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Biological elimination of dilute methane through multi-channel Taylor flow capillary bioreactors

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ABSTRACT

Elimination of dilute methane (<5 % v/v) was investigated in several multi-channel capillary bioreactor configurations and evaluated for operating conditions and parameters relevant to long-term reliable performance. Although all reactors showed a high methane removal capacity, the addition of only surfactant or only silicone oil did not show enhancement in methane removal. The capillary bioreactor containing both silicone oil (up to 20 % v/v, 20 cSt) and surfactant (BRIJ 58) treated methane with very high elimination capacities of >200 g per m^3 internal capillary channel per hour at gas contact times around 30 s, which is one order of magnitude lower than gas contact times of conventional biological gas treatment methods. No accumulation of biomass on the walls of capillary channels was observed during the 300 days of operation. Internal gas recirculation was applied to decouple gas-liquid turbulence conditions from the actual gas retention time. This work revealed that a capillary bioreactor can be a useful platform for the abatement of dilute methane emissions.

1. Introduction

The global economic output, as measured in gross domestic product, is estimated to double between 2020 and 2040, with greenhouse gas emissions rising by ~30 % [1]. However, even emissions at the current level are already leading to unquestionable environmental changes and global warming.

The need for mitigating methane (CH_4) emission is increasing dramatically as research indicates that CH_4 has greater climatic impact than previously thought [2]. As global CO_2 mitigation falters, it now appears that aggressive CH_4 mitigation is a lower cost means to reduce climate change while the mitigation cost strongly varies among emission sources [3,4]. Methane is responsible for ~30 % of the rise in global temperatures since the Industrial Revolution. Although emissions of CH_4 are much smaller than those of CO_2 by mass, CH_4 is about 28 times more potent than CO_2 per unit mass when averaged over the most common 100-year time scale. Over a 20-year time scale, which is relevant to the near-term threat of climate change, methane is about 80 times more potent. The voluntary Global Methane Pledge, launched at COP 26 in November 2021, is supported by about 160 countries and aims at reducing CH_4 emissions from human activity. Some countries have also released national methane action plans and many countries are in the process of doing so. Despite these initiatives, CH_4 emissions remain unchanged, even though reducing them is imperative for controlling near-term global warming as well as improving air quality [5].

About 60 % of global CH_4 emissions are caused by human activities according to the 2021 assessment by the Climate and Clean Air Coalition and the United Nations Environment Programme [6]. The major anthropogenic sources of CH_4 are primarily oil and natural gas systems (35 %), coal mines (12 %), waste treatment systems, mainly landfills and wastewater (20 %), agriculture, mainly manure and enteric fermentation (32 %) and rice paddies (8 %). Methane emissions from natural gas and oil systems are the result of system leaks, inefficiencies, and process upsets, while CH_4 emissions from enteric fermentation refers to its formation (methanogenesis) in the guts of ruminant livestock (cattle, goats, sheep). Anaerobic digestion for the conversion of organic waste to energy has significantly expanded over the last decades, but unfortunately results in liquid effluents containing a substantial amount of dissolved CH_4 [7]. The release of CH_4 from the liquid effluent and fugitive emissions undermines the sustainability of biogas as renewable energy source.

The CH_4 contained in ventilation gases is often too lean for self-sustaining combustion. Indeed, >55 % of anthropogenic CH_4 emissions have a concentration below the lower explosive limit of CH_4 in air

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mixtures of 5 % v/v and incompatible for energy recovery or for chemical oxidation processes devoted to the removal of CH_4. Most technologies are therefore not economically viable when the CH_4 concentration is below 5 % v/v, which equals 50,000 ppm_v or about 31,000 mg m^{-3} at ambient temperatures and pressures [8]. Examples of dilute CH_4 emissions are those from landfills (0–20 % v/v), from ventilated coal mines (0.1–1 % v/v), from liquid manure storage tanks (0–3 % v/v), or animal houses (0–0.015 % v/v) [9–11].

Biotechnologies are increasingly applied for gas treatment, but methane's poor solubility in water, together with its high volatility and chemical stability, hampers their application for its abatement due to its limited bioavailability to the microbial community. Studies of biological systems treating dilute methane all required long gas contact times of several minutes exemplifying that the methane bioavailability hampers biological methane elimination processes. Alternative biological approaches are in urgent need to especially increase the mass transfer of CH_4 from the gas phase to the biomass. The most studied and most applied biological gas treatment reactors (biofilters and biotrickling filters) operate generally under laminar flow conditions. Laminar flow occurs when a gas or liquid flows in parallel layers, with minimal disruption between the layers, being characterized by high diffusion and low advection. Therefore, improved advection (e.g., through mixing) will improve contaminant mass transfer through a water film. Mixing is typically applied in liquid reactors to enhance reactions that are mass transfer limited, but requires high energy inputs, which is in general a critical parameter for the design and application of process equipment.

In this context, capillary reactors can combine good mass transfer with relatively low pressure drop, two important factors affecting cost effectiveness for many industrial applications. Capillary gas-liquid contactors are structures of parallel straight microchannels (small round or square capillary channels) separated by a thin wall. The hydrodynamics of gas-liquid flow in capillary channels have been extensively studied within the context of chemical reaction engineering [12–14]. Examples of study areas and applications are discussed in Haase et al. [15] and Kreutzer et al. [12]. Despite that capillary gas-liquid bioreactors have shown to be an effective gas treatment platform [16,17], they have not been systematically studied for dilute methane abatement.

The removal of dilute gaseous CH_4 was herein investigated using different capillary bioreactor configurations, optimizing channel diameter, channel length, internal gas recirculation, and circulating liquid characteristics. The results were evaluated under relevant operating conditions (i.e., gas contact time, gas-liquid ratio, gas-liquid slug face velocity) considering both energy input requirements and long-term reliability (i.e., inlet CH_4 transient conditions and biomass control).

2. Materials and methods

2.1. Capillary bioreactor set-up

Biotic experiments were conducted in multi-channel capillary bioreactors to investigate how variations in capillary channels, operational modes, liquid properties, and conditions relevant to long-term reliable performance influence methane biodegradation. Three long-term biotic experiments were undertaken to assess the dilute methane removal in a CBR according to Table 1.

- Test Series A involved a CBR containing capillary channels with an internal diameter of 1.7 mm and 1.5 m in length. The primary object was to quantify the effect of the addition of an oil to the recirculating liquid (3 % v/v silicone oil with a viscosity of 20 cSt).
- Test Series B involved a CBR containing capillary channels with an internal diameter of 2.4 mm and 1.0 m in length. This test was focused on quantifying the effect of the addition of a surfactant to the recirculating liquid (SDBS at a concentration up to 27.5 mg L^{-1}).

Table 1
Operational conditions of the capillary bioreactors to study dilute methane removal.

Test Series	Days (#)	CBR channels			Slug face velocities (m s^{-1})	ECRT (s)	Internal gas recirculation (Yes/No)	G/L ratio in channel (−)	Methane conc. (ppm_v)	Methane IL[c] (g m^{-3} h^{-1})	Liquid medium of CBR[d]
		Diameter[a]	Length	No.							
A	98	1.7 mm (PTFE[b])	1.5 m	25	1.7–6.0	0.5–4.0	No	0.5–1.9	~250	125–1250	Stage I: Medium only Stage II: 3 % v/v Silicone oil (20 cSt)
B	238	2.4 mm (glass)	1.0 m	25	0.15–0.84	4.5–9.0	No	0.4–2.5	350–7850	100–2000	Stage I: Medium only Stage II: SDBS (27.5 mg L^{-1})
C	305	2.4 mm (glass)	1.0 m	25	1.3–2.5	4.1–33.9	Yes	0.5–2.0	900–6500	300–1100	Stage I: Medium only Stage II: BRIJ 58 (80 mg L^{-1}) Stage III: BRIJ 58 (80 mg L^{-1}) 5 % v/v Silicone oil (20 cSt) Stage IV: BRIJ 58 (160 mg L^{-1}) +5 % v/v Silicone oil (20 cSt) Stage V: BRIJ 58 (160 mg L^{-1}) +20 % v/v Silicone oil (20 cSt)

[a] Internal diameter.
[b] PTFE = polytetrafluoroethylene (Teflon™).
[c] IL: Inlet methane load per total internal volume of all capillary channels.
[d] Concentration of the liquid phase in the CBR.

