Modelling biofilm anaerobic reactor with effluent from hydrolytic/acidogenic reactor as substrate

Marisol Vergara Mendoza and Rodrigo Torres Sáez

ABSTRACT

This work presents modelling of an anaerobic biofilm reactor using ceramic bricks as support. The results were compared with the experimental data. It was observed that the substrate concentration curves showed the same tendency. The methane formation curves showed significant differences. The substrate removal efficiency was 83%. In the steady state, the experimental data were higher than the model, from the result the substrate degrading bacteria grew enough to reach biofilm and that the effect of the shear stress was more significant as the biofilm increased in thickness. To the methane production, the model in steady state reached a maximum value of 0.56 m^3 CH_4/m^3 *d and the experimental data reached 0.42 (m^3 CH_4/m^3 * d). The biofilm thickness calculated by the model was 14 μm.

Key words | acidogenic, biofilm, methanogenic, modelling

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INTRODUCTION

The anaerobic digestion of organic fraction of urban solid waste (OFUSW) can be carried out in two-phase anaerobic systems, in which the methanogenic phase is carried out in a biofilm reactor searching increased methane production. The anaerobic biofilm reactors are attractive for handling high organic loads, high biomass concentration, resistance to organic overloads and do not require mechanical mixing. This reactor compared to conventional systems reduces start-up time and increases organic loading speeds up to five times (Borkar *et al.* 2013; Takriff *et al.* 2014).

In the industry, different biofilm reactors have been successfully evaluated (Rajagopal *et al.* 2013). The anaerobic filter (AF) is a common biofilm reactor and its hydraulic retention time (HRT) is effective for wastewater treatment, but the organic loading rate is a limitation for handling complex wastewater; this water may contain fermentable substrates but also alternating electron acceptors, such as nitrates and sulfates. The AF can work with one or several feeds, up or down flow or horizontally (Rajinikanth *et al.* 2009). The packed bed reactors contain materials that provide a high surface area to attach microorganisms, an efficient mixing inside the reactor favors the dispersion of the volatile acids in the solution and biogas release (Rodgers *et al.* 2004). Fluidized bed reactors have better transfer characteristics when compared to fixed bed reactors (Campos-Pineda *et al.* 2012). These reactors have a high attached biomass which is rich in microbial diversity and recovers quickly after load instability conditions (Wang *et al.* 2009; De Amorin *et al.* 2015).

Mathematical models can be simple empirical correlations or algorithms that describe three-dimensional morphology of the biofilm. Analytical models employ simplified assumptions, such that the flow of the substrate inside the biofilm can be calculated without numerical techniques (Boltz *et al.* 2009). In mathematical models, the effects of each term, variable or parameter can be analyzed directly. Pseudoanalytical models are an alternative when simplifications are eliminated, achieving a more real representation of the system. The pseudoanalytical solutions comprise a set of equations that can be solved by means of simple programming (Sarkar & Mazumder 2015). The numerical models in one dimension represent the biofilm in a dimension perpendicular to the substrate. The equations can be solved numerically and the simulations with a certain degree of complexity through software (Boltz *et al.* 2010). In multidimensional numerical models, the biofilm is modeled as a two- or three-dimensional structure. All the components can vary in the multidimensional space, as well as in time. The premise of these models is that by capturing the spatial and temporal chemical, physical and biological heterogeneity, it is possible to obtain an evaluation of the biofilm and the interactions at a micro level (von der Schulenburg *et al.* 2009; Taherzadeh *et al.* 2012). Rittmann *et al.* 2018 developed a guide on how to apply a biofilm model to obtain accurate

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and meaningful results. These authors present an overview of biofilm models since it began in the mid-1970s.

In biofilm models, the computing time is often a limiting factor, the excess complexity increases the calculation time in orders of magnitude. On the other hand, a complex biofilm model requires providing many input parameters that are difficult to determine. The choice of a too complex model will incur many penalties to produce results that do not improve the utility of the model. For these reasons, the simplified model of a biofilm reactor provides the necessary information for monitoring the performance of the system.

