Optimization strategy to maximize the hydrogen production in a dark fermenter

Ixbalank Torres [*,**] **Alejandro Vargas** [**] **Germán Buitrón** [**]

[*] *División de Ingenierías - Universidad de Guanajuato, campus Irapuato-Salamanca. Carretera Salamanca - Valle de Santiago Km. 3.5 + 1.8, Comunidad de Palo Blanco, Salamanca, México*
[**] *Laboratorio de Investigación en Procesos Avanzados de Tratamiento de Aguas, Unidad Académica Juriquilla del Instituto de Ingeniería - Universidad Nacional Autónoma de México. Blvd. Juriquilla No. 3001, 76230 Querétaro, México (email: ixbalank@gmail.com, avargasc@ii.unam.mx, gbuitronm@ii.unam.mx)*

Abstract: Biohydrogen production represents a real alternative to the energetic problem, indeed, the 21^{st} century is called the century of the hydrogen. However, once a biohydrogen production process has been developed, it must be optimized in order to maximize the resulting production rate. In this context, we propose a simple heuristic optimization strategy based on the relation between the organic loading rate and the hydrogen production rate. A nonlinear optimization problem is first solved to maximize the hydrogen production rate by optimizing the flow rate at the reactor input. Since the organic loading rate depends on both, the flow rate and the substrate at the reactor input, a robust observer is used to estimate this last concentration. Finally, an anti-windup super-twisting controller tracks the maximum productivity computed by controlling the flow rate at the reactor input. Simulations demonstrate the feasibility of this strategy for future implementation in a real process.

Keywords: Biohydrogen production, model-based optimization, Luenberger observer, super-twisting algorithm.

1. INTRODUCTION

Biological production of hydrogen (biohydrogen), using (micro) organisms, is an area of technology development that offers the potential production of usable hydrogen from a variety of renewable resources. Biological systems provide a wide range of approaches to generate hydrogen, and include direct biophotolysis, indirect biophotolysis, photo-fermentations, and dark-fermentation (Levin et al., 2004).

Once a biological process to produce hydrogen has been developed, the operational conditions have to be optimized in order to achieve a desirable process performance.

In the last decade, optimization methods have been developed in order maximize the hydrogen production into fermenter bioreactors. For example, Aceves-Lara et al. (2010) apply model predictive control (MPC) to optimize the hydrogen production in continuous anaerobic digesters using the influent flow rate as the main control variable. Huang et al. (2012) apply fuzzy control-based real-time optimization of pH and temperature to achieve the best growing environment and hydrogen production rate control as well as enhance hydrogen production into a dark fermentation reactor.

This article presents an improved version of the heuristic optimization strategy to maximize on-line the hydrogen production into a dark fermenter proposed by Ramírez et al. (2015). In such work, a non-linear programming (NLP) optimization problem was formulated, considering the relation between the hydrogen production rate (HPR) and the organic loading rate (OLR) as objective function, and the flow rate at the reactor input as optimization variable. Since the OLR depends on both, the flow rate and the substrate at the reactor input, this last concentration was maintained constant.

Nevertheless, in real applications the substrate concentration does not remain constant along the bioreactor operation. Hence, an observer is used in this work to estimate the substrate concentration and estimate therefore the OLR. This OLR estimation allows us to solve the NLP problem and compute the maximum HPR. Furthermore, a super-twisting controller is used to track this maximum HRP (considering the optimal flow rate as initial condition).

The biohydrogen production process considered in this work, depicted in Figure 1, has two inputs: the substrate concentration (an uncontrolled input) and the flow rate (a controlled input) at the reactor input. On the other hand, the total gas flow rate and the hydrogen fraction at the reactor output are the

Reserva de Derechos No. EN TRÁMITE, ISSN. EN TRÁMITE

measured outputs. Using these measurements, the produced hydrogen flow rate can be calculated.