- Test Series C involved a CBR containing capillary channels with an internal diameter of 2.4 mm and 1.0 m in length. The aim was to quantify the effect of the combined addition of a surfactant and oil to the recirculating liquid (Brij 58 at concentrations of 80 and 160 mg L^{-1}, and silicone oil at concentrations of 5 and 20 % v/v).

The schematic representation of the CBR set-up in the three studies is shown in Fig. 1. In Test Series A and Test Series B the gas-liquid mixing was conducted by pushing air into the liquid through a flat 3 mm thick PDMS membrane through which about 400 needle holes were perforated using a 0.4 mm diameter needle. In Test Series C the gas-liquid mixing was conducted by injecting the air via a 4 mm supply tubing into the liquid that contained 6 mm scrubber Kaldness K1 packing rings.

Internal gas recirculation at different ratio's was only applied in Test Series C using an EVO 10 compressor (EAD, Model H5P3 P 1, Spain), where the recycled gas stream was subsequently mixed with fresh inlet air containing CH_4 before resupplied into the bottom liquid reservoir as illustrated in Fig. 1. Internal gas recirculation was previously shown to be beneficial for CH_4 removal in a biotrickling filter study [18] and was here adapted as a strategy to decouple optimum turbulent conditions inside the channels from the gas contact time to potentially enhance CH_4 removal efficiency in the CBR. During the first 175 days of Test Series C different operating conditions were applied, while the same operating conditions were maintained during the testing of the liquid additives (the last 130 days of Test Series C): an up-flow segmented flow face velocity inside the capillary channels of 2.2 m s^{-1} and an internal gas recirculation (recycled gas to fresh inlet air) ratio of 25, which resulted in an empty channel gas residence time of 23 s. The he surfactant BRIJ 58 used in Test Series C was selected based on results of the experimental work reported elsewhere [19].

The fresh inlet air was clean dry air from which all the CO_2 and humidity was removed before CH_4 was introduced using a flow control meter (Aalborg, Model GFC 17). The clean dry supply airflow and the recirculating gas flow were measured with a rotameter (Aalborg, S/N 51588-2). The temperature of the recirculation liquid of the CBR was maintained at 24 ± 1 °C. CH_4 concentrations were measured at the inlet and outlet of the CBRs typically twice a day and where each measurement is the average of three analysed gas samples. Liquid samples were withdrawn once per week for the analyses of the biomass concentration, the total organic concentration (TOC), the total nitrogen concentration (TN), the pH and the Electrical Conductivity.

No recirculating liquid was replaced during Test Series A and Test Series B, while during Test Series C 800 mL of recirculating liquid was removed from the CBR five days per week from day 87 onwards and replaced with fresh medium to avoid nutrient limitation and accumulation of inhibitory metabolites. During this medium replacement, the biomass and silicone oil were recovered and returned to the capillary reactor through centrifugation of the liquid twice (5000 rpm for 10 min) in a refrigerated centrifuge (Eppendorf, Model 5439 R).

The performance of the CBR was evaluated mainly by the following four operating parameters:

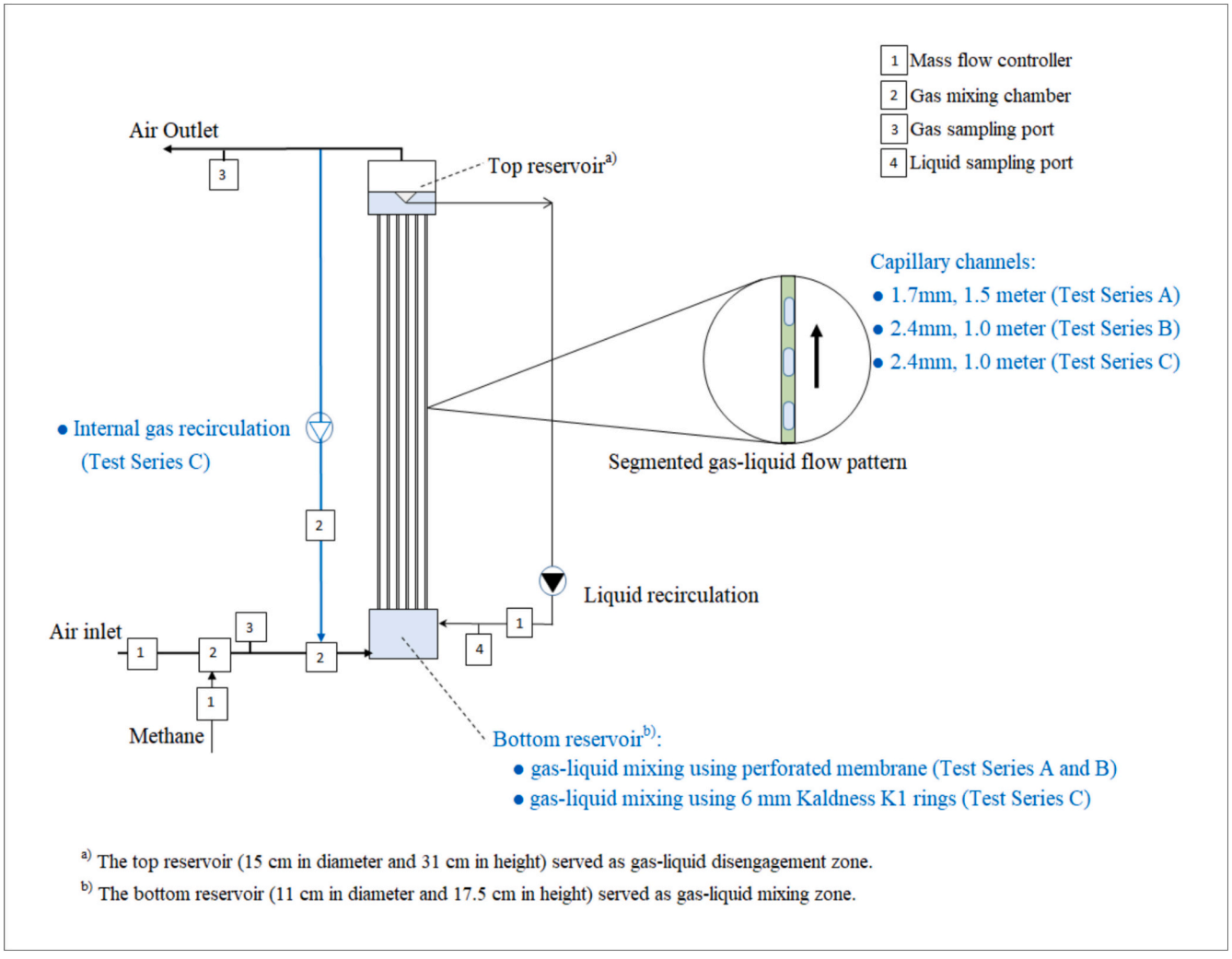

Fig. 1. Set-up of the capillary bioreactor to optimize dilute methane removal.

1. The inlet methane load (*IL*), which is defined as follows (Eq. (1)):

$$IL\ (g\,m^{-3}\,h^{-1}) = Q_g \times C_i/(V_c \times n_c) \quad (1)$$

where Q_g is the inlet air flow rate ($m^3\ h^{-1}$), C_i is the inlet CH_4 concentration ($g\ m^{-3}$), V_c is the internal volume of a capillary channel (m^3), and n_c the number of capillary channels (−).

2. The CH_4 elimination capacity (*EC*), which is defined as follows (Eq. (2)):

$$EC\ (g\,m^{-3}\,h^{-1}) = (C_i - C_o) \times Q_g/(V_c \times n_c) \quad (2)$$

with Co standing for the outlet CH_4 concentration ($g\ m^{-3}$).

3. The removal efficiency (*RE*), which is defined as follows (Eq. (3)):

$$RE\ (\%) = (C_i - C_o)/C_i \times 100 \quad (3)$$

4. The empty channel residence time (*ECRT*) is defined as follows (Eq. (4)):

$$ECRT\ (s) = (V_c \times n_c)/(Q_g) \quad (4)$$

2.2. Microbial inoculum, chemicals and analytical methods

The analytical methods applied as well as the medium composition and the chemicals used were as described by Kraakman et al. [19]. The capillary reactor was inoculated with fresh activated sludge from Valladolid wastewater treatment plant (Spain) in Test Series A and Test Series B. In Test Series C a mixed inoculum was used from two sources: fresh activated sludge from Valladolid water resource recovery facility (Spain) and post-composted anaerobically digested sludge from Five Ford wastewater sludge treatment facility (United Kingdom). The microbial consortium in Test Series C was acclimatised in the CBR during the first 175 days before starting the study on day 175 to determine the effect of the surfactant and oil addition to the recirculating liquid in the capillary reactor on the methane removal capacity.

