EVALUATION OF A MECHANISTIC MATHEMATICAL MODEL OF A PACKED-BED ANAEROBIC REACTOR TREATING WASTEWATER

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***Abstract* - A mechanistic mathematical model is proposed and evaluated for simulating the performance of a bench-scale packed-bed anaerobic reactor that uses polyurethane foam as biomass support. The model developed under rational criteria was based on the study of mass transfer and biochemical kinetics, also considering the hydrodynamic characteristics of the reactor. The data generated by the model adhered well to the experimental data obtained from the operation of the reactor applied to the treatment of a glucose-based substrate. The liquid-phase mass transfer coefficient was found to be the main parameter in the model, and its precise estimation is essential for the model to be successfully applied. Additionally, a case study was used in order to verify the applicability of the model for designing full-scale reactors. The simulations performed permitted to demonstrate the importance of the choice of convenient liquid superficial velocity and polyurethane foam matrix size which have direct influence on the solid- and liquid-phase mass transfers resistance and, consequently on the volume of the designed reactor.**

Keyword* - Wastewater treatment, anaerobic process, packed-*bed reactor, HAIB reactor, performance simulation

1. INTRODUCTION

Most of the anaerobic reactor configurations containing immobilized sludge have been developed essentially based on empirical criteria. The predominance of empirical over rational criterion arises as a consequence of the variety and complexity of interactive processes occurring in such heterogeneous units.

According to Wentzel and Ekama (1997), two extremes in mathematical models can be identified: empirical and mechanistic. The mechanisms and processes are ignored in the empirical ("black box") approach, while the mechanistic models are based on some fundamental phenomena. Both approaches can be useful for design and simulation purposes, but the mechanistic one provides more reliable bases for optimization than the empirical approach. On the other hand, mechanistic models demand wide knowledge about chemical, physical and biological aspects of the system. Such knowledge is not always available and, sometimes, the parameters required cannot be estimated.

Mechanistic models can be very complicated, especially in anaerobic digestion, due to the many microbial interactions, such as inhibition by intermediary products formed and competition for substrate. In this case, a complex model can be used for better understanding the system, but its use for simulation can be limited. Such limitation derives from the difficulty of estimating all parameters involved. Moreover, the mechanistic model derived is frequently too complex and difficult to be solved. The proposition of a mechanistic model presupposes the existence of reliable methods for estimating its parameters. Otherwise, even a well conceived model would be useless, if the required parameters were not accessible.

This work aims to supply elements for modeling a packed-bed anaerobic reactor, the Horizontal-Flow Anaerobic Immobilized Biomass (HAIB) reactor, under rational criteria. A simple mechanistic mathematical model was tested to simulate the performance of a lab-scale bioreactor used to treat a glucose-based synthetic substrate. Such a model was based on the study of phenomena as mass transfer, biochemical kinetics and hydrodynamic characteristics.

2. EXPERIMENTAL EQUIPMENT

A 2-liters horizontal-flow anaerobic immobilized biomass (HAIB) reactor was composed by a glass tube of 1.0 m with 0.05 cm diameter and length to diameter ratio (L/D) of 20 (Fig. 1). A perforated tube (0.9 cm of diameter) for gas collection was installed along the reactor in its upper part. Intermediate sampling ports were allocated along the reactor at L/D of 4, 8, 12 and 16.

A performance test was carried out in a previous work (Zaiat *et al.*, 1997a) using a synthetic substrate composed of glucose as the main carbon source (chemical oxygen demand-2090 mg COD.l^{-1}). The hydraulic detention time (θ_h) was 8 hours and temperature was controlled at 30°C. The reactor was filled with polyurethane foam cubic particles, previously inoculated with anaerobic biomass. Data of chemical oxygen demand (COD) along the reactor

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length were obtained during the operation in steady-state regime.