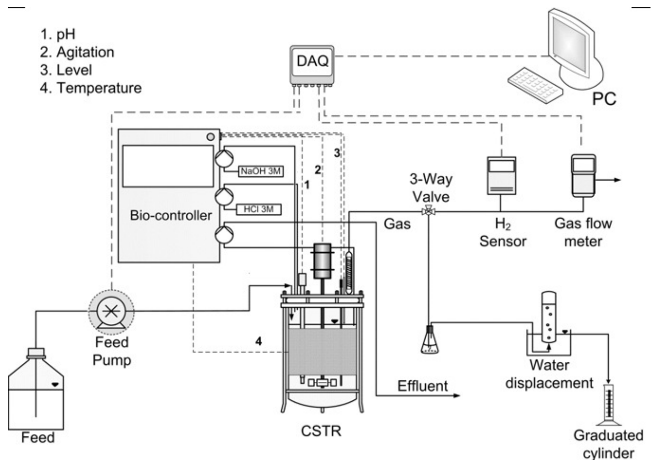

Fig. 1. Biohydrogen production process scheme.

The rest of the article is organized as follows: in Section 2 the model of the hydrogen production bioreactor is presented. In Section 3 the optimization strategy proposed is explained in detail. Section 4 is devoted to simulate the optimization strategy and to discuss the results obtained. In section 5 some conclusions about the work presented in this article are stated.

2. MODEL OF THE BIOREACTOR

The anaerobic hydrogen production reactor considered in this work is modeled, as proposed by Aceves-Lara et al. (2008) and Torres Zúñiga et al. (2015), by the following set of ordinary differential equations (ODE):

$$\begin{bmatrix} \dot{Glu} \\ \dot{Ace} \\ \dot{Pro} \\ \dot{Bu} \\ Et\dot{O}H \\ \dot{X} \\ \dot{CO_2} \\ \dot{H_2} \end{bmatrix} = Kr - D \begin{bmatrix} Glu - Glu_{in} \\ Ace \\ Pro \\ Bu \\ EtOH \\ X \\ CO_2 \\ H_2 \end{bmatrix} - \begin{bmatrix} 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ q_{CO_2,gas} \\ q_{H_2,gas} \end{bmatrix} \qquad (1)$$

where Glu, Ace, Pro, Bu, $EtOH$, X, CO_2 and H_2 represent the concentrations in gL^{-1} of glucose, acetate, propionate, butyrate, ethanol, biomass, carbon dioxide and hydrogen, respectively, in the liquid phase. The vector r describes the kinetics of the involved biological reactions (in $gL^{-1}d^{-1}$), D is the dilution rate (d^{-1}) and $q_{CO_2,gas}$ and $q_{H_2,gas}$ the gas flow rates of carbon dioxide and hydrogen expressed in $gL^{-1}d^{-1}$, respectively. Finally, $K \in \mathbb{R}^{8\times 2}$ represents the matrix of pseudo-stoichiometric coefficients.

The reaction pathway is described by two reactions occurring in parallel. Thus, the vector r is composed of the specific glucose uptake rate multiplied by the biomass concentration in the reactor:

$$r = \begin{bmatrix} \dfrac{\mu_{max1} Glu}{K_{Glu1} + Glu} \\ \dfrac{\mu_{max2} Glu}{K_{Glu2} + Glu} \end{bmatrix} X$$

where $\mu_{max,l}$ is the maximum specific growth rate of the microorganisms in $g[Glu]g[X]^{-1}d^{-1}$ and $K_{Glu,l}$ is the half-saturation constant in gL^{-1}.

Furthermore, the differential equations for the gas phase with constant gas volume are:

$$\frac{dCO_{2,gas}}{dt} = -\frac{CO_{2,gas}Q_{gas}}{V_{gas}} + \rho_{CO_2}\frac{V}{V_{gas}} \qquad (2)$$

$$\frac{dH_{2,gas}}{dt} = -\frac{H_{2,gas}Q_{gas}}{V_{gas}} + \rho_{H_2}\frac{V}{V_{gas}} \qquad (3)$$

with:

$$Q_{gas} = \frac{RT_{amb}}{P_{atm} - p_{vap,H_2O}} V \left(\frac{\rho_{H_2}}{M_{H_2}} + \rho_{CO_2}\right) \qquad (4)$$

$$\rho_{H_2} = k_L a_{H_2}(H_2 - M_{H_2}K_{H,H_2}p_{H_2,gas}) \qquad (5)$$

$$p_{H_2,gas} = \frac{H_{2,gas}RT_{reac}}{M_{H_2}} \qquad (6)$$

$$\rho_{CO_2} = k_L a_{CO_2}(CO_2 - K_{H,CO_2}p_{CO_2,gas}) \qquad (7)$$

$$p_{CO_2,gas} = CO_{2,gas}RT_{reac} \qquad (8)$$

where $CO_{2,gas}$ and $H_{2,gas}$ are, respectively, the carbon dioxide concentration, in $molL^{-1}$, and the hydrogen concentration, in gL^{-1}, in the gas phase.