2.3. Evaluation of factors important for long-term reliable operation

Microbial responses to transient conditions to gain deeper understanding on the microbial reactor system in order to define its reliability, which is the combination of robustness and resilience. Process robustness reflects the capacity of a system to maintain functionality with changes such fluctuations in inlet loading rate, inlet loading interruptions, or operational upsets, while resilience is the rate at which a system returns to its original state after being disturbed. Biological systems may be impacted by sudden changes or longer-term changes in parameters such as nutrient concentration or accumulated biomass.

The following reliability related parameters of the CBR were studied during Test Series C to get an understanding of its stability and its long-term operation:

- Methane transient conditions (inlet load shocks for about 5 h and a 6-day inlet load interruption)
- Nutrient concentration (minimum total nitrogen concentration)
- Increased surfactant concentration (beyond the threshold of potentially causing microbial inhibition)
- Biomass control (risk of biomass accumulation inside capillary channels)

In addition, operating conditions relevant to key input requirements (i.e., energy) have been evaluated.

3. Results and discussion

3.1. CBR performance evaluation Test Series A – addition of silicone oil

During Test Series A, the inlet concentration (Ci) was maintained relatively low, averaging 250 ± 15 ppm_v during the entire 98-day study. No fresh medium was added during this period, only demineralised water was occasionally supplemented to compensate for evaporation losses and to maintain the reactor's liquid working volume at approximately 8.5 L. The pH of the recirculated liquid remained stable at ~7.4 ± 0.05 throughout the study.

The results of the CBR for the exact same operational conditions (0.6 s ECRT, a slug face velocity of 4.6 m s^{-1}, and a gas-to-liquid ratio between 1.0 and 1.1), the CH_4-RE during Stage I (Medium only) ranged between 16 and 25 %, while the RE during Stage II (with 3 % v/v silicone oil) ranged between 4 and 13 %. Fig. 2a shows the results for the periods only with similar operating conditions to illustrate the impact of the additives. Even when the ECRT was increased from 0.6 to 2.6 s and then to 5.1 s in Stage II, the RE increased only slightly to 10 ± 2 % and 16 ± 5 %, respectively. Therefore, it can be concluded that the addition of silicone only as second liquid phase to a CBR treating dilute CH_4 does not support any improved removal efficiency under the conditions tested and may hamper its performance.

For optimal mass transfer, the preferred flow pattern in capillary channels is segmented flow (also called Taylor flow), which is a bubble train of alternating liquid slugs and air bubbles with gas and liquid flowing co-currently. Although this flow regime seems to be laminar, the internal liquid circulation increases the mixing of the liquid phase. The mass transfer between the gas and liquid phases is boosted by the internal recirculation within the liquid slug, while mass transfer also benefits from the relatively large gas-liquid interfacial area and small diffusion paths. After silicone oil addition to the CBR, the CH_4 removal efficiency was less, which may be explained by the higher overall liquid viscosity. A high liquid viscosity can hamper the gas-liquid mass transfer in a capillary channel under segmented (Taylor) flow regime as discussed elsewhere [20]. Moreover, the higher viscosity increases the viscous drag forces relative to the surface tension forces, which may also compromise capillarity. The Capillary number (Ca) represents this relation between viscous drag forces and capillary forces (Eq. (5)):

$$Ca\ (-) = \mu \times u/\gamma \quad (5)$$

where μ is the viscosity (Pa s), u the liquid velocity (m s^{-1}), and γ the surface tension of the liquid in the gas phase (N m^{-1}). Increased viscous drag forces slow down the internal liquid recirculation in the liquid slug, the vortex that enhances mass transfer through advection rather than diffusion. Thulasidas et al. [21] found that the liquid internal recirculation velocity reduces sharply and ultimately becomes zero with increasing the Ca number, with Ca > 0.6 being the theoretical value where the internal vortex becomes zero in a downward flow. In our study, the calculated Ca numbers with the 20 cSt viscosity silicone oil were 0.06 and 0.19 for the 0 % v/v and 3 % v/v oil, respectively. This assumes that the overall liquid viscosity is proportional to the oil-liquid fraction, and the gas-liquid surface tension is 35.7 mN m^{-1}. This assumption is a simplification but shows that adding silicone oil may increase the Capillary number beyond the threshold where internal recirculation is reduced, and mass transfer is compromised.

Despite the low REs recorded during Test Series A (only 8 % during Stage II and 21 % during Stage I as summarized in Table S-2 in Supplementary material), the overall average EC was nevertheless 188 ± 45 g m^{-3} h^{-1} and 46 ± 29 g m^{-3} h^{-1} during Stage I and Stage II, respectively (Fig. 2a). It can be concluded that under the conditions tested that the addition of the silicone oil does not improve the CH_4 gas-liquid mass transfer.

After 100 days, Study A was stopped because 3 of the 25 capillary channels (1.7 mm internal diameter) became non-functional. This

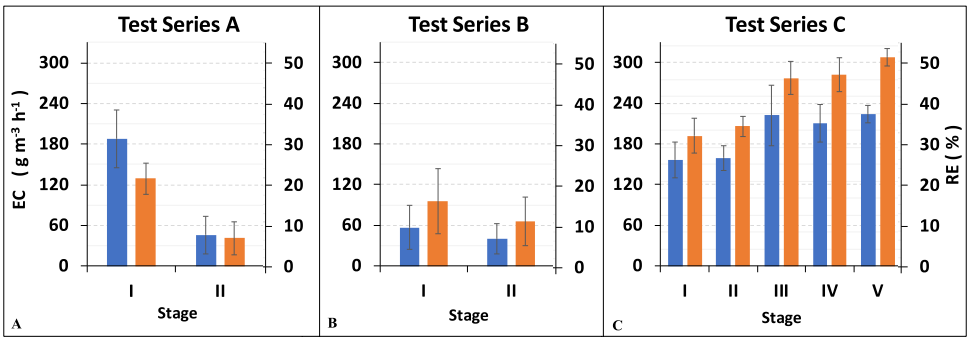

Fig. 2. Methane elimination capacity (blue) and methane removal efficiency (red) in the different capillary bioreactor configurations during Test Series A (effect of silicone oil), Test Series B (effect of surfactant), and Test Series C (effect of silicone oil + surfactant). Data is shown for the periods for each Test Series where the operating conditions were similar to illustrate the impact of the additives only.

failure was attributed to biomass aggregates detaching from the recirculation liquid tubing and/or pump, which subsequently obstructed the inlet side of these capillary channels preventing liquid flow.

3.2. CBR performance evaluation Test Series B – addition of surfactant

During Test Series B, the addition of a surfactant was investigated according to Table 1. The inlet concentration averaged 2277 ± 1043 ppm_v but was also changed up to a maximum concentration of 3818 ± 246 ppm_v. The surfactant tested in this Test Series B was sodium dodecylbenzenesulfonate (SDBS) at a concentration of 27 mg L^{-1}. A concentration lower than 30 mg L^{-1} has shown to avoid microbial inhibition while increasing the RE in a conventional biotrickling filter treating CH_4 and various other hydrophobic short-chain alkanes [22]. Occasionally, fresh mineral salt medium was added to compensate for water evaporation and for nutrient replenishment (on average 154 mL per week, which represented ~2 % per week of the total liquid volume in the reactor). The pH and the electric conductivity were on average 8.0 ± 0.4 and 179 ± 22 uS cm^{-1}, respectively, while TSS and VSS were also measured in Test Series B, which averaged 1.78 ± 0.88 and 1.33 ± 0.67 g L^{-1}, respectively.

