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Influence of the gas-liquid flow configuration in the absorption column on photosynthetic biogas upgrading in algal-bacterial photobioreactors

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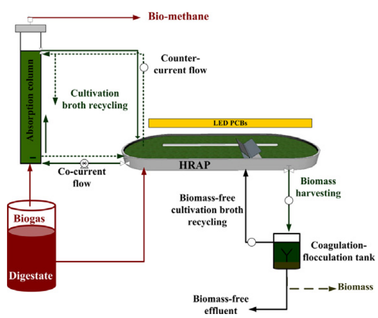
HIGHLIGHTS

- EU standard bio-methane was obtained regardless of the gas-liquid flow configuration.
- Optimum bio-methane composition was achieved at a L/G = 0.5 under co-current operation.
- Counter-current operation decreased biomass productivity and the cultivation broth pH.
- High C, N, P and S recoveries were achieved by decoupling the HRT from the SRT.

GRAPHICAL ABSTRACT



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ABSTRACT

The potential of an algal-bacterial system consisting of a high rate algal pond (HRAP) interconnected to an absorption column (AC) via recirculation of the cultivation broth for the upgrading of biogas and digestate was investigated. The influence of the gas-liquid flow configuration in the AC on the photosynthetic biogas upgrading process was assessed. AC operation in a co-current configuration enabled to maintain a biomass productivity of 15 $g\,m^{-2}\,d^{-1}$, while during counter-current operation biomass productivity decreased to 8.7 ± 0.5 $g\,m^{-2}\,d^{-1}$ as a result of trace metal limitation. A bio-methane composition complying with most international regulatory limits for injection into natural gas grids was obtained regardless of the gas-liquid flow configuration. Furthermore, the influence of the recycling liquid to biogas flowrate (L/G) ratio on bio-methane quality was assessed under both operational configurations obtaining the best composition at an L/G ratio of 0.5 and co-current flow operation.

1. Introduction

Anaerobic digestion is a sustainable platform technology to reduce the environmental impact of biodegradable organic wastes. During anaerobic digestion, ~20–95% of this residual organic matter is biologically converted into biogas (consisting of 50–70% of CH_4, 30–50% of CO_2 and trace gases such as H_2S, H_2 and N_2 (Appels et al., 2011)) and digestate (a nutrient rich liquid effluent) (Möller and Müller, 2012). Biogas is a renewable energy source typically used in industry for heat and power generation or as natural gas substitute after upgrading. Nowadays, the high energy and chemicals consumption associated to conventional physical-chemical technologies for biogas upgrading (to a CH_4 content of at least 95% as required by most international bio-methane standards) limits their environmental and economic sustainability (Muñoz et al., 2015). On the other hand, digestate is applied in agriculture as biofertilizer, although environmental problems such as ammonia emission, nitrate leaching or phosphorus soil saturation might derive from inappropriate digestate handling, storage and application (Holm-Nielsen et al., 2009).

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In this context, photosynthetic biogas upgrading coupled to nutrient removal from digestate can enhance the sustainability and economic viability of biogas and digestate management (Bahr et al., 2014; Posadas et al., 2015; Serejo et al., 2015; Yan et al., 2016a,b). During photosynthetic biogas upgrading, microalgae use light energy to fix the CO_2 from biogas via photosynthesis, while sulphur-oxidizing bacteria oxidize H_2S to sulphate using the O_2 photosynthetically produced. Both microalgal and bacterial growth can be supported by the N and P contained in wastewaters from different sources (*i.e.* digestate or livestock waste compost), with the subsequent reduction of their eutrophication potential (Bahr et al., 2014; Zhu and Hiltunen, 2016). Recently, an innovative high rate algal pond (HRAP) operational strategy based on decoupling the hydraulic retention time (HRT) from the solids (biomass) retention time (SRT) was developed. This strategy allows maximizing nutrient recovery from high-strength wastewaters (*i.e.* digestate) in the form of algal-bacterial biomass while maintaining biomass concentration below light limiting values (Toledo-Cervantes et al., 2016). The algal–bacterial biomass produced during photosynthetic biogas upgrading can be used as slow-release bio-fertilizer or as a feedstock for biofuel production, thus contributing to improve the economic and environmental viability of this innovative technology (Posadas et al., 2014).

Despite the high potential of photosynthetic biogas upgrading, N_2 and O_2 stripping from the recycling cultivation broth to the upgraded biogas often results in CH_4 concentrations <95%. (Muñoz et al., 2015). N_2 and O_2 are often present in the recycling cultivation broth at concentrations of $\sim$14 mg-N_2 L^{-1} and >8 mg-O_2 L^{-1} as a result of its direct contact with the atmosphere (in open HRAPs) and the intensive microalgal photosynthetic activity in the photobioreactor, respectively (Toledo-Cervantes et al., 2016). In fact, the O_2 stripped out from the cultivation broth is a function of the biomass productivity, which is directly linked to the irradiation impinging into the cultivation broth. All studies evaluating the performance of this technology to date were conducted under low light intensities (75–420 μmol m^{-2} s^{-1}), which could have partially biased the results obtained in terms of final bio-methane quality (Posadas et al., 2015; Serejo et al., 2015; Toledo-Cervantes et al., 2016). On the other hand, the liquid to biogas flow (L/G) ratio in the external absorption column (AC) has been recently identified as one of the key operational parameters determining the final composition of bio-methane. Unfortunately, the influence of the biogas/recycling liquid flow configuration in the AC (counter-current *vs* co-current) on bio-methane composition has not been yet systematically assessed. Meier et al. (2015) operated a counter-current flow bubble column interconnected to a stirred tank photobioreactor and reported a bio-methane O_2 content of $\sim$1.2% at an L/G of 6.3. Likewise, bio-methane O_2 concentrations ranging from 0.7 to 1.2 were recorded by Posadas et al. (2015) in a HRAP interconnected to a bubble column operated at co-current flow. These O_2 concentrations were significantly higher than the limit of 0.3% required by most international regulations for bio-methane injection into natural gas networks, which entails the need for a systematic optimization of biogas scrubbing in the absorption column of photosynthetic biogas upgrading systems.