As shown in equation (4), the total gas flow at the reactor output is the sum of the hydrogen gas flow plus the carbon dioxide gas flow. The carbon dioxide and the hydrogen gas flow rates are calculated by considering the transfer of the gas out from the liquid phase to the gas phase. The carbon dioxide and the hydrogen concentrations at the liquid-gas interface in equilibrium are calculated by considering the Henry law. The pressure of each gas component can be calculated using the ideal gas law for the two gases.

The physico-chemical, pseudo-stoichiometric and kinetic parameters used in this work are defined in (Torres Zúñiga et al., 2015).

3. OPTIMIZATION STRATEGY

3.1 Optimization problem

Ramírez et al. (2015) propose an heuristic optimization strategy to maximize the hydrogen production in a dark fermenter by considering the effect of the organic loading rate (OLR) on the hydrogen production rate (HPR). In order to describe the effect of the OLR on the HPR, a model was proposed as:

$$HPR = a_3 OLR^3 + a_2 OLR^2 \tag{9}$$

On the other hand, the OLR is defined as:

$$OLR = \frac{Q_{in} Glu_{in}}{V} \tag{10}$$

In order to maximize the HPR, the following non-linear programing (NLP) problem was then proposed:

$$\begin{aligned} &\max_{Q_{in}} HPR(OLR(Q_{in}, Glu_{in})) \\ &\text{such that:} \\ &HRT_{min} \leq HRT \leq HRT_{max} \end{aligned} \tag{11}$$

where HRT represents the hydraulic retention time. As can be regarded in equation (11), the equation (9) is used as objective function. Furthermore, the flow rate at the reactor input (Q_{in}) was selected as the optimization variable.

In order to compute the OLR, the substrate at the reactor input (Glu_{in}) must be provided. Since measuring Glu_{in} on-line is not practical, Torres Zúñiga et al. (2015) proposed a robust observer to estimate it.

3.2 Estimation of the substrate at the reactor input

Let the state vector $x \in \mathbb{R}^4$ be defined as:

$$x = \begin{bmatrix} Glu \\ X \\ H_2 \\ H_{2,gas} \end{bmatrix}$$

Let us define in addition $u = Q_{in}$ as a controlled input and $w = Glu_{in}$ as a disturbance.

A reduced nonlinear system can be defined as:

$$\dot{x}(t) = f(x, u, w) \tag{12}$$

By linearizing the non-linear model (12) around an operating point (x^*, u^*, w^*), a reduced linear state space model is obtained as:

$$\dot{\bar{x}}(t) = A\bar{x}(t) + B_u \bar{u}(t) + B_w \bar{w}(t) \tag{13}$$

where:

- A is the Jacobian matrix $J_f(x)|_{(x^*,u^*,w^*)}$.
- B_u is the Jacobian matrix $J_f(u)|_{(x^*,u^*,w^*)}$.
- B_w is the Jacobian matrix $J_f(w)|_{(x^*,u^*,w^*)}$.
- $\bar{x}(t) = x(t) - x^*$.
- $\bar{u}(t) = u(t) - u^*$.
- $\bar{w}(t) = w(t) - w^*$.