The OFUSW can be treated by anaerobic digestion. The suspended reactors are commonly used for this kind of substrate. The organic loads for the treatment of this substrate are high, which can lead to operational failures in the reactors. Biofilm reactors offer the possibility to handle these high organic loads. The modeling of these reactors allows obtaining information related to the feasibility of its use in the treatment of this kind of waste. In this work, the modeling of the methanogenic phase was carried out in a biofilm reactor with substrate from a hydrolytic-acidogenic reactor of OFUSW.

MODEL DEVELOPMENT

In this work, a simplified approach for analyzing and modelling the biofilm process was used.

To model the substrate degradation, the following five assumptions were taken into account. First, the biofilm on the support is homogeneous. Second, the substrate concentration inside the biofilm varies only in the normal direction of the biofilm surface. Third, the substrate is transported from the bulk of the liquid to the biofilm by molecular diffusion. Fourth, the growth of the biofilm does not affect the flow pattern of liquid in the reactor. Fifth, the resistance to internal and external mass transfer are negligible.

The description of the mathematical model begins with the approach of biofilm substrate consumption rate based on Fick's law and its boundary conditions:

$$\frac{\partial S_f}{\partial t} = \mathcal{D}_f \frac{\partial^2 S_f}{\partial z_f^2} - \frac{kS_f}{K_s + S_f} * \mathrm{X_f} \tag{1}$$

$$IC:\ S_f|_{t=0} = S_{b0} \tag{2}$$

$$FC:\ \left.\frac{\partial S_f}{\partial z}\right|_{z=0} = 0 \tag{3}$$

$$LC:\ K\,(S_b - S_s) = \left. D_f \frac{\partial S_f}{\partial z_f}\right|_{z=Lf} \quad t > t_0 \tag{4}$$

where S_f is the substrate concentration in the biofilm (kg/m^3), D_f is the diffusion coefficient in the biofilm (m^2/d), k is the Monod maximum specific utilization rate (kg COD/kg VSS.d), K_s is the Monod half-velocity coefficient (kg COD/m^3), X_f is the density of the biofilm (kg VSS/m^3) and z_f is the radical distance in the biofilm (m).

The biofilm reactor was assumed as a fully mixed biofilm reactor. All biomass suspended in the liquid/biofilm interface is exposed to the same substrate concentration. The substrate mass balance and the biomass suspended in the biofilm reactor can be described according to the following equations.

Limiting substrate mass balance equation is:

$$\frac{dS_b}{dt} = \frac{Q}{V\varepsilon}(S_{b0} - S_b) - K_f * (\mathrm{S_b} - \mathrm{S_s}) * \frac{\mathrm{A}}{\mathrm{V}\varepsilon} - \frac{\mathrm{kS_b}}{\mathrm{K_s} + \mathrm{S_b}} * \mathrm{X_b} \tag{5}$$

$$IC:\ S_b|_{t=0} = S_{b0} \tag{6}$$

Biomass suspended mass balance equation is:

$$\frac{dX_b}{dt} = \left(\frac{Y * \mathrm{k} * \mathrm{S_b}}{K_s + S_b} - b - \frac{Q}{V\varepsilon}\right) * \mathrm{X_b} + \frac{\mathrm{A}}{\mathrm{V}\varepsilon} \mathrm{b_s L_f X_f} \tag{7}$$

$$IC:\ X_b|_{t=0} = 0 \tag{8}$$

where S_{b0} is the initial concentration in the feed (kg/m^3), X_b is the suspended biomass concentration in bulk liquid (kg VSS/m^3), X_{b0} is the suspended biomass concentration in the feed (kg VSS/m^3), Q is the flow rate of the feed substrate (m^3/d), V is the effective reactor volume (m^3), A is total surface area of the media (m^2) and ε is reactor porosity.

