ICA2025OSLO
14th IWA Specialised Conference on
Instrumentation, Control and Automation

# Modelling of a trickling filter bioreactor for ex-situ hydrogenotrophic methanation with gaseous effluent feedback

F. A. Ortiz-Ricardez*, K. M. Muñoz-Páez**, A. Vargas-Casillas***.

*fortizr@iingen.unam.mx; **kmunozp@iingen.unam.mx; ***avargasc@iingen.unam.mx

**Abstract:**

A mathematical model for a hydrogenotrophic methanation process in a trickling filter bioreactor (TFB) is formulated and simulated. The model separates the physical space in an arbitrary number of vertical levels and uses the first Fick's diffusion law along its vertical axis and along the thickness of the biofilm layer attached to the inert bed material. According to the general ideal gas law, computing the gas flows among levels requires to consider the fixed amount of moles of gas in each TFB level. Simulations of the proposed model were compared to experimental results of ex-situ hydrogenotrophic methanization in a TFB. Model performance compared to experimental results of the real reactor fitted remarkably well the effluent proportions and productivity, this considering an ideal scenario in which the model formulation assumes that hydrogenotrophic methanation by archaea as the only biological reaction. A key model-based enhancement is the back-feeding of the gaseous effluent flow, which resulted in a remarkable improvement for fitting the experimental results regarding biomethane productivity. A qualitative analysis of important model parameters was crucial for fitting simulation results to real experimental data. In this sense, influent raw biogas proportions near the ideal stoichiometry for hydrogenotrophic methanation is detrimental to purity and productivity of the biomethane effluent. Interestingly, the numerical effort needed for simulations was substantially minor than expected, as large-sized models may result using our modelling approach.

**Keywords**: Bioenergy production; Modelling and identification; Monitoring; Parameter and state estimation

## 1. Introduction

Frequently used as pollutant removal schemes in water and gas treatment, trickling filter bioreactors (TFB) are appealing due to their versatility. Computer fluid dynamics (CFD) has been used to study the mass transfer processes occurring in biotrickling filters; the resulting distributed parameter models, founded on partial differential equations derived from the Navier-Stokes' equations, may describe precisely the fluid dynamics involved. Nevertheless, they are not handy for control purposes, since simpler models may help to set out control directives on variables that can directly be handled via an online actuator, such as a modification of the gaseous and/or hydraulic retention times; thus, lumped-parameter models based on ordinary algebraic-differential equations are more desirable. For biogas refinement, hydrogenotrophic methanation is a process in which archaea metabolize hydrogen ($H_2$) and carbon dioxide ($CO_2$) to produce methane ($CH_4$) as a metabolite (Burkhardt and Busch, 2013). Raw biogas contains mostly $CO_2$ and $CH_4$ as predominant components, present in volumetric concentrations of $CH_4$ between 53–70%, and between 30–47% of CO2 originated from anaerobic digestion of sewage sludge, livestock manure or agro-industrial biowastes (Muñoz et al., 2015). Afterwards, raw biogas is converted into biomethane, whose composition complies with the natural gas quality on the market, and can be straightforwardly put in infrastructure that complies with the directives of transportation of natural gas (Cappannelli et al., 2020).

Experimental enhancing of biomethane from raw biogas was successfully tested by Muñoz-Páez et al. (2025), favouring biomethane productivity using raw biogas with low volumetric ratios of around $H_2$:$CO_2$ = 4:3; hence the formulation of a mathematical model is attractive for studying such an advantageous process and proposing feedback control laws for increasing productivity and/or purity, as well as for robustification. Modelling approaches based on the Fick's first diffusion law, and the segmentation into levels of the trickling bed, as proposed by Dupnock and Deshusses (2021), are starting points of this work, but in addition, it is taken into account the physical restriction of a maximum total number of moles in a given TBF level volume according to the general gas equation.

The objective of this paper is to mathematically model a hydrogenotrophic methanation process in a trickling filter-based reactor, and to numerically simulate such model to compare its performance against the experimental results of a real reactor set-up, obtained by Muñoz-Páez et al. (2025). The proposed model considers gaseous diffusion transport through the trickling filter, the mass transfer among liquid and gaseous phases, and also the reaction of absorbed gases in the biofilm attached to the bed. Wet (i.e. completely soaked) and moist (i.e. sufficiently humidified) biofilms, gas volume conversion from $H_2$ and $CO_2$ into $CH_4$, and axial dispersion are all included in the model. In addition, gaseous effluent recirculation to the gaseous influx of the reactor is considered, as this is expected to improve the biomethane purity of the effluent.