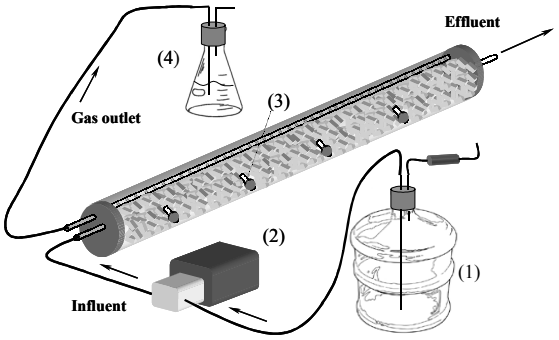

Figure 1. Scheme of the bench-scale HAIB reactor. (1) Substrate reservoir, (2) Peristaltic pump, (3) Sampling ports, (4) Hydraulic seal

3. MATHEMATICAL MODEL

A classical mathematical model is proposed to simulate the steady-state operation of the horizontal-flow anaerobic immobilized biomass reactor. Such a model was developed to describe the HAIB reactor behavior in a simplified way.

The development of Horizontal-Flow Anaerobic Immobilized Biomass (HAIB) Reactors consisted several experiments allowing evaluating fundamental parameters. The experimental planning involved the following stages: methodology for anaerobic biomass immobilization in polyurethane foam; performance evaluation of the HAIB reactor in the treatment of recycled paper industry wastewater; studies on cell wash out from polyurethane foam; studies on intraparticle mass transfer; studies on liquid-phase mass transfer and kinetic studies (Zaiat *et al*., 1997b).

The mathematical model proposed for performance simulation and designing of HAIB reactor is based on kinetic and mass transfer parameters. The flow pattern is considered to be properly described by a plug-flow model. This assumption is based on previous hydrodynamic studies carried out by de Nardi *et al*. (1999). Substrate consumption is considered to follow a first order kinetics, based in previous kinetic studies (Vieira *et al*., 1997).

According to these assumptions and assuming isothermal condition, the substrate mass balance under steady state resulted in:

$$E_{COD} = \left\{1 - \exp\left[\frac{-(L/D).k_1.\eta.D.\overline{X}}{\varepsilon.v_s}\right]\right\}.100 \qquad (1)$$

In the Eq. (1), E_{COD} is the chemical oxygen demand (COD) removal efficiency; L, the length of the reactor; D, diameter of the reactor; k_1, the first order intrinsic kinetic constant; $\overline{X}$, the mean biomass concentration within the reactor on its useful volume; η, the overall effectiveness factor; ε, bed porosity and v_s is liquid superficial velocity.

The overall effectiveness factor for first order kinetics was calculated by the classical expression obtained from a mass balance in a particle's shell (Bailey and Ollis, 1986):

$$\eta = \frac{1}{\phi}\left(\frac{1}{\mathrm{tgh}(3.\phi)} - \frac{1}{3.\phi}\right)\left\{\frac{Bi}{\left[3.\phi / \mathrm{tgh}(3.\phi)\right] + Bi - 1}\right\} \qquad (2)$$

In the Eq. (2), Bi is the Biot number and ϕ is the Thiele modulus, which are defined as:

$$Bi = \frac{k_s.R_p}{D_e} \qquad (3)$$

$$\phi = \left(\frac{R_p}{3}\right)\sqrt{\frac{k_1.\overline{X}}{D_e}} \qquad (4)$$

In Eqs. (3) and (4), k_s is the liquid-phase mass transfer coefficient; D_e, the substrate effective diffusivity in the bioparticle; R_p, the equivalent particle radius of the bioparticle; k_1, the intrinsic first order kinetic parameter and $\overline{X}$ is the mean biomass concentration based on the useful volume of the reactor.

Application of such a mechanistic model aims to predict the COD removal efficiency profile along the reactor length. Initially, several parameters must be provisioned to the model: reactor diameter (D), diameter of the tube for gas collection (D_{TS}), liquid flow-rate (Q), the first order intrinsic kinetic parameter (k_1), effective diffusion of substrate in the particle (D_e), equivalent particle radius (R_p) and bed porosity (ε). Viscosity and density of the liquid should be known when some relationships of literature were used to estimate liquid-phase mass transfer coefficient (k_s).