The biofilm uses the substrate as a carbon source for biosynthesis and respiration. The biofilm density is assumed constant, therefore, the biofilm volume and the thickness should increase with time according to its growth. The growth rate of the biofilm can be expressed as:

$$\frac{dL_f}{dt} = \int_0^{L_f} \left(\frac{Y * \mathrm{k} * \mathrm{S_f}}{K_s + S_f} - b - b_s\right) dz_f \tag{9}$$

$$IC:\ L_f|_{t=0} = L_{f0} \tag{10}$$

where L_f is biofilm thickness (m), Y the yield coefficient of the biomass (kg VSS/kg), b is the biomass decay coefficient (d^{-1}), b_s the biofilm shear-loss coefficient (d^{-1}) and L_{f0} is initial biofilm thickness (m).

MATERIALS AND METHODS

The biofilm methanogenic reactor was maintained at 25 ± 2 °C, fed with effluent from the hydrolytic/acidogenic reactor, with a hydraulic retention time (HRT) of 15 days and an organic loading of 4 kg COD/m^3 * d. The pH, volatile fatty acids (VFA), total solids (TS), volatile suspend solids (VSS), suspended solids (SV), chemical oxygen demand (COD) and biogas quality were evaluated. The variables validated in the model were the evolution of the substrate concentration (S_b), the evolution of the biomass concentration (X_b), the biofilm thickness (L_f), biogas formation (RCH_4) and the substrate flow in the biofilm. During the biofilm reactor run, VFA and biogas production were measured daily. The VSS, SV, TS and COD were measured twice in the week according to standard methods (APHA 1995). Measurements of pH were carried out with a general-purpose pH electrode (Hanna Instruments HI 8314). Biogas volumetric composition was determined using a Bacharach GA-94 *in situ* analyzer, which detects gases using an electrochemical cell and an infrared cell of dual wavelength. The VFA was measured according to a titration procedure described by Anderson & Yang (1992).

To solve mathematical model, firstly the biofilm mass balance equation was discretized for approximation by central differences, for a point i inside the biofilm. On the other hand, the use of Bode rule ($n = 4$) to calculate biofilm thickness was necessary. The mathematical model was supported for differential equations solved by POLYMATH 5.1 software using the STIFF method.

RESULTS AND DISCUSSION

The kinetic parameters obtained for the reactor are shown in Table 1. These kinetic parameters were calculated from experimental data and literature review. The values for Monod maximum utilization rate (k) and Monod half velocity coefficient (K_s) were 5.8 kg COD/kg VSS * d and 0.83 kg COD/m^3 for the reactor. The values of k are similar, in order of magnitude to those reported by Hsien & Lin (2005), Lin & Hsien (2009) for organic compounds degradation in biofilm reactors. In the same way, Nava *et al.* (2014) found K_s values similar in magnitude order for oil refineries wastewater treatment in attached film reactor; these values were greater than substrate concentration in the system, which showed low affinity of microorganisms for the substrate. In this work, the K_s values obtained were lower than the substrate concentration (see Table 1).

Table 1 | Biokinetic and reactor parameters used in the modelling biofilm reactor

Parameter	Symbol	Unit	Value
Feed concentration	S_{b0}	kg/m^3	60
Yield coefficient	Y	kg VSS/kg COD	0.0234
Monod maximum utilization rate	k	kg COD/kg VSS.d	5.8
Monod half velocity coefficient	K_s	kg COD/m^3	0.83
Decay coefficient	b	d^{-1}	0.0157
Shear loss coefficient	b_s	d^{-1}	0.095
Diffusion coefficient	D_f	m^2/d	0.000082
Film transfer coefficient	K_f	m/d	0.25
Biofilm density	X_f	kg VSS/m^3	0.00147
Initial concentration of suspend biomass	X_{b0}	kg VSS/m^3	0.3
Initial biofilm thickness	L_{f0}	m	0.00005
Reactor porosity	ε		0.7
Effective reactor volume	V	m^3	0.0015
Influent flow rate	Q	m^3/d	0.0001
Total surface area support	A	m^2	0.00236

