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Liquid phase optimisation in a horizontal flow biofilm reactor (HFBR) technology for the removal of methane at low temperatures

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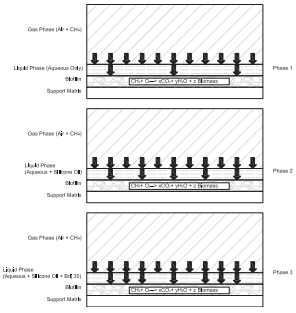
HIGHLIGHTS

- Methods for the optimisation of a horizontal flow biofilm reactor for the removal of methane gas are investigated.
- Ammonium salts appeared to have a positive influence on the performance of HFBR 3.
- Addition of silicone oil to the liquid phase led to significant performance improvements.
- Addition of a non ionic surfactant led to further significant improvements.
- CO_2 analysis revealed good correlation between CO_2 production and CH_4 oxidation.

GRAPHICAL ABSTRACT



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ABSTRACT

Methane (CH_4) is a potent greenhouse gas often emitted in low concentrations from waste sector activities. Biological oxidation techniques have the potential to offer effective methods for the remediation of such emissions. In this paper, methods of improving the CH_4 oxidation performance of a horizontal flow biofilm reactor (HFBR) technology, operated at low temperatures, are investigated.

Three pilot scale HFBRs were operated over three studies (Study 1, 2 & 3) lasting 310 days in total. The reactors were loaded with 13.2 g $CH_4/m^3/h$ during each study and operated at an average temperature of 10 °C.

In Study 1, nutrients were added to the biofilm via a liquid nutrient feed (LNF) comprising water and nutrient mineral salts. Average removals were 4.2, 3.1 and 2.3 g $CH_4/m^3/h$ for HFBRs 1, 2 and 3 respectively.

In Study 2 silicone oil was added to the LNF of all three HFBRs. Average removals increased, when compared to Study 1, by 31%, 79% and 78% for HFBRs 1, 2 and 3 respectively.

In Study 3 a non ionic surfactant (Brij 35) was added to the LNF and silicone oil liquid phase of HFBRs 1 and 2. The operating conditions of HFBR 3 were not changed and it was used as a control. A concentration of 1.0 g Brij 35/L proved most effective in improving reactor performance; with removal rates increasing by 105% and 171% for HFBRs 1 and 2 respectively when compared to Study 1.

These results indicate the potential of liquid phase optimisation for improving mass transfer rates and removal performances in biological reactors treating CH_4.

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1. Introduction

Methane is a prominent greenhouse gas with a global warming potential 25 times that of carbon dioxide (CO_2) and comprises almost a quarter of worldwide greenhouse emissions [16,33]. 55% of anthropogenic methane (CH_4) emissions are below the lower explosive limit (LEL) for CH_4 and cannot be thermally oxidised [4]. In such cases, biological waste treatment technologies can be an effective mitigation measure against these emissions [10,20]. Biofilm reactors are a practical alternative for the control and mitigation of these emissions [20,38,18]. Biofilm reactors are low cost, energy efficient and are simple to construct and operate [38]. Biofilm reactors treating CH_4 have been previously shown to be capable of achieving high removals of up to 100 g $CH_4/m^3/h$ [33,27] and have successfully been deployed at site scale [23].

There are, however, a number of challenges when designing CH_4 biofilm reactor, foremost of which is the low solubility of CH_4 in water. This presents a barrier to mass transfer and necessitates long hydraulic retention times, especially at low temperatures [33]. Biofilm reactors treating CH_4 have typically required retention times 100 times greater than biofilm reactors treating odorous compounds such as hydrogen sulphide or ammonia [36], with required empty bed retention times (EBRT) of over 1 h previously reported [14,23,13].

Recent studies have shown, however, that the limiting effect of low solubility can be alleviated in a number of different ways. Optimising the nutrients in the liquid phase to maximise methanotrophic activity in the biofilm can significantly improve performance [8,25]. The use of a secondary organic liquid phase with a higher affinity for methane than water such as polydimethylsiloxane (silicon oil) or hexadecane have been shown to result in greater rates of mass transfer in both a packed bed biotrickling filter and in a stirred tank reactor and lead to improved oxidation rates [15,7,24]. Addition of silicone oil leads to improved methane solubility as the partition coefficient of methane in silicone oil is approximately 10 times lower than in water; thus at equilibrium, the ratio of concentrations of methane dissolved the oil and water phases will be 10:1 [33].