The liquid-phase mass transfer coefficient (k_s) is an important parameter in simulation, since mass transfer resistance can be the limiting step of overall conversion rate. According to Zaiat *et al.* (2000a), overall reaction rate in a bench-scale HAIB reactor was mainly affected by liquid-phase mass transfer. So, it was expected the liquid-phase mass transfer coefficient would be the key parameter in the model. By this way, this parameter deserved special attention and three relationships were chosen to be applied in the model:

(i) Relationship presented by Zaiat *et al*. (1996a)

Liquid-phase mass transfer coefficient (k_s) was calculated as a function of bed porosity (ε), equivalent particle radius (R_p) and liquid superficial velocity (v_s):

$$k_s = \frac{\varepsilon.R_p}{3.(1-\varepsilon)}\left[-0.244 + 0.271.e^{(1.796.v_s)}\right] \qquad (5)$$

Equation (5) was obtained from experiments in differential reactor containing polyurethane foam particles.

(ii) Relationship presented by Sarti *et al.* (2001)

The k_s relationship from Sarti *et al.* (2001) was obtained in a fixed-bed reactor treating synthetic substrate simulating domestic sewage and subjected to liquid superficial velocities between 0.003 and 0.0139 $cm.s^{-1}$

$$k_s = 0.033.e^{0.0217.v_s} \quad (6)$$

In this expression, k_s and v_s are expressed in $cm.h^{-1}$.

(iii) Relationship presented by Perry and Chilton (1985)

This is the most complete correlation, since mass transfer coefficient is related to liquid properties, as:

$$J_D = (0{,}81 \pm 0{,}05).(Re_p)^{-0.50} \quad (7)$$

In the Eq. (7), J_D is the Colburn factor for mass transfer and Re_p is the particle Reynolds number based on the cross-section area of the reactor, defined as:

$$Re_p = \frac{v_s.\rho_L.d_p}{\mu_L} \quad (8)$$

$$J_D = Sh.(Sc)^{-1/3}.(Re_p)^{-1} \quad (9)$$

In the Eq. (9), Sh is Sherwood number, which relates mass transfer by convection with mass transfer by diffusion in liquid-phase; Sc is the Schmidt number that relates the momentum with mass transfer. Such parameters are defined as:

$$Sh = \frac{k_s.d_p}{D_L} \quad (10)$$

$$Sc = \frac{\mu_L}{\rho_L.D_L} \quad (11)$$

In the Eqs. (8), (10) and (11), d_p is bioparticle diameter, D_L, substrate diffusion in liquid medium, μ_L, liquid viscosity, ρ_L, liquid density; k_s, liquid-phase mass transfer coefficient and v_s is the liquid superficial velocity in the bed, which was calculated through the expression:

$$v_s = \frac{Q}{\varepsilon.A} \quad (12)$$

In Eq. (12), Q is liquid-flow rate, ε is the bed porosity and A is the cross-section area of the reactor.

4. RESULTS AND DISCUSSION

The mathematical model was applied to simulate the COD removal efficiencies along the bench-scale horizontal anaerobic reactor treating a glucose-based synthetic substrate. Fundamental and physical parameters used for HAIB reactor simulation are presented in Table 1. Effective diffusivity (D_e) was estimated by Vela *et al*. (1999) in finite-batch assays, using an orbital shaker. A Brookfield viscosimeter and a densimeter were used to estimate liquid viscosity (μ_L) and density (ρ_L), respectively.

Experimental efficiencies obtained by Zaiat *et al*. (1997a) and the theoretical ones, obtained by the proposed model, are presented in Table 2. The simulation was performed using three different relationships for k_s prediction. The model adjustment to experimental points is presented in Figs. 2 and 3.

Values of E_{COD} predicted by the mathematical model are extremely low when the Eq. (5) is used for k_s prediction, leading to underestimation of the effectiveness factor (η). The use of such expression for HAIB reactor design would result in overestimated units. For example, the model predicts that it would be necessary a 21 liters reactor to achieve E_{COD} of 98% and 84 hours of hydraulic detention time.

