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A capillary bioreactor to increase methane transfer and oxidation through Taylor flow formation and transfer vector addition

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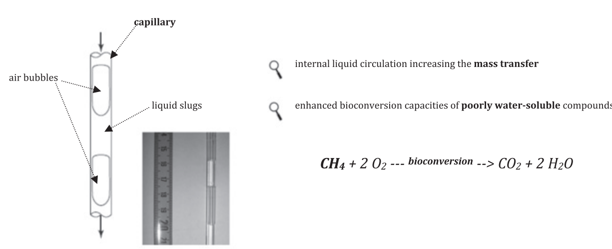
HIGHLIGHTS

- ▶ We studied methane oxidation in a capillary gas treatment bioreactor.
- ▶ A new bioreactor in which Taylor flow with transfer vector addition are combined.
- ▶ Superior mass transfer (k_La) is obtained when compared to conventional bio-contactors.
- ▶ Improved methane removal obtained when compared to conventional bio-contactors.
- ▶ A method demonstrated to improve bio-treatment of gaseous hydrophobic compounds.

GRAPHICAL ABSTRACT



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ABSTRACT

The impact of two strategies to enhance the mass transfer of hydrophobic compounds, Taylor flow (or segmented flow) and the addition of an organic transfer vector (silicone oil), were investigated under abiotic and biotic conditions in a capillary bioreactor. The capillary bioreactor consisted of a capillary column (where Taylor flow was produced in a gas/liquid flow) and a gas–liquid separator at the outlet of the capillary column which was operated as a stirred tank with superficial aeration. It was shown that the system was limited by mass transfer and not by the biological reaction. Taylor flow in the capillary resulted in an increase of up to two orders of magnitude for the volumetric oxygen transfer coefficient (k_La) when compared to the coefficient for the gas–liquid separator, or values previously obtained in other turbulent contactors. The bioconversion rates of methane in the capillary column where found to be significantly higher than for conventional systems. Silicone oil addition increased k_La up to 38% in the gas–liquid separator, but reduced it with 38% in the capillary. Contrary to observations during abiotic k_La determinations, silicone oil addition increased the CH_4 removal and O_2 consumption by the methanotrophic consortium in both, gas–liquid separator and capillary. Increased gas flow rate gave an 19% increase in methane removal in the capillary bioreactor, an additional increase of 8% was obtained adding 5% of silicone oil at the same flow, while an additional increase of 47% was obtained adding 10% of silicone oil at the same flow with inoculum pre-adapted to transfer vector. The contribution of the capillary channel to the overall methane removal in the system was high considering that the volume of this channel was just 0.64% of the total volume in the bioreactor, indicating a good potential of further optimization of the reactor system.

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1. Introduction

Biodegradation of poorly water-soluble air pollutants, such as methane, is in applied biological gas treatment systems generally limited by the mass transfer from the gas to the aqueous phase where the microorganisms are present [1]. A strategy to increase the mass transfer of hydrophobic compounds in air treatment applications is the addition of an organic phase (solid or liquid) with more affinity for the target compound than water [2]. The systems amended with the organic phase are called in generic way two-phase partition bioreactors (TPPBs) and the organic phase is known as *transfer vector*. TPPBs have shown to improve toluene [3], hexane [4], and methane removals [5]; the oxygen transfer rate [6–8], and the performance under transient conditions [9,10].

In turbulent systems (stirred tank and airlift reactors) the transfer vectors can increase the gas/water interfacial contact area (a) by disruption of the gas bubbles as shown by Galindo et al. [11] and Quijano et al. [12]. Also, the driving force for mass transfer is increased by the higher solubility of the hydrophobic compounds in the organic phase (up to 10 times more for methane and oxygen in silicone oil than in water) as shown by Rocha-Rios et al. [5]. Finally, it is possible that the microorganisms extract the pollutants *directly* from the vector, without intermediate transfer to water as suggested by McLeod and Daugulis [13] and Rocha-Rios et al. [5,8]. These three mechanisms explain the observed higher degradation velocities in stirred tanks added with a vector transfer.

Higher degradation rates alone, however, are not sufficient. The effectiveness of the transfer vector, in economical rather than process-dynamical terms, depends on the cost of the vector, which is usually expensive [14]. Moreover, dispersing the vector in water requires energy [1]. Both the increased power consumption in the system [8,15] and the organic phase cost severely impede the application of these systems on a commercial scale. For highly soluble compounds, dispersion and mass transfer are less important and in that case classical laminar bioreactors (biofilters, biotrickling filters and bioscrubbers) have adequate performance.