This research assessed for the first time the influence of the gas-liquid flow configuration (co-current and counter-current) in the AC on bio-methane quality and nutrient recovery performance from real digestate. Biogas-liquid mass transfer in the absorption column strongly impacts the quality of the bio-methane, which is of key importance for complying with the legal specifications for bio-methane injection in the natural gas grid or use as autogas. Additionally, the influence of the L/G ratio (0.3–1) on bio-methane quality was investigated under steady state at the two target gas-liquid flow configurations in order to minimize both O_2 and N_2 content.

2. Materials and methods

2.1. Experimental set-up operation

The experimental set-up consisted of a HRAP interconnected to a bubble column (referred to as absorption column, AC) and to a harvesting tank via recirculation of the cultivation broth (Fig. S1, Supplementary information). The system was operated indoors at the Dept. of Chemical Engineering and Environmental Technology at University of Valladolid (Spain). The HRAP dimensions were 170 cm length and 82 cm width, with a working volume of 180 L and an illuminated area of 1.210020m^2. The HRAP was continuously agitated at an internal liquid recirculation velocity of 20 cm s^{-1} and illuminated at 1500 ± 600 μmol m^{-2} s^{-1} by six high intensity LED PCBs (Phillips SA, Spain) using 14:10 h light:dark cycles. The composition of the rendering digestate fed continuously at an influent flow rate of 1 L d^{-1} was (mg L^{-1}): ammonium (NH_4^+) 1668 ± 249, total nitrogen (TN) 1815 ± 109, total phosphorous (TP) as P-PO_4^{-3} 48 ± 2, chemical oxygen demand (COD) 1745 ± 413, inorganic carbon (IC) 1500 ± 168 and sulphate (SO_4^{-2}) 15 ± 2. Tap water was daily supplied to the HRAP to compensate for evaporation losses. The AC (165 cm height and 4.4 cm diameter) was fed with a synthetic biogas mixture (70% of CH_4, 29.5% of CO_2 and 0.5% of H_2S, Abello Linde (Barcelona, Spain)) and cultivation broth from the HRAP at a similar flow rate of 1.6 m^3 m^{-2} h^{-1} (flow rate referred to the AC cross sectional area). The algal-bacterial cultivation broth exiting the AC was returned to the HRAP. A fraction of the cultivation broth (26 L d^{-1}) was transferred to an external stirred tank for biomass harvesting, thus decoupling biomass productivity from the HRT. A polyacrylamide-based flocculant solution (Chemifloc CV-300, (de Godos et al., 2011)) was dosed at 120 mg L^{-1} to recover the algal-bacterial biomass by coagulation-flocculation. The biomass-free cultivation broth was then returned to the HRAP. This harvesting method represents a low cost alternative for algal-bacterial broths with a sludge volume index >100 mL g^{-1}. The effluent from the system was removed at 0.5 L d^{-1} from the harvesting tank, along with the flocculated biomass in the stirred tank, in order to minimize the effluent discharged into the environment while avoiding the accumulation of potentially toxic compounds present in the digestate.

2.2. Influence of the gas-liquid flow configuration on biogas upgrading and nutrients recovery

The HRAP was inoculated with *Mychonastes homosphaera* (Skuja) Kalina & Puncochárová (a taxonomic synonym of *Chlorella minutissima* Fott & Nováková) from a previous culture grown in synthetic anaerobically digested stillage (Toledo-Cervantes et al., 2016). The AC was operated under co-current flow for 94 days (stage I) and for 110 days (stage II) under a counter-current flow configuration. Samples of 100 mL from the rendering digestate and the cultivation broth were collected twice a week to measure the pH and concentration of IC, TN, NH_4^+, TP, nitrite (NO_2^-), nitrate (NO_3^-), SO_4^{2-} and TSS. The inlet and outlet biogas flow rate and composition (CO_2, H_2S, O_2, N_2, and CH_4) were also recorded twice a week. Temperature and dissolved O_2 concentration were *in-situ* determined in the HRAP. Algal-bacterial cultivation broth samples were drawn at each steady state to characterize the structure of the population of both microalgae and bacteria, and their elemental composition (C, N, P and S).

2.3. Influence of the L/G ratio on bio-methane composition under co-current and counter-current operation

Liquid to biogas flow rate ratios ranging from 0.3 to 1.0 were tested under co-current and counter-current operation. The