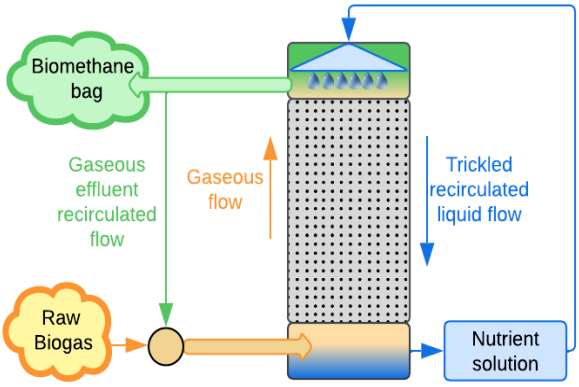


**Figure 1.1** Trickling filter bioreactor (TFB) scheme.

The TFB is assumed as fed with a gas flow from below and with a liquid flow from above that recirculates from the bottom, and the gaseous outflow is assumed to exit the system from above (see Figure 1.1). In addition, the hydrogenotrophic methanation reaction is stoichiometrically described by Gujer and Zehnder (1983) as $CO_2 + 4H_2 \rightarrow CH_4 + 2H_2O$, where gaseous and liquid proportions of water are conventionally neglected due to their relatively insignificant contribution to the overall liquid volume. The proposed model considers the following important assumptions:

- The TFB is modelled with $n$ vertical levels, indicated with the subscript $i$, such that $i$=1 is the lowermost level and $i = n$ the uppermost one; the total volume of each level (contained in the centre dashed square of Figure 1.2) is fixed.

- On each level $i$, 4 media coexist divided in layers: a gas phase (one layer), a liquid phase (one layer), and two types of biofilm with $p$ layers, where the active biomass is only present. These are: wet biofilm, in contact with the layer of liquid phase, and moist biofilm, in contact with the gas phase (actually the liquid layer is assumed irrelevantly thin). Index used for all layers is $j$, so $j$=1 is the innermost layer of moist biofilm, $j$=$p$+1 is the gas phase, $j$=$p$+2 is the liquid phase, and $j$=2$p$+2 is the innermost layer of wet biofilm.

- Components are the gases or other metabolites (if needed); currently only 3 are considered and represented by index k ∈ {1,2,3} for $CH_4$, $CO_2$ and $H_2$, respectively.

- Concentrations for the k-th component, at the i-th level, in the j-th layer or phase are represented by $C_{j,k,i}$; this quantity may have units g/L, kg/m$^3$, etc.

- The volume a gas can occupy in a level is $V_{G,i}$ and is assumed constant; hence, volumes for liquid, inert material bed and biofilm in a level are also constant.

- Biomass transfer between levels and layers, and a biological reaction in the liquid phase are assumed non-existent.

For ease of presentation, the indices $j=p+1$ or $j=p+2$ is sometimes replaced by $G$ or $L$ to emphasize that it is the gas or the liquid phase, while the indices $j=p$ and $j=p+3$ are replaced by $M$ and $W$ for the outermost layer of the moist or wet biofilm, respectively.

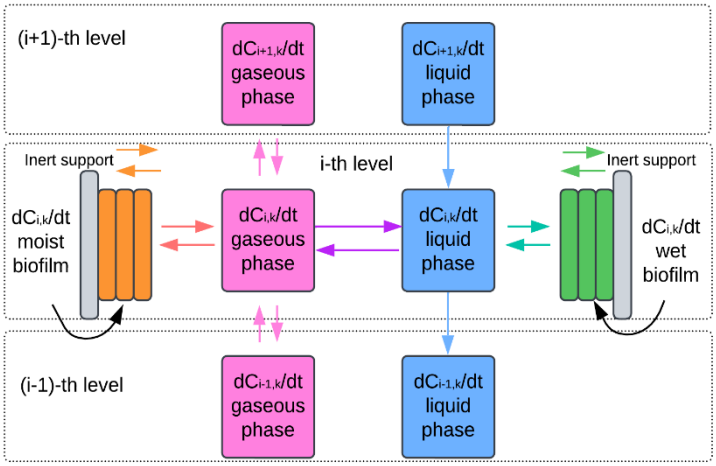


**Figure 1.2** TFB levels and interactions between layers.

## 2. Findings and results

A mathematical lumped-parameter model for a trickling filter bioreactor was proposed and simulated using MATLAB©. The execution time of the simulation resulted surprisingly faster than expected, considering the potentially large-sized models originated using the presented approach. Shown in Figure 2.1, the simulation results fitted remarkably well the experimental data originated from a 69 days experiment set up on the real trickling filter bioreactor, and model simulation condensed experimental time in an execution time of less than 3 minutes on average using a regular laptop computer.

