 1). The temperatures at the GC injection, oven and detection ports were 250, 120 and 250 °C respectively. CO_2 concentrations were measured using another Hewlett – Packard 5890 GC fitted with a thermal conductivity detector (TCD). The CO_2 concentrations were determined at an injection temperature of 90 °C, an oven temperature of 25 °C and using a TCD at 100 °C. The pall rings and perlite packing material were taken from the BTF and BF, respectively, and subjected to SEM analysis (Fig 2). Examinations were performed with a JOEL JSM – 6400 SEM working at a voltage of 20 kV and a working distance of 15 mm, and with Oxford Instruments EDX equipment.

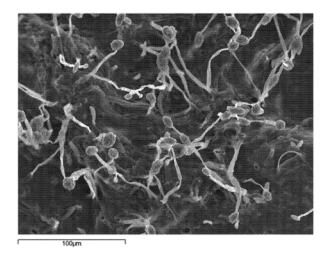


Fig. 2. SEM photograph for the one – stage biotrickling filter.

3. Results and discussion

3.1 Two - stage bioreactor

3.1.1 Reactor performance

After start – up, experiments were carried out in five phases, at flow rates of 0.12, 0.24, 0.36, 0.5 and 1 m 3 h $^{-1}$ corresponding to different empty bed residence times (EBRTs) and inlet loading rates of H₂S, methanol and α – pinene vapours. H₂S and methanol were removed in the first stage (BTF) with maximum ECs of 45 g m $^{-3}$ h $^{-1}$ for H₂S and 894 g m $^{-3}$ h $^{-1}$ for methanol, while a high EC of α – pinene was observed in the second stage BF (138 g m $^{-3}$ h $^{-1}$). It was further delineated that, low loading rates of methanol (100 g CH₃OH m $^{-3}$ h $^{-1}$) and moderate loading rates of H₂S would be ideally preferred in the first – stage BTF, in order to achieve high removal efficiencies without mutual inhibition of both the pollutants. The high EC achieved in the second – stage BF, 138 g m $^{-3}$ h $^{-1}$, for a hydrophobic VOC, α – pinene, was plausible due to the inoculation of that reactor with the fungus *Ophiostoma stenoceras* and considering similar previous results with that fungus grown on α – pinene, (Jin et al., 2006). The maximum EC (EC_{max}) achieved at different EBRT, for each pollutant in either the BTF or BF, was highly dependent on the ILR.

3.1.2 Effect of transient state operations

Shock loading studies were carried out in two steps, viz., as long term (66 h – low to medium) and short term (12 h – low to high) shock loads, with the target pollutants. Increasing the loading rate from 12 to 35 g m $^{-3}$ h $^{-1}$ and from 70 to 230 g m $^{-3}$ h $^{-1}$ for H_2S and methanol in the BTF reduced their removal efficiencies by 40% and 25% respectively. However, α – pinene removal in the BF dropped by 10%, when a medium shock load of 55 g m $^{-3}$ h $^{-1}$ was applied.

On the other hand, during high shock loads, the performance of the first stage BTF and second stage BF decreased significantly. At a shock load of 100 g m⁻³ h⁻¹, H_2S removal decreased by almost 72% in the BTF, while methanol removal also declined significantly (28%) at a shock load of 1150 g m⁻³ h⁻¹. Both medium and high shock loads did not have any significant effect on α – pinene removal in the second stage BF.

3.2 One - stage BTF

3.2.1 BTF performance

Experiments were carried out at different inlet concentrations of H_2S , methanol and α – pinene, and at two different flow rates, corresponding to EBRTs of 38 and 26 s, respectively. The highest ECs were 191 g m $^{-3}$ h $^{-1}$ for H_2S , 307 g m $^{-3}$ h $^{-1}$ for methanol and 123 g m $^{-3}$ h $^{-1}$ for α – pinene, corresponding to inlet loading rates of 131, 313 and 123 g m $^{-3}$ h $^{-1}$, respectively (Fig. 3).

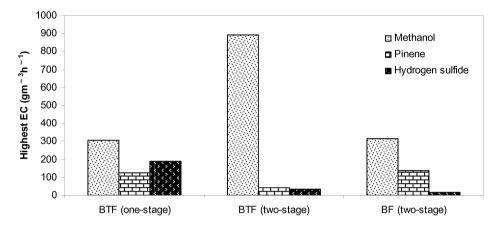


Fig. 3. Hightest elimination capacities reached for the three compounds in the one – stage and two – stage systems

3.2.2 Effect of transient state operation

Experiments were carried out at a constant EBRT of 26 s, and by imparting a sudden shock load to the BTF, by varying the inlet concentrations from low to high values. These shock loads were applied for 5 h, to test the ability of the BTF to withstand transient operating conditions, similar to those expected in chemical processing plant operation. The response of the BTF was monitored after the shock load to envisage the recovering phase of the removal profiles. The inlet loading rate was increased from 52 to 196, 24.8 to 260, and 42.2 to 257 g m⁻³ h⁻¹, for H₂S, methanol and α – pinene, respectively, during the shock load. It was observed that, methanol and α – pinene removal efficiencies dropped by 13 and 14%, respectively, while the removal of H₂S remained unaffected, despite being subjected to very high loading rates.

4. Conclusions

The results from this study provide sufficient information on the antagonistic and synergistic effects occurring during the biological treatment of a complex gaseous

mixture containing organic, inorganic, hydrophilic and hydrophobic pollutants. The results show that high ECs are achievable in both the one – stage and two – stage bioreactor configurations, under a wide range of steady and non – steady state operating conditions, which are common to effluents from pulp and paper industry. According to the results, in terms of removal efficiency, VOCs were better removed in the two-stage system, while H₂S biodegradation proved to be significantly higher in the one-stage reactor. Both reactor configurations, two – stage system and one – stage system, showed good ability to withstand sudden shock loads when subjected to transient – state operations.

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