The results of the CBR for the exact same operational conditions (9.0 s ECRT, a slug face velocity of 0.18 m s^{-1}, and a gas-to-liquid ratio of 1.5) showed a CH_4 RE during Stage I (Medium only) of 16 ± 8 %, while the RE during Stage II (with surfactant SDSB) was slightly less and on average 11 ± 6 %. The ECs during these periods with similar operational conditions accounted for 57 ± 33 g m^{-3} h^{-1} (Stage I) and 40 ± 22 g m^{-3} h^{-1} (Stage II), as illustrated in Fig. 2b. It can be concluded that the addition of the surfactant SDSB to a CBR treating dilute CH_4 does not provide any improved RE under the conditions tested.

Despite the low REs during Test Series B, the average ECs during Stage I in Test Series B were nevertheless in similar range as the average EC in Test Series A, averaging 186 ± 166, and slightly higher during Stage II than the average EC in Test Series A, averaging 72 ± 52 g m^{-3} h^{-1}. The additional supplementation of 13 mg SDSB L^{-1} by day 226 did not exert a positive nor a negative effect on the CH_4 REs. During the entire 238-day duration of Test Series B all of the 25 capillary channels stayed functional, which indicate that 2.4 mm diameter channels (Test Series B) are more appropriate than 1.7 mm diameter channels (Test Series A).

3.3. CBR performance evaluation – Test Series C – addition of silicone oil with surfactant

During Test Series C, the CBR with the 2.4 mm diameter and 1.0 m long channels was tested under medium slug velocities ranging from 1.3 to 2.5 m s^{-1}, while internal gas recirculation was applied to decouple the gas-liquid turbulence conditions inside the capillary channel from the actual gas retention time. BRIJ 58 was selected as the surfactant to be tested in the CBR because of its potential to enhance CH_4 gas-liquid mass transfer in the presence of silicone oil as well as its ability to enhance the oil-in-water Emulsion Capacity and oil-in-water Emulsion Stability at a concentration low enough to eliminate the risk of microbial inhibition. BRIJ 58 have also shown to enhance the cell hydrophobicity of CH_4 oxidizing bacteria and can improve overall the bioavailability of dilute CH_4 as shown elsewhere [19].

During the first 175 days of Test Series C, and prior the testing of surfactant and silicone oil as additives in the CBR, the microbiology was exposed to CH_4 as the sole energy and carbon source, in which optimal and stable process conditions of the CBR were established. During this period, the operating conditions were changed in terms of gas contact time, slug velocity, internal gas recirculation rate and G/L ratio. The CH_4 RE was during this initial phase in general lower than 15 % during operation at a short ECRT of 2.7 s and was 22 ± 6 % at a higher ECRT of 13.6 and 23.6 s. The modification of the G/L ratio did not significantly change the performance under the conditions tested (see Table S-3 in Supplementary material). The slug velocity was changed at two different ECRTs (2.7 and 13.6 s) by changing the internal gas recirculation rate, which allowed decoupling the slug velocity from the overall gas retention time in the channels. Interestingly, the optimum slug velocity at both ECRTs ranged from 2.0 to 2.5 m s^{-1} as illustrated in Fig. 3. It was concluded that the optimum slug velocity for the CBR set up in Test Series C was ~2.2 m s^{-1} and this operating condition was applied as such during the testing of liquid additives in the CBR.

During the testing of the liquid additives in the CBR the same operating conditions were maintained: an up-flow segmented flow face velocity inside the capillary channels of 2.2 m s^{-1} and an internal gas recirculation (recycled gas to fresh inlet air) ratio of 25, which resulted in an ECRT of 22.6 s. The inlet CH_4 concentration was maintained at ~4500 ppm_v. Just prior the start of testing surfactant and silicone oil as additives in the CBR, the biomass concentration was measured multiple days and showed a TSS of 1.8 ± 0.3 g L^{-1} (82 ± 11 % VSS). The pH and the electrical conductivity of the recirculating medium at the start were 7.3 ± 0.04 and 510 ± 10 uS cm^{-1}, respectively, and remained relatively constant during the entire test period of 130 days (7.4 ± 0.1 and 470 ± 20 uS cm^{-1}). The TN concentration was maintained between 40 and 90 mg N L^{-1} for the whole experiment and was on average 62 ± 15 mg N L^{-1}. At the beginning of Stage I, the TN concentration was ~90 mg N L^{-1}, slowly decreasing over time despite medium replenishment, reaching a concentration of 40 mg N L^{-1} by day 60 after the start of Stage I. Thus, 50 mg N L^{-1} as sodium nitrate was added to restore the initial nitrogen concentration of 90 mg N L^{-1}, steadily decreasing again to ~40 mg N L^{-1} by the end of Stage V. The TOC was measured once a week and was on average 189 ± 40 mg C L^{-1}.

The influence of the addition of surfactant and silicone oil on CH_4 removal is illustrated in Fig. 2c. During Stage I, when no additives were added, the operational conditions of the CBR resulted in a RE of 32 ± 4 %, corresponding to an EC of 156 ± 26 g m^{-3} h^{-1}. The addition of the

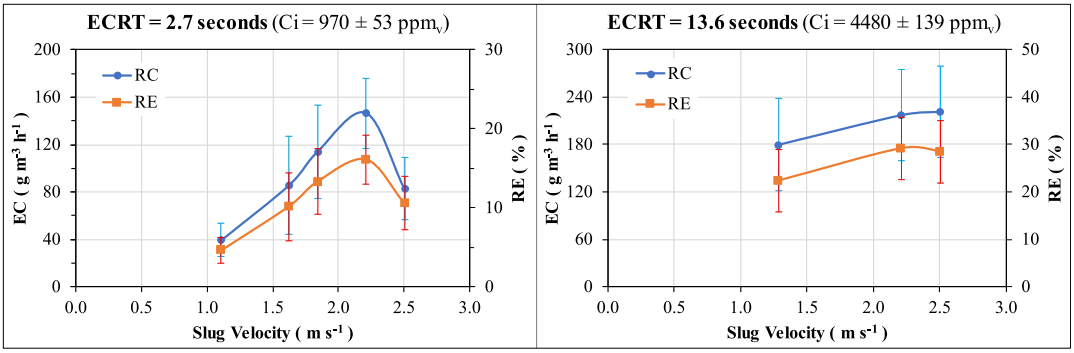

Fig. 3. Methane removal efficiency (RE) and methane elimination capacity (EC) at 2.7 s of gas contact time (left) and 13.6 s of gas contact time (right) during the initial 130 days of Test Series C.

surfactant in Stage II did not result in any significant change in the RE and the EC, remaining at 34 ± 3 % and 159 ± 18 g m^{-3} h^{-1}, respectively. In contrast, when silicone oil at 5 % was added in Stage III, both the RE and the EC increased by ~40 % up to 46 ± 4 % and 222 ± 45 g m^{-3} h^{-1}, respectively. The surfactant supported a superior gas-liquid mass transfer in a capillary channel, but only when combined with silicone oil. The surfactant enhanced emulsification of the oil in the medium, which appears to be the main mechanism rather than altering the gas-liquid mass transfer partial coefficient of CH_4.

No significant enhancement on the CH_4 removal performance was observed after the increased surfactant addition in Stage IV, with average RE and EC values in this stage of 48 ± 4 % and 214 ± 27 g m^{-3} h^{-1}, respectively. However, increasing the silicone oil to 20 % (v/v) in Stage V did further increase, though slightly, the RE and the EC to 53 ± 6 % and 231 ± 30 g m^{-3} h^{-1}, respectively (Fig. 2c). This confirmed that the addition of silicone oil beyond 5 % v/v is beneficial in this case, especially since during this Stage V the inlet concentration was somewhat lower compared to the average concentration in earlier stages in Test Series C (4195 ± 195 ppm_v during Stage V vs 4404 ± 250 ppm_v on average during Stages I to IV).

The performance of the CBR was also assessed under constant inlet CH_4 load and variations in the inlet gas flow rate (see Fig. S-1 in Supplementary material), leading to different inlet concentrations and gas contact times. The inlet gas flow rate was adjusted during the day, and CH_4 inlet and outlet concentrations were measured three times, 1 h after each adjustment. The measurements were repeated the next day under the same conditions. A low inlet gas flow rate of 0.2 L min^{-1} (=33.9 s of ECRT) resulted in a RE of ~60 % (62 ± 1.5 % during Stage IV and 59 ± 1.3 % during Stage V), while at a gas flow rate of 0.9 L min^{-1} (=7.5 s of ECRT) the RE decreased to ~25 % (24 ± 0.7 % during Stage IV and 28 ± 0.3 % during Stage V). The performance in both stages was similar regardless of the fraction of silicone oil applied. At a low inlet gas flow rate (high ECRT), 5 % v/v silicone oil (Stage IV) resulted in a slightly higher RE. Conversely, at a high inlet gas flow rate (low ECRT), 20 % v/v silicone oil (Stage V) showed slightly better performance.