Laminar contactors such as biofilters, biotrickling filters and bioscrubbers, with commercially attractive low power consumption, typically have removal efficiencies, for soluble compounds, above 90% [16]. In contrast, when these systems are used for the removal of poorly water-soluble compounds, the removal efficiency can be as low as 40% even at residence times of several minutes [5]. It is questionable if the addition of a vector helps to improve mass transfer in these systems. Without the energy input to break up a transfer vector as silicone oil into small droplets, one obtains in general a non-homogeneous poor dispersion. The benefit of adding a vector in such systems is therefore inconclusive [17].

Another novel strategy to increase gas–liquid mass transfer is the monolithic reactor. Monolith reactors are increasingly significant as multiphase reactors, considering the advantages that they offer in comparison to conventionally used bed and slurry systems for a host of processes. These advantages, which include low pressure drop, high gas–liquid mass transfer rates, and minimum axial dispersion (plug flow), stem from the uniquely structured multichannel configuration of monoliths [18,19]. In essence, a monolith block is composed of an array of uniformly structured parallel channels, often of square or circular geometry, typically having hydraulic diameters between 1 and 5 mm. Thus, the monolith can be viewed as a structure that is comprised of many repeating building blocks, where the basic building block is a single channel. It can be argued that data obtained from studies on a single channel, or what may be called a capillary, can be used in scaling up a monolith reactor, provided that a uniform gas and liquid distribution (such as that obtained) occurs in the monolith block [20,21].

Among several possible flow patterns in capillaries, segmented flow (Taylor flow), gives the best mass transfer properties [22]. In this flow pattern, the flow through the capillary channel consists of liquid slugs well separated from each other by distinct gas bubbles. This flow recirculates within the liquid slugs and increases the mass transfer from the gas to the liquid [23]. Taylor flow in capillaries makes it possible to obtain mass transfer coefficients equivalent to stirred tanks, but with one order of magnitude lower power consumptions or in other words, k_La one order of magnitude higher than stirred tank reactors for the same power consumption [21].

Monolith packages have recently gained importance in biotechnological applications as relatively high mass transfer rates can be obtained at relatively low energy input [1,21]. Ebrahimi et al. [24] and Ebrahimi et al. [25] studied the possible clogging of the channels by biomass growth, while Jin et al. [26] used a monolith bioreactor to treating air polluted with volatile organic compounds (VOCs). Monoliths are often operated at higher superficial velocities than trickle beds, and the residence time may be too short to achieve full conversion in a single pass [27].

In this work the effects of Taylor flow and the addition of a transfer vector on methane biodegradation in a capillary bioreactor were studied compared to a control system without these tested variables. This is the first report of methane oxidation in a capillary bioreactor, and moreover it is the report of a bioreactor with both Taylor flow and the transfer vector addition. The aim of this work was to investigate the feasibility of two-phase partition capillary bioreactors to extent the application field of biological air treatment.

2. Materials and methods

2.1. Microorganisms and culture conditions

A methanotrophic community (with *Methylobacterium organophilum* as the predominant strain) was enriched from an activated sludge sample at UAM-Iztapalapa wastewater treatment plant (México City). Culture maintenance, inoculum preparation and mineral salt medium composition were carried out as previously described by Rocha-Rios et al. [5].

2.2. Chemicals

Natural gas with an average methane concentration of 93% was diluted with air to obtain an average methane concentration of 4.2% (v/v) or 32 g m^{-3}. Silicone oil (polydimethylsiloxane) with 100 cSt of kinematic viscosity (S100) was purchased from Sigma–Aldrich. Silicone oils are not biodegraded or toxic for this methanotrophic community as shown by Rocha-Rios et al. [28].

2.3. Experimental set-up

The diagram of the system used for methane oxidation experiments is shown in Fig. 1. The capillary bioreactor consisted of an acrylic tube (polymethylmetacrylate) of 1 m of length and 0.003 m of inner diameter. A gas–liquid contactor similar to that described by Simmons et al. [29] was used to minimize the pressure shock during the contact in the entrance of capillary. The biphasic flow in the capillary was co-current downward. The gas was recirculated in a closed loop using an oil-free diaphragm pump (Wisa, Germany). The liquid was recirculated using two peristaltic pumps (Cole–Parmer, USA) connected in parallel to reduce the pulsations. The gas flow through the capillary was controlled with a rotameter and a needle valve. A jacketed flask with magnetic agi-