Contrarily, the use of the Eq. (7) for k_s estimation resulted in E_{COD} values higher than the one observed experimentally, since it led to a 192% overestimation of the effectiveness factor (η). The model predicts that the substrate will be completely consumed at L/D of 8 and that application of a hydraulic detention time of 2.8 hours would be sufficient to attain the E_{COD} of 98%. Therefore, the use of such a model for designing HAIB reactors operated at low values of v_s would result in underestimated units.

Good adjustment of the values generated by the mathematical model can be observed when the relationship (6) is used to estimate liquid-phase mass transfer coefficient (k_s). Such a correlation was obtained under conditions of low liquid superficial velocities, as effectively applied to the horizontal reactor. This fact can explain the better adjustment obtained when Eq. (6) is used.

The data obtained from the application of the mathematical model indicate that the liquid-phase mass transfer coefficient is a key parameter when low liquid superficial velocities are applied. Most of the expressions available for k_s prediction are reliable only for high v_s values. So, the performance simulation and design of immobilized cell reactors under low v_s conditions must be performed with caution, since high deviations can be observed by using an inadequate correlation.

Equation (6) obtained by Sarti *et al.* (2001) for k_s prediction was found to provide good results. However, such a relationship is recommended to be used for liquid superficial velocities lower than 0.014 $cm.s^{-1}$, as affirmed by the authors.

Table 1: Parameters used for HAIB reactor simulation applied to glucose-based substrate treatment at 30°C.

Parameter	Value	Source
R_p	0.31 cm	Experimental condition
v_s	0.0035 cm.s^{-1}	Operating condition
ε	0.4	Experimental condition
θ_h	8 h	Operating condition
k_1	7.12 x 10^{-5} *l*.mg VSS^{-1}.h^{-1} *	Vieira *et al*. (1997)
D_e	0.027 cm^2.h^{-1}	Vela *et al*. (1999)
D_L	0.027 cm^2.h^{-1}	Perry and Chilton (1985)
μ_L	0.008 poise	Experimentally obtained
ρ_L	1.19 g.m*l*$^{-1}$	Experimentally obtained

*VSS = Volatile Suspended Solids

Table 2: Experimental and generated by the theoretical model values of COD concentration in bulk liquid (S_b) and COD removal efficiency (E_{COD}) along HAIB reactor treating glucose-based synthetic substrate at 30°C.

L/D	$S_{b\ exp.}$ (mg COD.*l*$^{-1}$)	S_b* (mg COD.*l*$^{-1}$)	S_b** (mg COD.*l*$^{-1}$)	S_b*** (mg COD.*l*$^{-1}$)	$E_{COD\ exp.}$ (%)	E_{COD}* (%)	E_{COD}** (%)	E_{COD}*** (%)
0	2090	2090	2090	2090	0	0	0	0
4	582	1939	690	217	72	7	67	90
8	206	1799	230	23	90	14	89	99
12	108	1670	85	0	95	20	96	100
16	55	1548	21	0	97	26	99	100
20	41	1437	0	0	98	31	100	100

*Generated values using the Eq. (5) for k_s estimation.
* *Generated values using the Eq. (6) for k_s estimation.
* * *Generated values using the Eq. (7) for k_s estimation.

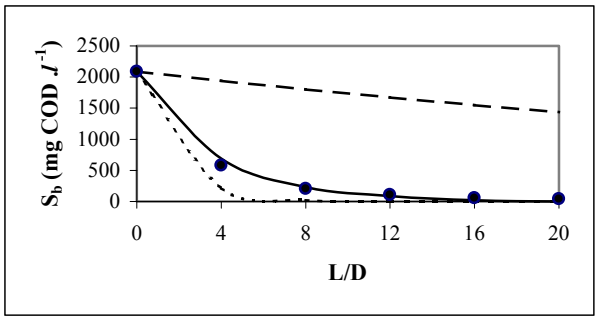

Figure 2: Substrate concentration in the bulk liquid (S_b) along the HAIB reactor treating synthetic substrate (•) and values generated by the theoretical model using relationships 5 (— —), 6 (—) and 7 (---) for k_s estimation.