To substrate concentration in the bulk of liquid (S_b), Figure 1 shows that model and experimental data have the same trend. The analysis of variance (ANOVA) (Statgraphics Centurion XV, 2013) showed no significant statistical difference ($P \geq 0.5$) between the results of the two curves. In the first stage of the curve until day 25, the substrate concentration increased until reaching 31 kg/m^3. In this period, there was no significant substrate degradation by the microorganisms. In the second stage from day 25 to day 80, the substrate concentration decreased drastically as a result of the activity of the microorganisms. In the third part of the curve from 80 days, the substrate concentration reached a steady state around 10 kg/m^3. The substrate removal efficiency was 83%. In the steady state, the experimental data were higher than model, as a result the substrate degrading bacteria grew enough to reach biofilm and that the effect of the shear stress was more significant as the biofilm increased in thickness. The rise in decay biofilm augmented suspended biomass. Suspended biomass decomposes and releases soluble microbial products which may slightly step up the substrate concentration in the effluent (Qi *et al.* 2008; Molobela & Ilunga 2012).

The suspended biomass concentration X_b, according to Figure 2, have the same trend for model and experimental data, although when the experimental data reached the

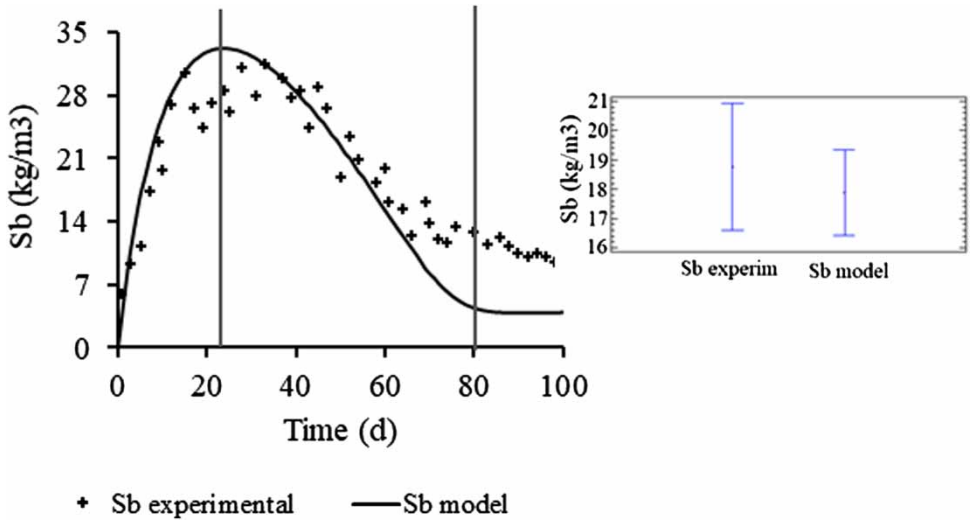

Figure 1 | Model and experimental results of the substrate concentration in the bulk of liquid (S_b). Correlation factor of 0.9401.