As mentioned in Section 1, the output of the system is the hydrogen gas flow rate at the reactor output. Thus, according to equation (4) , the measured output is defined as:

$$y(t) = Cx(t) = \frac{RT_{amb}}{P_{atm} - p_{vap,H_2O}} V \left(\frac{\rho_{H_2}}{M_{H_2}} \right) \tag{14}$$

By regarding equations (5) and (6) it is easy to verify that matrix C takes the following form:

$$C = \begin{bmatrix} 0 & 0 & c_{H_2} & c_{H_{2,gas}} \end{bmatrix}$$

with:

$$c_{H_2} = \frac{RT_{amb} V k_L a_{H_2}}{(P_{atm} - p_{vap,H_2O}) M_{H_2}}$$

$$c_{H_{2,gas}} = -\frac{R^2 T_{amb} V k_L a_{H_2} K_{H,H_2} T_{reac}}{(P_{atm} - p_{vap,H_2O}) M_{H_2}}$$

The measured output is defined in terms of $\bar{x}$ as:

$$\bar{y}(t) = y(t) - Cx^* = C\bar{x}(t) \tag{15}$$

The following Luenberger observer is proposed to estimate x without knowledge of w:

$$\dot{\hat{\bar{x}}}(t) = A\hat{\bar{x}}(t) + B_u \bar{u}(t) + L\left(\bar{y}(t) - \hat{\bar{y}}(t)\right) \tag{16}$$

On the other hand, the glucose dynamics is modeled by:

$$\begin{aligned} \dot{Glu} &= k_{11} r_1 + k_{12} r_2 - D(Glu - Glu_{in}) \\ \dot{Glu} &= DGlu_{in} + h(Glu, X) \end{aligned}$$

where $h(Glu, X) = k_{11} r_1 + k_{12} r_2 - DGlu$. $DGlu_{in}$ is unknown but it is an absolutely continuous function of time, its dynamics can therefore be modeled as:

$$\frac{d(DGlu_{in})}{dt} = \delta_2(t)$$

Thus, the dynamics of Glu and $DGlu_{in}$ is modeled by the following ODE system:

$$\begin{aligned} &\dot{Glu} = DGlu_{in} + h + \delta_1(t); \quad |\delta_1| \leq c_1, \ c_1 > 0 \\ &(D\dot{Glu}_{in}) = \delta_2(t); \quad |\delta_2| \leq c_2, \ c_2 > 0 \end{aligned} \tag{17}$$

Note that $\delta_2(t)$ captures the uncertainties about $DGlu_{in}$ while $\delta_1(t)$ captures the uncertainties about r, Glu and X.

A super-twisting observer is then proposed to estimate Glu_{in} as:

$$\begin{aligned} \dot{\hat{Glu}} &= (D\hat{Glu}_{in}) + h(Glu, X) + \gamma_1\phi_1(\epsilon_1) \\ (\dot{D\hat{Glu}_{in}}) &= \gamma_2\phi_2(\epsilon_1) \end{aligned} \tag{18}$$

where:

$$\epsilon = \begin{bmatrix} \epsilon_1 \\ \epsilon_2 \end{bmatrix} = \begin{bmatrix} Glu - \hat{Glu} \\ DGlu_{in} - (D\hat{Glu}_{in}) \end{bmatrix}$$

$$\phi_1(\epsilon_1) = |\epsilon_1|^{1/2}\text{sign}(\epsilon_1)$$

$$\phi_2(\epsilon_1) = \frac{1}{2}\text{sign}(\epsilon_1)$$

Since the super-twisting observer (18) needs the glucose and the biomass concentrations to compute the term $h(Glu, X)$ and the error ϵ_1, it uses the estimations $\hat{x}_1$ and $\hat{x}_2$ made by the Luenberger observer (16).

3.3 Output tracking problem

Now, we address the problem of tracking the HPR_{max} computed by solving the NLP problem (11). Let us first define $\sigma = HPR - HPR_{max}$ and

$$HPR = \frac{Q_{H_2,gas}}{V} = \frac{y}{V}$$

By derivating σ with respect to time, it is easy to verify that $\dot{\sigma}$ has the form:

$$\dot{\sigma} = g_1(x,t) + g_2(x,t)u(t) \tag{19}$$

The system described by the dynamics (12) and the controlled output σ has therefore relative degree 1.

The following anti-windup super-twisting controller is proposed to track the HPR_{max} computed by solving the NLP problem (11):

$$u = -\lambda|\sigma|^{1/2}\text{sign}(\sigma) + u_1,\ \dot{u}_1 = \begin{cases} -u, & |u| > u_{max} \\ -\alpha\text{sign}(\sigma), & |u| < u_{max} \end{cases} \tag{20}$$

The precedent super-twisting control law guarantees the appearance of a 2-sliding mode $\sigma = \dot{\sigma} = 0$ in system (19), which attracts the trajectories in finite time. The control $u(t)$ enters in finite time the segment $[-u_{max}, u_{max}]$ and stays there. It never leaves the segment, if the initial value is inside at the beginning (Shtessel et al., 2014).