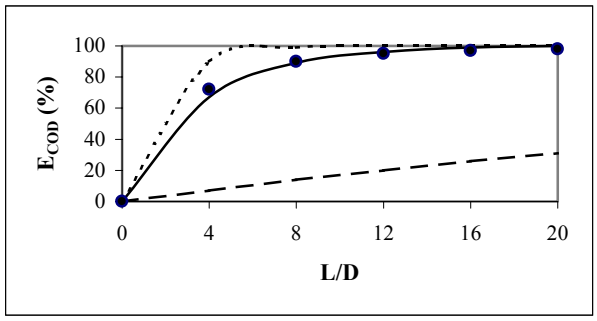

Figure 3: COD removal efficiency along the HAIB reactor treating synthetic substrate (•) and values generated by the theoretical model using relationships 5 (— —), 6 (—) and 7 (---) for k_s estimation.

Probably the relationship presented by Perry and Chilton (1985) would provide a more reliable k_s data when high liquid superficial velocities are applied. Such operating condition is preferable in design procedures, since high v_s implies high k_s, thus improving organic matter transfer to the bioparticles and thus the conversion rates. However, the application of high superficial velocities will be limited by critical v_s, above

which biomass is washed out. According to Zaiat *et al*. (1996b), liquid superficial velocities above 1.5 $cm.s^{-1}$ are critical for anaerobic mixed culture wash out from polyurethane foam matrices.

The proposed mathematical model was evaluated previously for designing a pilot-scale HAIB reactor applied to domestic sewage treatment (Zaiat *et al*., 2000b). Relationship for k_s prediction (7) proposed by Perry and Chilton (1985) was used in the model for a liquid superficial velocity of 0.1 $cm.s^{-1}$. Verification of the mathematical model was carried-out by monitoring organic matter concentration, expressed as COD, along the reactor's length. The model was found to be suitable though some deviations between experimental and theoretical data were observed.

In this work, the model is applied to design a reactor to be applied to treat recycled paper industry wastewater. Dimensions and characteristics of the designed HAIB reactor could be compared to an up-flow anaerobic sludge blanket (UASB) reactor of 600 m^3 installed in the industry. Such a reactor operates with COD removal efficiency ranging from 80% to 85%

Average initial substrate concentration after primary treatment was 3,000 mg $COD.l^{-1}$ and the mean liquid flow-rate, 26 $m^3.h^{-1}$. Average temperature was assumed to be around 25°C and the substrate effective diffusivity (D_e) was considered as 0.48 x 10^{-5} $cm.s^{-2}$. This value was obtained from Perry and Chilton (1985) for organic compounds diffusion in water with molecular weight ranging from 100 to 1000. Wastewater viscosity was admitted as being the same as water at 25°C and wastewater density of 1.19 $g.cm^{-3}$ was determined experimentally. Bed porosity was considered to be 0.4 and it was chosen, initially, polyurethane foam cubic matrices with side of 1 cm, thus resulting in equivalent sphere radius of 0.62 cm. The experimental first order intrinsic kinetic parameter (k_1) was estimated as 1.26 x 10^{-5} $l.mg^{-1}$ $SSV.h^{-1}$ using the proper wastewater and sludge taken from the up-flow anaerobic sludge blanket (UASB) reactor operating in the industry (Vieira, 1996).

Initially, v_s value was arbitrarily fixed in 0.5 $cm.s^{-1}$ and k_s value was estimated using the relationship (7) presented by Perry and Chilton (1985), since this correlation was used previously by Zaiat *et al*. (2000b) for designing a pilot-scale reactor, thus providing suitable results for v_s of 0.1 $cm.s^{-1}$.

Relationship between the diameter of the perforated tube for gas collection and diameter of the reactor (D_{TS}/D) in the bench-scale HAIB reactor was of 0.19. It is convenient, therefore, that values between 0.1 and 0.2 be fixed for D_{TS}/D ratio. The smallest D_{TS}/D value will reduce total volume of the reactor due to smallest available volume for gas separation. An intermediate value of 0.15 was adopted in this study.