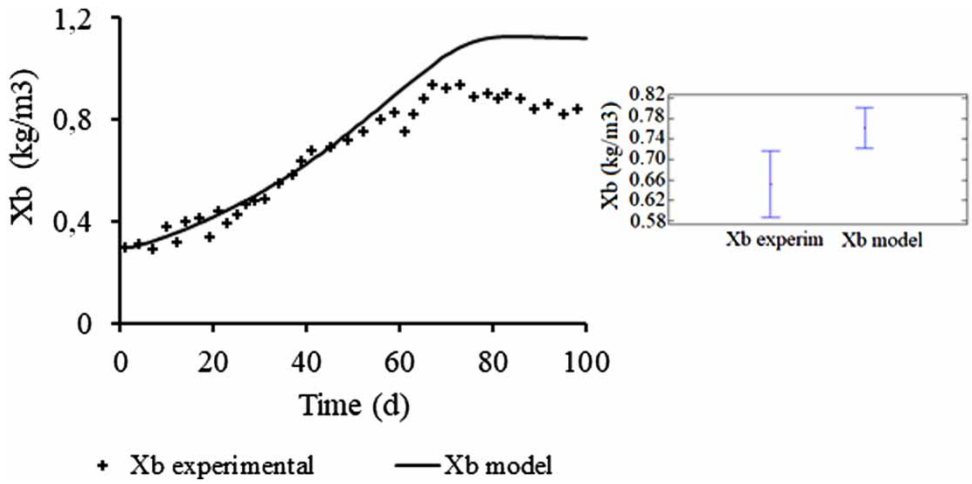

Figure 2 | Model and experimental results of the suspend biomass concentration (X_b). Correlation factor of 0.9695.

steady state, the concentration is lower than the model. The ANOVA analysis (Statgraphics Centurion XV, 2013) gave significant statistical difference ($P < 0.5$) between the experimental results and model. The experimental data showed a high utilization of the substrate in the first 70 days of experimentation. In the model, X_b reached a steady state at 1.12 kg/m^3, while to the experimental data, X_b reached a maximum concentration of 0.92 kg/m^3, but diminished in steady state to 0.84 kg/m^3. According to these results, both in the model and in the experimental data, suspended biomass concentration increased with the biomass contribution for shear stress ($b_s = 0.095\ d^{-1}$), which for the experimental data, should have been greater to achieve the maximum concentration given by the model.

In the model, it was observed that the decay coefficient (b) not influenced to X_b in the steady state, since this concentration did not decline. The X_b values in the experimental data before reach the steady state diminished, this can be attributed in part to the influence of decay coefficient, b (0.0157 kg/m^3). In steady state, X_b reached values of 1.12 kg/m^3 and 0.84 kg/m^3 to model and experimental data, respectably. According to these data, it is observed that in the experimental results, the release of active biomass was lower than its contribution to the model. In the reactor, cell death can occur in the biofilm, which is released and causes decrease in X_b. The biomass suspended concentration in the model and experimental data is lower than the value of 3.2 kg/m^3 reported by Leyva-Díaz *et al.* (2013) in the modeling wastewater treatment in a bed reactor.

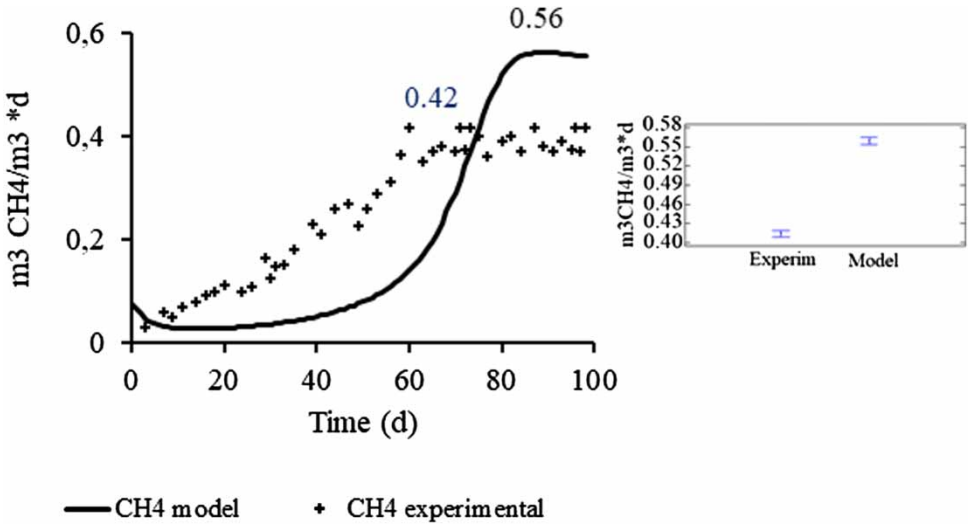

Figure 3 | Model and experimental results of methane formation (RCH_4). Correlation factor of 0.7801.