The removal capacities reported herein are high compared to conventional biological gas treatment system treating dilute CH_4 emissions, especially when considering the extremely short gas contact time and relatively low inlet concentrations. Other studies with biological systems treating dilute CH_4 all required long gas contact times of several minutes and indicate that the bioavailability of the CH_4 hampers these bioprocesses. Studies performed within the last 15 years with biological systems treating dilute CH_4 were reviewed and summarized in Table 2. All these study set-ups required several minutes of gas contact time to obtain elimination capacities ranging from 9 g m^{-3} h^{-1} (6 min gas contact time) to 65 g m^{-3} h^{-1} (20 min gas contact time). Only the stirred tank bioreactor was capable of obtaining ~100 g m^{-3} h^{-1} but required 4.8 min gas contact time and high energy input, while a previous study with a capillary bioreactor was able to obtain relatively high elimination capacities (up to 77 g m^{-3} h^{-1}) but operated at lower superficial gas-liquid velocities and without liquid additives.

Table 2
Overview of biological system studies performed within the last 15 years on the abatement of dilute CH_4 (<5 % v/v = 50,000 ppm_v = ~31,000 mg m^{-3}).

System design	Inlet concentration (mg m^{-3})	EBRT (min)	Maximum elimination (g m^{-3} h^{-1})	References
BF	800–6000	3.2–17.5	60	[35]
	500–6300	4.1	~50	[36]
	160–2800	4.1	~15	[49]
	1250–3100	4.0–6.5	~10	[37]
	31,000	20.0	~ 65	[38]
	6200	1.6–19.5	~27	[11]
	11,700–22,200	4.4	11	[39]
	600–8000	6.0	~45	[40]
	40	0.25	~3	[41]
	1050–22,500	7.4–42.8	~13	[42]
	5580	70	~2	[43]
	430–1370	6.0	9	[44]
	4800	4.2	45	[45]
	13,600	20	37	[24]
BC	27,000–57,000	24–186	~9	[46]
BTF	15,300	4.0	30	[18]
	14,300	4	60	[24]
	11,100	4.8	51	[47]
STR	15,900	4.8	106	[47]
HFBR	9900	45–55	5	[23]
CBR	25,000	<1	77	[48]

BF = Biofilter, BC = Bio-cover, BTF = Biotrickling filter, STR = Stirred Tank Reactor, HFBR = Horizontal flow bioreactor, CBR = Capillary Bioreactor.

3.4. Test Series C – evaluation of factors important for stable bioreactor operation

3.4.1. Methane shock load

Transient conditions of a large increase in inlet concentration were investigated during Stage I and Stage V. Increasing the CH_4 inlet concentration instantly showed a similar increase in the CH_4 EC during both Stages (see Fig. S-2 in Supplementary material). This increase in EC confirmed that the operation of the bioreactor was mass transfer limited rather than kinetically limited during both Stage I and Stage V, with the CH_4 mass transfer from the gas phase to the liquid phase limiting the process performance. Nevertheless, the initial response in CH_4 removal during Stage V (with silicone oil) during the transient condition appears to be slightly quicker than during Stage I (without the oil). This may be an indication of the beneficial buffering capabilities of silicone oil in the CBR.

3.4.2. Methane supply interruption

The interruption of CH_4 supply was investigated when the CH_4 inlet

flow was stopped (day 291) and resumed again after six days (day 297), while keeping the rest of the CBR operational unchanged. No disruption of the CH_4 removal was observed when measured 30 min after the restart of the CH_4 supply (Fig. 4a). The CH_4 removal before and directly after the six-day interruption was similar: RE was 52 ± 2 % before vs 51 ± 1 % after, and EC was 219 ± 9 g m^{-3} h^{-1} before vs 227 ± 4 g m^{-3} h^{-1} after. It can be concluded that the silicone oil present in the culture broth indeed provided CH_4 buffering capacity, which was further confirmed by the CO_2 produced during the following days after resuming CH_4 supply. CO_2 production increased from 76 ± 7 % of the amount of CH_4 removed recovered as CO_2 to 122 ± 11 % after supply resumption. Indeed, more CH_4 was converted by the methanotrophic bacteria in the CBR than CH_4 was removed from the air stream by the CBR during the following days after resuming CH_4 supply. Although studies [23,24] have shown that the RE of CH_4 in a biological reactor can benefit from the addition of silicone oil, no studies have demonstrated the beneficial buffering capabilities of silicone oil during CH_4 supply interruption to overcome a starvation period.

3.4.3. Minimum nitrogen concentration

The nitrogen concentration in the experiment was maintained between 40 and 90 mg N L^{-1}. This nitrogen concentration was not limiting the microbial activity as the above experiment proved that the bioreactor was operating under mass transfer limiting conditions both at the beginning during Stage I (when TN concentration was ~80 mg N L^{-1}) and at the end during Stage V (when TN concentration was ~40 mg N L^{-1}). This nitrogen concentration range is much lower than the minimum required concentration in more laminar type of bioreactors such as biofilters and biotrickling filter, where the biomass is mostly growing as a fixed film on a carrier material rather than suspended in the liquid as is in our CBR. Estrada et al. [18] and Veillette et al. [25] showed that a TN concentration of 100 mg N L^{-1} should be maintained in a biotrickling filter and a biofilter treating CH_4, respectively.

3.4.4. Elevated surfactant concentrations

Surfactants are known to inactivate enzymes on the bacterial outer membrane or to cause cell membrane disruption, thus inhibiting biological conversions [26,27]. An increasing surfactant concentration was tested at the end of Test Series C to determine how a sudden increase of surfactant concentration would affect CH_4 removal in the CBR. Additional BRIJ 58 surfactant was added three days in a row with increasing concentration. During this period, the CH_4 RE dropped from 51 % to 44, 38 and 37 % after the addition of 80, 160 and 320 mg surfactant L^{-1}, respectively (Fig. 4b), while foam formation was observed after the second addition (day 306), which is indicative of major microbial cell lysis.

3.5. Biomass control

No accumulation of biomass on the walls of the 2.4 mm capillary glass channels was observed during the entire periods of CBR operation (238-days operation during Test Series B and >300-days operation during Test Series C). These observations are consistent with the observations in other long-term CBR studies where no biofilm attachment was observed inside capillaries [16,17]. Biofilm formation would start with the adhesion of bacteria cells on a surface, which may be influenced by factors including the characteristics of the surface, the bacterial cell wall, and the liquid flowing along the surface [28,29]. The surface characteristics may involve material surface roughness and surface hydrophobicity, the bacterial cell wall characteristics may involve cell hydrophobicity and filamentous appendages such as pili and fimbriae, while the liquid characteristic may involve the fluid hydrodynamic forces. Multiple studies have shown that hydrodynamic forces, particularly the shear stress of the liquid on a surface, is the key parameter factor on biofilm formation in terms of the initial adhesion, the biofilm firmness, as well as the composition of the bacterial community [30–32].