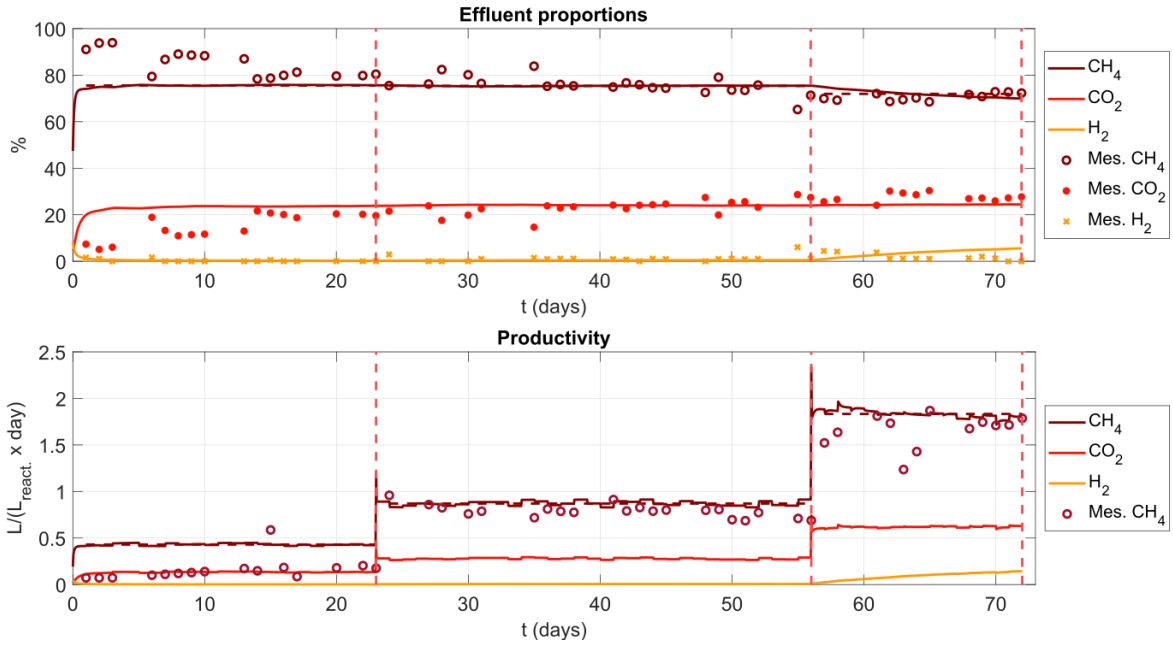


**Figure 2.1** Gaseous effluent proportions and $CH_4$ productivity.

The addition of gaseous outflow storage dynamics and the recirculation of the gaseous effluent resulted in a remarkable fitting to experimental results both in productivity and effluent volumetric proportions. Simulations fitted well the measured volumetric composition of gaseous effluent in the experiment. Productivity was not fitted as accurately as volumetric composition due to disregarding the influence of acetoclastic archaea in the proposed model, which produce additional $CO_2$, and due to also possible gaseous outflow proportions measurement errors; hence, simulation results for $CH_4$ productivity overestimated the measured productivity data of the experiment. In consequence, qualitatively sensitive variables such as the volumetric influent $H_2$:$CO_2$ ratio; and parameters such as the kinetic reaction rate, and the stoichiometric coefficients were crucial for fitting the model. In contrast, considering multiple biofilm layers had almost no effect on productivity or composition of the gaseous effluent.

As suggested by experimental results in other works, several simulations evidenced volumetric ratios for the gaseous influent near the stoichiometric ideal $H_2$:$CO_2$ = 4:1 for hydrogenotrophic methanation as detrimental for enhancing productivity and purity of biomethane. Diffusivity parameters defining Fickian diffusion were also relevant, but they are easily constrained due to physical limitations of influent flow, reactor volume, and liquid influent or mineral substrate liquid recirculation rate. Additional refinements of the model such as proposing other kinetic reaction functions, incorporation of variable liquid volume in the trickling filter levels, or considering phenomena related to acetoclastic archaea could be further developed. In addition to the gaseous influent, and the trickling liquid recirculation fluxes; the consideration of the gaseous effluent feedback as a new degree of freedom variable opens an alternative perspective for the control of ex-situ hydrogenotrophic methanization processes for producing bioenergy.

## REFERENCES

Burkhardt, M. & Busch, G. 2013. Methanation of hydrogen and carbon dioxide. *Applied Energy*, **111**, 74-79.

IEA 2020. *Outlook for biogas and biomethane: Prospects for organic growth.* International Energy Agency.

Dupnock, T.L. & Deshusses, M.A. 2021. Development and validation of a comprehensive model for biotrickling filters upgrading biogas. *Chemical Engineering Journal.*

Gujer, W. & Zehnder, A. 1983. Conversion processes in anaerobic digestion. *Water Science & Technology.*

Muñoz, R., Meier, L., Diaz, I., & Jeison, D. 2015. A review on the state-of-the-art of physical/chemical and biological technologies for biogas upgrading. *Reviews in Environmental Science and Bio/Technology.*

Muñoz-Páez, K.M., Ramos-Aréchiga, K.Y., & Buitrón, G. 2025. Ex-situ biogas enrichment by hydrogenotrophic methanogens at low H2/CO2 ratios: Effect of empty bed residence time. *Fuel*, **381**, 133330.