In Figure 3, the model in steady state reached a maximum of 0.56 m^3 CH_4/m^3 * d and the experimental data 0.42 m^3 CH_4/m^3 * d. The methane values are higher in the model than experimental data. Although detachment coefficients were included in the model, other factors were not taken into account; such as: inhibition by pH, substrate or methanogenesis that affect methane production. The presence of some of these inhibitions during the experimental development could have influenced the reduction in methane production (Chen *et al.* 2008; Zhai *et al.* 2015). The difference between the values of the model and the experimental data could be due that the observed reaction rates are usually lower than the rates predicted by the reaction kinetics. The available substrate concentration to the microorganisms is lower than the liquid bulk substrate concentration, the consequence of control exerted by molecular diffusion on the penetration of the substrate into the biofilm (Sun *et al.* 2014).

The predicted biofilm thickness increased until stable value of 14 μm (Figure 4). According to these results, in the initial stage the substrate removal was low due to the emerging formation of the biofilm and the removal would be done by the suspended microorganisms from the inoculum. The biomass grew until the biofilm was developed. The initial stage is very sensitive, unstable and inefficient process for organic removal. In this initial stage there is a complex relationship between acidogenic, acetogenic and methanogenic microorganisms.

According to Rittmann & McCarty (2001), the biofilm thickness is determined by substrate flow inside biofilm from bulk liquid, growth rates and detachment of bacteria

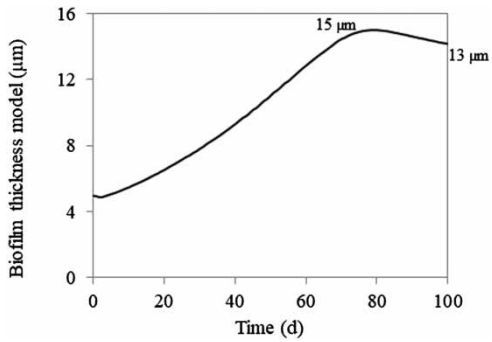

Figure 4 | Model results of biofilm growth.

in the biofilm. In this way, the mass in the biofilm is greater when the substrate concentration in the liquid is high; this is in agreement with the model. In addition, the biofilm is negatively affected by high shear stress that increase the detachment.

The values of biofilm thickness obtained by the model were between 5 to 14 μm. Langer *et al.* (2014) measured biofilm thickness in reactors with polypropylene support for the treatment of wastewater with organic loads of 45 kg SV/m^3 and 15 kg SV/m^3. The thickness of the reactor samples with high organic load varied from 5 to 160 μm, while for the low organic load microcolonies were observed instead of biofilm.

CONCLUSIONS

In the mathematical model, the values obtained for maximum utilization (k) and Monod half velocity coefficient

(K_s) were comparable (same order of magnitude) with those obtained by other researchers who used biofilm reactors. The mathematical model was validated by the results in the case of substrate concentration (Sb) for the reactor using ceramic brick as a support for cellular immobilization. In the steady state, the experimental data were higher than in the model, as a result the substrate degrading bacteria grew enough to reach biofilm and that the effect of the shear stress was more significant as the biofilm increased in thickness. The substrate removal efficiency was 83%.

The predictions of the model for active biomass concentration (Xb) and methane production have similar trends to experimental data with a variation in the steady state values. To Xb, this variation can be attributed in part to the influence of decay coefficient. To methane production, the difference between the values of the model and the experimental data could be because the observed reaction rates are usually lower than the rates predicted by the reaction kinetics. According to the model, the biofilm thickness reached 14 μm. The increase in the biomass concentration was allowing the biofilm development.

The development of this mathematical model allowed to establish a practical monitoring of the behavior of a biofilm reactor, in spite of not having some parameters in the model that could provide more specific information. This model gives the possibility of stopping the process according to the response of generated biogas volume by the system, the profile of substrate concentration and the biofilm profile with the time of operation.

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