Hence, the flow rate at the reactor input computed according to the equation (20) will drive the HPR to the value HPR_{max}.

3.4 Optimization algorithm

The algorithm proposed to maximize on-line the hydrogen production in the dark fermenter consists of the following steps:

(1) Estimate the glucose concentration at the reactor input using the coupled observer (16)-(18).
(2) Using the Glu_{in} estimation, solve the NLP problem (11) to compute the maximum HPR and the optimal Q_{in} of the biohydrogen production process.
(3) By considering the HPR_{max} (as reference) and the optimal Q_{in} (as initial value) computed in the precedent step, use the super-twisting controller (20) to track the maximum hydrogen production rate by controlling the flow rate at the reactor input.

4. RESULTS AND DISCUSSION

In the following simulations, the value of the parameters of the model (1), used in addition by the coupled observer (16)-(18), were taken from (Torres Zúñiga et al., 2015), while the parameters of the model (9), used as objective function in the NLP problem (11), were taken from (Ramírez et al., 2015). On the other hand, the super-twisting controller parameters considered were $\lambda = 0.05$ and $\alpha = 0.5$.

The model of the hydrogen production reactor and the optimization strategy were simulated during 67 days in *MATLAB* considering a sample period $T = 4h$. Furthermore, the ODEs were solved using the *ode15s* solver. The observer started after one day from the process beginning. The optimization problem was solved and the maximum HPR was tracked after three day from the process beginning, once the observer converged.

Figure 2 shows the glucose at the reactor input (solid blue line) considered in this simulation and the glucose estimated (dashed red line) by the coupled observer (16)-(18). As can be observed, the estimations are very close to the 'real' concentrations along the simulation.

Figure 3 shows the model (9) in solid blue line, while in red balls the optimal points (HPR_{max}, OLR_{opt}) computed by solving the NLP problem (11) for $5h \le HRT \le 12h$. Points around $HPR_{max} = 10L[H_2]L^{-1}d^{-1}$ correspond to substrate estimations around $\hat{Glu}_{in} = 10gL^{-1}$. Points close to $HPR_{max} = 15L[H_2]L^{-1}d^{-1}$ correspond to substrate estimations around $\hat{Glu}_{in} = 15gL^{-1}$. Points around $HPR_{max} = 20L[H_2]L^{-1}d^{-1}$ correspond to substrate estimations around $\hat{Glu}_{in} = 20gL^{-1}$. It must be pointed out that the maximum HPR of the model (9) is not reached because the constrain in the NLP problem (11) was defined for a minimum HRT of $5h$. This is due to the fact that the Luenberger observer (16) was designed to estimate the vector state x around $HRT = 8h$. Simulations for $HRT < 5h$ demonstrated that the Luenberger observer does not converge and therefore, the substrate at the reactor input is not correctly estimated.

Once an optimal point (HPR_{max}, OLR_{opt}) has been computed by solving the NLP problem (11), the maximum produc-

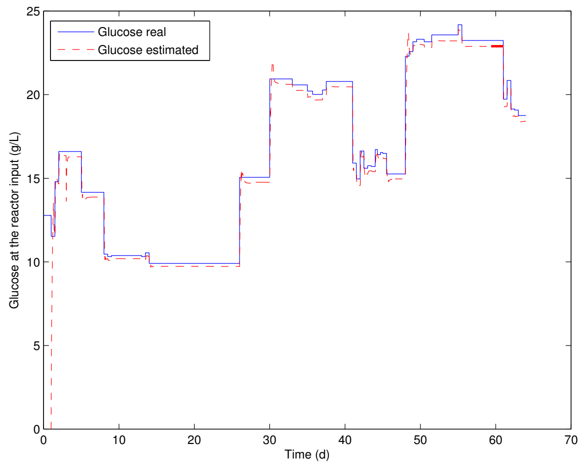

Fig. 2. Estimation of the input glucose concentration. In solid blue line the 'real' concentration and in dashed red line the estimated one.