The shear stress in capillary channels can be estimated assuming that the liquid slugs behave as a fully developed laminar flow in a cylindrical tube, which is characterized by the following velocity distribution (Eq. (6)):

$$u_z(r) = 2\,U_s \times \left(1 - (r/R)^2\right) \tag{6}$$

where U_s is the superficial liquid slug velocity (m s^{-1}), and R the radius of the capillary tube. The liquid surrounding the gas bubbles follows the hydrodynamic of a falling liquid film [21]. The gas bubbles are surrounded by a liquid film with a thickness (δ) of R - R_b, R_b being the radius of the bubble, where the velocity distribution in a cylindrical falling film of thickness δ is (Eq. (7)):

$$u_z(r) = -(g \times \rho)\Big/4\,\mu \times \left(R^2 - r^2\right) - (g \times \rho)\Big/2\,\mu \times (R - \delta)^2\, ln\,(r/R) \tag{7}$$

where g is the gravitational constant, μ the liquid viscosity (N s^2 m^{-2} or Pa s), and ρ the liquid density (kg m^{-3}). The shear stress at the walls of the capillary channel will be equal to (Eq. (8)):

$$\tau_{rz} = -\mu \times (du_z/dr)_{r=R} \tag{8}$$

This expression, for the liquid slugs leads to Eq. (9):

$$\tau_{rz} = \mu \times 4 \times (U_s/R) \tag{9}$$

and for the regions around the bubbles (Eq. (10)):

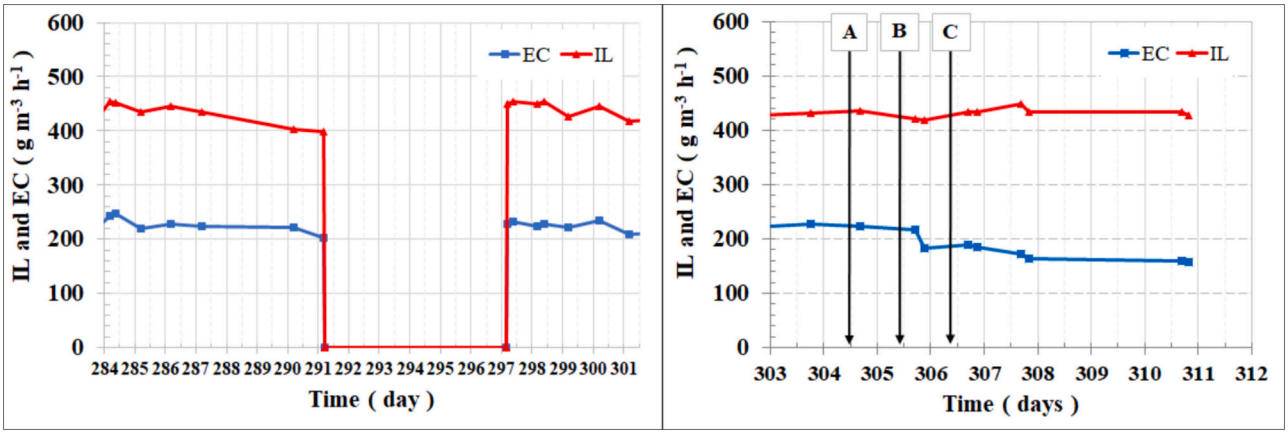

Fig. 4. Methane elimination capacity (EC) and inlet load (IL) before and after a six-day methane supply interruption (left), and before and after increasing the surfactant concentration (right) with (A) the additional 80 mg BRIJ 58 L^{-1}, (B) the additional 160 mg BRIJ 58 L^{-1}, and (C) the additional 320 mg BRIJ 58 L^{-1}.

$$\tau_{rz} = -\rho \times g \times R/2\left(1-((R-\delta)/R)^2\right) \quad (10)$$

The negative sense indicates that the force points downwards. The film thickness was estimated using the correlation proposed by Liu et al. [33]. The shear stress in the capillary channel along the liquid slug would increase with the liquid velocity and the liquid viscosity as illustrated in Fig. 5a below.

Saur et al. [28] showed in a Couette-Taylor reactor that the shear stress of the liquid on a surface wall can strongly impact the initial bacterial adhesion. Their study showed that an increasing shear stress up to 3.7 Pa initially stimulated adhesion in their experimental set-up (likely due to the increased liquid transport facilitating the access of bacteria to the wall surface), while a higher shear force of 7.3 Pa reduced biofilm formation (likely due to the increased detachment forces). However, biofilm formation was not prevented at 7.3 Pa, which is consistent with other studies where high shear forces (6 to 20 Pa) alone could not prevent biofilm formation [31,34].

Our long-term experiments using a 2.4 mm capillary channel and liquid velocities between 0.15 and 0.84 m s $^{-1}$ for 238 days (Test Series B), or liquid velocities between 1.3 and 2.5 m s $^{-1}$ for 305 days (Test Series C), did not show accumulation of biomass nor any signs of biofilm formation on the capillary channel walls. The shear stress in our experiments can be expected to be in a similar range (Fig. 5a) as the shear stress estimated in the experiments of Saur and coworkers, and therefore the wall shear stress generated by the liquid slugs alone cannot explain the absence of biofilm formation on the capillary channel wall in our studies.

Other explanations may be the difference in wall material (glass in our study versus plastic in Saur and coworkers' study, as probably less shear force is necessary to avoid cell attachment in the glass compared to plastic) and/or the different experimental set up (capillary reactor with small diameter capillary channels versus a Couette-Taylor reactor consisting of two concentric glass cylinders, a rotating inner cylinder and a non-rotating outer cylinder). Saur et al. [28] operated the Couette-Taylor reactor with a 28 mm gap between the cylinders under conditions that the Taylor vortex inside the liquid would not be present, while our capillary reactor with the 2.4 mm channels was operated in a way that the Taylor vortex would be expected. The presence of the Taylor flow containing a recirculating liquid vortex could be the critical factor preventing the accumulation of biomass on the inner walls of the channels, since it causes shear stress alteration as illustrated in Fig. 5b. In an up-flow configuration where the bubble train flows upward against gravity, the shear force is upward when the liquid slug passes, but downwards when the gas bubble with falling film passes any point on the capillary channel wall. This creates a pulsating shear stress possibly contributing to limiting biomass adhesion which may explain the absence of biomass on the inner walls of the channels. Nevertheless, further studies would be required to better understand the contribution of these factors involved in preventing accumulation of biomass on the walls of the capillary channels.

3.6. Energy input evaluation

The energy required for the operation of a CBR would be mainly determined by the pressure loss when the gas-liquid slugs flow through the capillary channels. The total pressure loss per length unit in a capillary channel with gas-liquid segmented flow is caused by several frictions: (1) the wall friction of the liquid slug, (2) the static head of the liquid in the capillary channel (in case of vertical channel configuration), and (3) the wall friction of the gas bubble. The liquid wall friction can be estimated using the Hagen-Poiseuille equation (Eq. (11)), while the static head can be calculated using the volume and density of the liquid slugs (Eq. (12)).

$$dP_{LWF}/L = 32\,\mu \times u_L/d^2 \quad (11)$$

$$dP_{LSH}/L = \rho \times g \quad (12)$$

where dP_{LWF} stands for the pressure loss in a capillary caused by liquid wall friction (Pa), dP_{LSH} the liquid static head pressure loss (Pa), L the length of the capillary channel (m), μ the viscosity (Pa s), u_L the superficial velocity of the liquid slug (m s^{-1}), d the diameter of the capillary channel (m), ρ the liquid density (kg m^{-3}), and g the gravitational constant (m s^{-2}). The wall friction of the gas bubbles was determined experimentally and reported elsewhere [16].

Based on the measured pressure loss by the gas (air bubble) and the calculated pressure loss by the liquid (liquid slug), the total pressure loss per meter capillary channel can be estimated. The overall pressure drop is mainly influenced by the internal diameter of the capillary channel, the slug velocity through the capillary channel, as well as the gas-to-liquid ratio. Fig. 6 shows that the calculated pressure drop over a 2.4 mm capillary channel is 297 Pa per meter at slug velocity of 0.5 m s^{-1} and G/L of 9, while up to 6426 Pa per meter at slug velocity of 2.5 m s^{-1} and G/L of 1.

The contribution of (1) the wall friction of the liquid slug, (2) the static head of the liquid in the capillary channel, and (3) the wall friction of the gas bubble significantly differs depending on the gas-to-liquid ratio, the slug velocity and the diameter of the capillary channel. The gas wall friction forces are in general negligible but cannot be ignored for the larger channel diameters at the higher G/L ratio.