In other studies, non-ionic surfactants such as Brij 35 and Tween 20 have been used to improve reactor performance [4]. Non ionic surfactant molecules contain both hydrophilic and hydrophobic elements and when added to the aqueous phase of a biofilm reactor, can increase the solubility of low water soluble compounds such as methane [4,19]. Non ionic surfactants have successfully improved performances of packed bed biofilters [4,21] and are largely biodegradable and non toxic in low concentrations (<0.5% w/w), [5]. Brij 35 can also be used as an oil water emulsifier (its hydrophobic–lipophilic balance (HLB) number is 16.9 – within the range for solubilising oils into water).

To date limited work has focused on the combined use of transfer vectors such as silicone oil and non-ionic surfactants (e.g. Brij 35) to aid mass transfer of CH_4 into the liquid phase. Rocha Rios and Revah [30] found that the effectiveness of silicone oil as a transfer vector is dependent on the degree of oil dispersion in the liquid phase. While a number of studies use mechanical turbulence to create dispersion [3,33,28] previous studies have not examined the possibility of combining transfer vectors to both enhance mass transfer and improve oil dispersion in the water phase.

Furthermore, most studies are carried out at temperatures of 20 °C or more. In many scenarios (due to the facility in question or the climate) temperatures can be significantly lower.

The horizontal flow biofilm reactor (HFBR) has been previously applied successfully to both wastewater and waste gas treatment [18,9]. The unique flow regime in the HFBR ensures good contact with the biofilm in the reactor and alleviates problems that can be associated with conventional biofilm reactors such as clogging, channelling, compaction and pressure drop. In this study, the effect of adding silicone oil, both with and without Brij 35, to the liquid phase of HFBRs treating methane gas, was investigated.

2. Materials and methods

2.1. Horizontal flow biofilm reactor (HFBR)

Three HFBR units (HFBR 1, HFBR 2 and HFBR 3) were commissioned during these trials. Each HFBR comprised 55 horizontal plastic sheets with integrated frustums stacked vertically – one above the other. The sheet stack was placed in a sealed enclosure that could be opened for visual assessment and biofilm sampling. The working volume of each reactor was 18 L and the top plan surface area (TPSA) of the plastic media was 0.04 m^2, giving a total media plan area of 2.4 m^2. 6 intermediate sample ports were located along the vertical profile of each reactor allowing for intermediate samples of air and water to be taken.

The HFBR units were housed in a temperature-controlled laboratory, maintained at 10 °C. The influent gas mixture stream comprised a mixture of atmospheric air with a CH_4 gas supply. Mass flow controllers (Bronkhorst High Tech BV, Ruurlo, Netherlands), flowmeters (Key Instruments, Trevose, USA) and pressure regulators (GCE DruVa, Cheshire, United Kingdom) were used to control gas flow rates and gas mix proportions as required (Fig. 1).

The gas mixture, containing approximately 1.6% v/v CH_4, was introduced at the top of the reactor (Sheet 1) and flowed horizontally across each sheet before moving to the sheet below. Similarly a liquid phase, introduced onto Sheets 1 and 30 of the reactor, flowed over each sheet before dropping to the sheet below (i.e. the unit does not operate as a submerged reactor). The gas and liquid exited the reactor below Sheet 55 (the bottom sheet in the reactor). Operating parameters are summarised in Table 1.

Nutrients were added to each of the reactors in the form of a Liquid Nutrient Feed (LNF) mixture, similar to that used by Nikiema et al. [25] (Table 2). The LNF was delivered intermittently (10 min/h) via a peristaltic pump. The LNF was delivered in a step feed manner, i.e. 75% of the LNF (3 L/day) was dosed onto Sheet 1 and 25% of the LNF (1 L/day) onto Sheet 30.

2.2. Biofilm growth and inoculation

An enrichment strategy was employed to cultivate a methanotroph-rich biomass capable of methane oxidation which could be used to seed the HFBRs. A biomass mix comprising landfill cover soil, landfill leachate, composted organic fraction municipal solid waste (OFMSW) and compost leachate in a 1:1:1:1 ratio was used for the enrichment culture. Briefly, 2 ml of the biomass mix were placed in each of several 40-ml crimp-top, glass vials with 8 ml Adapted Whittenbury Medium (AWM; [40]. The vials were sealed and the headspace was adjusted to a methane concentration of 10% at atmospheric pressure. Vials were incubated in the dark at 10 °C shaking at 80 rpm. The headspace methane concentration was monitored by gas chromatography (GC; Varian CP-3800 Gas Chromatograph) analysis of twice-weekly headspace samples. Once the headspace methane concentration was <0.5%, the headspace was flushed with air and a 10% methane headspace was re-instated. Over the course of 4 months, the cultures were subcultured (c. 10% inoculum) to fresh medium and eventually were scaled to 2-l enrichment cultures to cultivate sufficient biomass for HFBR inoculation. The enriched culture was added to the HFBRs at the beginning of the trial and then re-circulated around the systems for several days to encourage biofilm formation.