According to the design procedures, a HAIB reactor with total volume of 397 m^3 and hydraulic detention time of 5.53 hours would be necessary to obtain the COD removal efficiency of 85%. The model predicted a reactor diameter of 2.25 m and 99.57-m length (L/D of 44) and the tube for gas collection would be 0.34-cm diameter.

Thus, HAIB reactor would be 1.5 times minor than the UASB reactor installed in the industry for obtaining the same COD removal efficiency. Moreover, applied hydraulic detention time would be smaller. However, it must be emphasized that the designed reactor was not tested and the study presented is just a preliminary evaluation of the proposed model. The diameter of the reactor and the diameter of the perforated tube for gas collection values should be changed to values commercially available. Length of the tube obtained in the design can complicate cleaning operations and bed maintenance. Configuration using several modules in series can minimize such problems.

Diameter of the designed reactor can be reduced as liquid superficial velocity is increased. For example, if v_s of 1 $cm.s^{-1}$ were chosen instead of 0.5 $cm.s^{-1}$, the total volume and diameter of HAIB reactor would be of 394 m^3 and 1.6 m, respectively.

As observed previously by Zaiat *et al*. (2000b), the smaller reactor volume would be obtained by utilization of smaller particles, since the overall reaction rate increases as the size of bioparticles decreases. However, small bioparticles can result in high-pressure drop in the bed.

That study demonstrates the importance of convenient choices of v_s and particle size, both having direct influence on the intraparticle and liquid-phase mass transfer resistances and, consequently, on the overall effectiveness factor. For example, a 397-m^3 reactor is suitable to provide COD removal efficiency of 85% when filled with polyurethane foam cubic matrices 1-cm side and operated at v_s of 0.5 $cm.s^{-1}$. On the other hand, the same efficiency would be obtained with a 796-m^3 reactor if the particle size is changed to 3 cm, maintaining the same v_s.

5. CONCLUSIONS

- The mathematical model based on hydrodynamic characteristics, kinetic parameters and mass transfer fluxes was found to predict adequately HAIB reactor performance. The model data fitted well plotted experimental data of a HAIB bench-scale reactor treating a glucose-based substrate.
- The success of model application in bench-scale units was found to be directly related to the choice of an adequate relationship for k_s prediction.
- Liquid-phase mass transfer plays an important role in global performance of bench-scale system whereas the solid-phase mass transfer is the limiting step of the overall conversion rate in full-scale system.
- The model presented in this work is simple and its application is dependent on the precise evaluation of three fundamental parameters only: liquid-phase mass transfer coefficient (k_s), substrate effective

diffusivity (D_e) and intrinsic kinetic parameter (k_1). The first two parameters can be adopted or estimated using several relationships, while the intrinsic kinetic parameter should be estimated for each specific case, using methods not always available easily.

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Nomenclature

A	cross-section area, $[L]^2$
Bi	Biot number
D	diameter of the reactor, [L]
D_e	effective diffusivity, $[L]^2.[T]^{-1}$
D_L	substrate diffusion in the liquid medium, $[L]^2.[T]^{-1}$
D_{TS}	diameter of the perforated tube for gas collection [L]
d_p	bioparticle diameter,[L]
E_{COD}	COD removal efficiency, [%]
k_1	first order intrinsic kinetic parameter, $[T]^{-1}$
k_s	liquid-phase mass transfer coefficient, $[L].[T]^{-1}$
L	length of the reactor, [L]
Q	liquid flow-rate, $[L]^3.[T]^{-1}$
R_p	equivalent particle radius, [L]
S_b	substrate concentration in the bulk liquid, $[M].[L]^{-3}$
v_s	liquid superficial velocity, $[L].[T]^{-1}$
$\overline{X}$	mean biomass concentration, $[M].[L]^{-3}$
θ_h	hydraulic detention time, [T]
ε	bed porosity
ϕ	Thiele modulus
η	effectiveness factor
μ_L	liquid viscosity, $[M].[L]^{-1}.[T]^{-1}$
ρ_L	liquid density, $[M].[L]^{-3}$

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