There are other forces that should be considered and may be important depending on the configuration and the operating conditions of the CBR system. First, the entrance pressure losses generated by the inlet side of the capillary channel where gas and liquid are mixed to form the gas-liquid bubble train need to be taken into account. Further

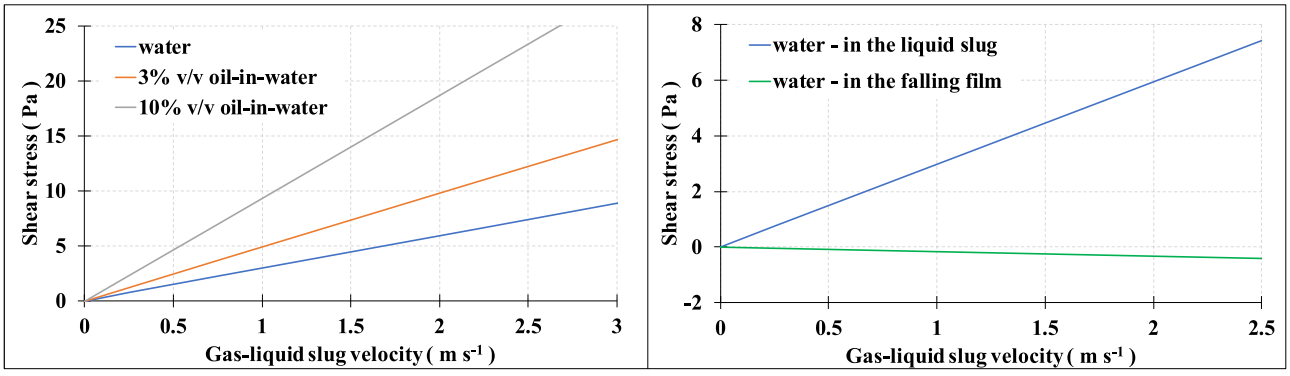

Fig. 5. The wall shear stress in the liquid slug (left) in the 2.4 mm capillary channel as a function of liquid superficial velocity for water (viscosity 0.89 mPa s) and two oil-in-water emulsions (oil viscosity 20 cSt = 20 mPa s). The wall shear stress in the liquid slug and in the falling film along gas bubble in the 2.4 mm capillary channel under segmented flow conditions as a function of gas-liquid superficial velocity for water (right).

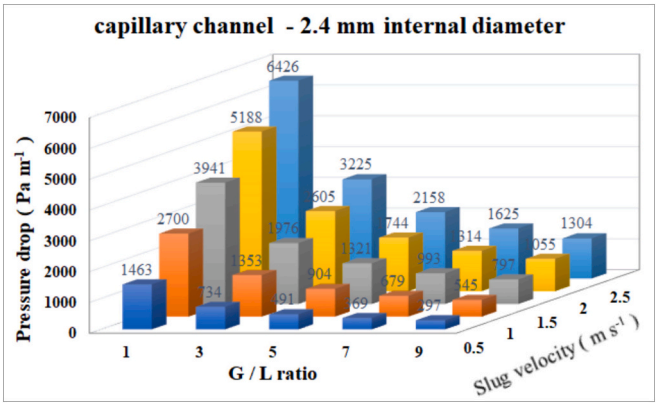

Fig. 6. The pressure drop as a function of the gas-to-liquid ratio and the slug face velocity for the 2.4 mm diameter capillary channel.

research on gas-liquid zones for multi-channel capillary bioreactors would therefore be valuable. Furthermore, the Laplace pressure, which is the pressure difference caused by the surface tension of the gas-liquid interface, governs also the pressure drop in Taylor flow reactors. The Laplace pressure increases with smaller capillary channel diameter and is proportional to the number of gas bubbles per unit length. Kreutzer et al. [12] determined that the Laplace pressure may become important for dimensionless slug lengths shorter than 10 times the capillary channel diameter. In this study the slug length observed in the CBR experiments (1.7 mm and 2.4 mm diameter channels) was typically between 2 and 3 cm, which means a dimensionless slug length between 8 and 15 and thus close to where the Laplace pressure may not be considered negligible. Finally, any non-aqueous liquid additive (i.e., silicone oil) should have a low viscosity to minimize not only the liquid wall friction (as illustrated in Eq. (11)), but also to minimize a possible pressure drop caused by the Laplace pressure losses.

4. Conclusions

This study showed that high methane removal capacities can be obtained in a capillary bioreactor, especially when using an altered liquid phase and when operated with internal gas recirculation. Although the addition of only surfactant or only silicone oil did not show any enhancement, the capillary bioreactor containing silicone oil and surfactant enhanced methane removal by 40 %. Silicone oil acting as buffer for methane was confirmed in experiments with transient methane conditions. No biomass accumulation on the walls of the 2.4 mm capillary glass channels was observed, possibly by the pulsating shear stress created by the segmented (Taylor) gas-liquid flow.

CRediT authorship contribution statement

Norbertus J.R. Kraakman: Writing – original draft, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Sergio Bordel:** Writing – original draft. **Raquel Lebrero:** Writing – review & editing, Supervision. **Raúl Muñoz:** Writing – review & editing, Supervision, Funding acquisition.

Declaration of competing interest

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jwpe.2025.107531.

Data availability

Data will be made available on request.

References

[1] IEA, World Energy Outlook 2021, International Energy Agency, Paris, France, 2021.
[2] F.M. O'Connor, O. Boucher, N. Gedney, C.D. Jones, G.A. Folberth, R. Coppell, P. Friedlingstein, W.J. Collins, J. Chappellaz, J. Ridley, C.E. Johnson, Possible role of wetlands, permafrost, and methane hydrates in the methane cycle under future climate change: a review, Rev. Geophys. 48 (2010) RG4005.
[3] J.H.M. Harmsen, D.P. van Vuuren, D.R. Navak, A.F. Hof, L. Hoglund-Isaksson, P. L. Lucas, J.B. Nielsen, P. Smith, E. Stehfest, Long-term marginal abatement cost curves of non-CO2 greenhouse gases, Environ. Sci. Pol. 99 (2019) 136–149.
[4] WMO, Greenhouse gas bulletin, in: The State of Greenhouse Gases in the Atmosphere Based on Global Observations Through 2010, World Meteorological Organization, Geneva, 2011.
[5] Methane Tracker, Analysis, IEA, 2023. https://www.iea.org/reports/global-methane-tracker-2023 (accessed, August 18, 2023).
[6] CCAC, Global methane assessment: benefits and costs of mitigating methane emissions, in: United Nations Environment Programme (Report), 2021, p. 173 (ISBN 978-92-807-3854-4).
[7] R.M. Gloria, T.M. Molla, P.V.O. Siva, P. Costa, E.M.F. Brandt, C.L. Souza, C.A. L. Chernicharo, Stripping and dissipation techniques for the removal of dissolved gases from anaerobic effluents, Braz. J. Chem. Eng. 33 (4) (2016) 713–721.
[8] I. Pecorini, R. Iannelli, Landfill GHG reduction through different microbial methane oxidation biocovers, Processes 8 (2020).
[9] BCME, British Columbia Ministry of Environment – landfill gas management facilities design guidelines. http://www.env.gov.bc.ca/epd/mun-waste/waste-solid/landfills/pdf/Design-guidelines-final.pdf, 2010.
[10] Q. Huang, Q. Zhang, N. Cicek, D. Mann, Biofilter: a promising tool for mitigating methane emission from manure storage, J. Arid. Land 3 (2011) 61–70.
[11] H. Limbri, C. Gunawan, B. Rosche, J. Scott, Challenges to developing methane biofiltration for coal mine ventilation air: a review, Water Air Soil Pollut. 224 (2013) 1–15.
[12] M.T. Kreutzer, F. Kapteijn, J.A. Moulijn, J.J. Heiszwolf, Multiphase monolith reactors: chemical reaction engineering of segmented flow in microchannels, Chem. Eng. Sci. 60 (2005) 5895–5916.
[13] T.A. Nijhuis, M.T. Kreutzer, A.C.J. Romijn, F. Kapteijn, J.A. Moulijn, Monolith catalysts as efficient three-phase reactors, Chem. Eng. Sci. 56 (3) (2001) 823–829.
[14] N. Shao, A. Gavriilidis, P. Angeli, Flow regimes for adiabatic gas-liquid flow in micro-channels, Chem. Eng. Sci. 64 (2009) (2009) 2449–2761.
[15] S. Haase, D.Y. Murzin, T. Salmi, Review on hydrodynamics and mass transfer in minichannel wall reactors with gas–liquid Taylor flow, Chem. Eng. Res. Des. 113 (2016) 304–329.
[16] N.J.R. Kraakman, J. González-Martína, E. Rodriguez, R. Lebrero, M.A. Deshusses, R. Muñoz, Hydrophobic air pollutants removal at one second gas contact in a multi-channel capillary bioreactor, J. Environ. Chem. Eng. 11 (2023) 110502.
[17] L.R. Lopez de Leon, K. Deaton, J. Junkin, M.A. Deshusses, Capillary microreactors for VOC vapor treatment: impacts of operating conditions, Chemosphere 258 (2020) 127286.
[18] J.M. Estrada, R. Lebrero, G. Quiano, R. Perez, I. Figueroe-Gonzalez, P.A. Garcia-Encina, R. Munoz, Methane abatement in a gas-recycling biotrickling filter – evaluating innovative operational strategies to overcome mass-transfer limitations, Chem. Eng. J. 253 (2014) 385–393.
[19] N.J.R. Kraakman, L. Villarreal-Heras, J. González-Martín, S. Cantera, R. Muñoz, R. Lebrero, Enhancing dilute methane treatment through liquid phase alteration in a capillary bioreactor, Chem. Eng. J. 510 (2025) 161383.
[20] S. Bordel, N.J.R. Kraakman, R. Muñoz, Theoretical analysis of gas–liquid mass transfer in Taylor flow capillary reactors, Chem. Eng. Sci. 292 (2024) 119949.
[21] T.C. Thulasidas, M.A. Abraham, R.L. Cerro, Flow patterns in liquid slugs during bubble-train flow inside capillaries, Chem. Eng. Sci. 52 (1997) 2947–2962.
[22] X. Wu, Y. Lin, Y.Y. Wang, S.H. Wu, X. Li, C.P. Yang, Enhanced removal of hydrophobic short-chain n-alkanes from gas streams in biotrickling filters in presence of surfactant, Environ. Sci. Technol. 56 (2022) 10349–10360.
[23] C. Kennelly, S. Gerrity, G. Collins, E. Clifford, Liquid phase optimisation in a horizontal flow biofilm reactor (HFBR) technology for the removal of methane at low temperatures, Chem. Eng. J. 242 (2014) 144–154.
[24] R. Lebrero, L. Hernandez, R. Perez, J.M. Estrada, R. Munoz, Two-liquid phase partitioning biotrickling filters for methane abatement: exploring the potential of hydrophobic methanotrophs, J. Environ. Manag. 151 (2015) 124–131.
[25] M. Veillette, M. Girard, P. Viens, R. Brzezinski, M. Heitz, Function and limits of biofilters for the removal of methane in exhaust gases from the pig industry, Appl. Microbiol. Biotechnol. 94 (2012) 601–611.
[26] P.A. Lamprea Pineda, K. Demeestere, M. Toledo, H. Van Langenhove, C. Walgraeve, Enhanced removal of hydrophobic volatile organic compounds in

biofilters and biotrickling filters: a review on the use of surfactants and the addition of hydrophilic compounds, Chemosphere 279 (2021) 130757.
[27] D. Zhang, L. Zhu, F. Li, Influences and mechanisms of surfactants on pyrene biodegradation based on interactions of surfactant with a Klebsiella oxytoca strain, Bioresour. Technol. 142 (2013) 454–461.
[28] T. Saur, E. Morin, F. Habouzit, N. Bernet, R. Escudié, Impact of wall shear stress on initial bacterial adhesion in rotating annular reactor, PLoS One 12 (2) (2017) e0172113.
[29] L. Wang, R. Keatch, Q. Zhao, J.A. Wright, C.E. Bryant, A.L. Redmann, E. M. Terentjev, Influence of type I fimbriae and fluid shear stress on bacterial behavior and multicellular architecture of early Escherichia coli biofilms at single-cell resolution, Appl. Environ. Microbiol. 84 (2018) (e02343-17).
[30] F. Habouzit, G. Gévaudan, J. Hamelin, J.-P. Steyer, N. Bernet, Influence of support material properties on the potential selection of Archaea during initial adhesion of a methanogenic consortium, Bioresour. Technol. 102 (2011) 4054–4060 (pmid: 21211965).
[31] S. Lecuyer, R. Rusconi, Y. Shen, A. Forsyth, H. Vlamakis, R. Kolter, et al., Shear stress increases the residence time of adhesion of Pseudomonas aeruginosa, Biophys. J. 100 (2011) 341–350.
[32] A. Park, H.-H. Jeong, J. Lee, K.P. Kim, C.-S. Lee, Effect of shear stress on the formation of bacterial biofilm in a microfluidic channel, Biochip J. 5 (2011) 236–241.
[33] H. Liu, C.O. Vandu, R. Krishna, Hydrodynamics of Taylor flow in vertical capillaries: flow regimes, bubble rise velocity, liquid slug length, and pressure drop, Ind. Eng. Chem. Res. 44 (14) (Jul. 2005) 4884–4897, https://doi.org/10.1021/ie049307n.
[34] L.C. Simões, I.B. Gomes, H. Sousa, A. Borges, M. Simões, Biofilm formation under high shear stress increases resilience to chemical and mechanical challenges, Biofouling 38 (1) (2022) 1–12, https://doi.org/10.1080/08927014.2021.2006189.
[35] J. Nikiema, M. Heitz, The influence of the gas flow rate during methane biofiltration on an inorganic packing material, Can. J. Chem. Eng. 87 (2009) 136–142.
[36] J. Nikiema, M. Heitz, The use of inorganic packing materials during methane biofiltration, Int. J. Chem. Eng. (2010) 8, https://doi.org/10.1155/2010/573149, 573149.
[37] S. Gomez-Cuervo, C. Alfonsin, J. Hernandez, G. Feijoo, M.T. Moreira, F. Omil, J. Clean. Prod. 143 (2017) 1191–1202.
[38] T.G. Kim, S. Jeong, K. Cho, Functional rigidity of a methane biofilter during temporal microbial succession, Appl. Microbiol. Biotechnol. 98 (2014) 3275–3286.
[39] J. Hernandez, S. Gomez-Cuervo, F. Omil, EPS and SMP as stability indicators during the biofiltration of diffuse methane emissions, Water Air Soil Pollut. 226 (2015) 343.
[40] M. Ferdowsi, M. Veillette, A.A. Ramirez, J.P. Jones, M. Heitz, Performance evaluation of a methane biofilter under steady state, transient state and starvation conditions, Water Air Soil Pollut. 227 (2016) 168.
[41] G. Ganendra, D. Mercado-Gracia, E. Hernandez-Sanabria, N. Peiren, S. De Campeneere, Ho., Adrian, Boon, N., Biofiltration of methane from ruminants gas effluent using autoclaved aerated concrete as the carrier material, Chem. Eng. J. 277 (2015) 318–323.
[42] E.M. Brandt, F.V. Duarte, J.P.R. Vieira, V.M. Melo, C.L. Zouza, J.C. Araujo, C.A. L. Cherbicharo, The use of novel packing material for improving methane oxidation in biofilters, J. Environ. Manag. 182 (2016) 412–420.
[43] M.T. Sun, Z.-M. Yang, S.F. Fu, Z.L. Fan, R.B. Guo, Improved methane removal in exhaust gas from biogas upgrading process using immobilized methane-oxidizing bacteria, Bioresour. Technol. 256 (2018) 201–207.
[44] M. Ferdowsi, B. Khabiri, G. Buelna, J.P. Jones, M. Heitz, Prolonged operation of a methane biofilter from acclimation to the failure stage, Environ. Technol. 45 (13) (2023) 2589–2598.
[45] A.A. Ramirez, B.P. García-Aguilar, J.P. Jones, M. Heitz, Improvement of methane biofiltration by the addition of non-ionic surfactants to biofilters packed with inert materials, Process Biochem. 47 (2012) 76–82.
[46] L. Fjedsted, C. Scheutz, A.G. Christensen, J.E. Larsen, P. Kjeldsen, Biofiltration of diluted landfill gas in an active loaded open-bed compost filter, Waste Manag. 103 (2020) 1–11.
[47] J. Rocha-Rios, S. Bordel, S. Hernandez, S. Revah, Methane degradation in two-phase partition bioreactors, Chem. Eng. J. 152 (2009) 289–292.
[48] J. Rocha-Rios, N.J.R. Kraakman, R. Kleerebezem, M.T. Kreutzer, M.C.M. van Loosdrecht, A capillary bioreactor to increase methane transfer and oxidation through Taylor flow formation and transfer vector addition, Chem. Eng. J. 217 (2013) 91–98.
[49] M. Girard, A.A. Ramirez, G. Buelna, M. Heitz, Biofiltration of methane at low concentrations representative of the piggery industry—Influence of the methane and nitrogen concentrations, Chem. Eng. J. 168 (2011) 151–158.