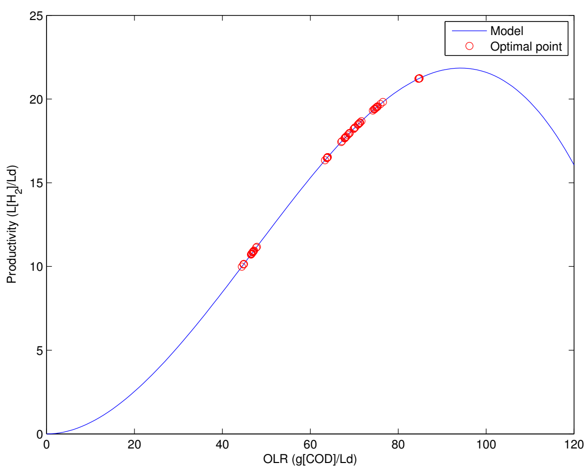

Fig. 3. Model $HPR(OLR)$ and optimal point computed by solving the NLP problem (11).

tivity (HPR_{max}) is tracked by the super-twisting controller (20), considering $Q_{in,opt}$ as initial value. Figure 4 shows in dashed red line the 'maximum' HPR tracked and in solid blue line the 'real' HPR of the bioreactor. As can be regarded, the super-twisting controller correctly tracks the HPR_{max} reference along the simulation, with a maximum transitory period of three days. It must be pointed out that in the time periods where a small error is observed ($< 1L[H_2]L^1d^{-1}$), it is due to the saturation of the control input (the NLP imposes that the minimum HRT allowed is $5h$).

Finally, Figure 5 shows the HRT of the bioreactor along the simulation. As can be observed, the super-twisting controller (20) correctly saturates the flow rate at the reactor input (($HRT = V/Q_{in}$)), since the minimum HRT computed is $5h$, as imposed by the constraint of the NLP problem (11).

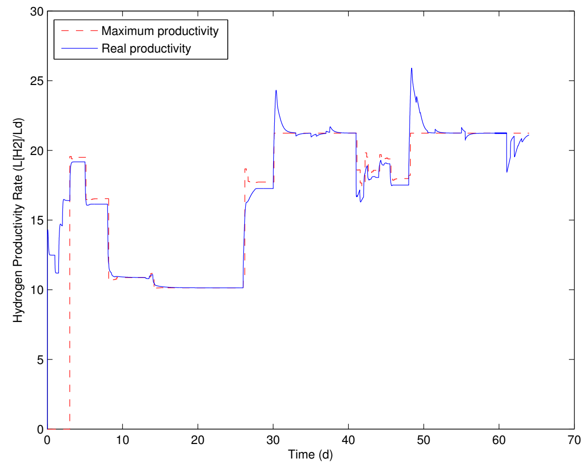

Fig. 4. Productivity of the hydrogen production bioreactor. In solid blue line the 'real' productivity and in dashed red line the 'maximum' one tracked.

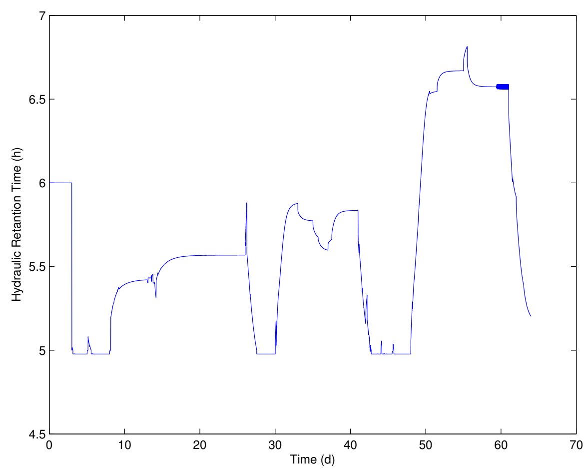

Fig. 5. HRT computed by the super-twisting controller (20).

5. CONCLUSIONS

In this work, an heuristic optimization strategy to maximize the productivity into a hydrogen production bioreactor was presented. The strategy consists of solving a nonlinear optimization problem to compute the maximum hydrogen production and the optimal flow rate at the reactor input and then, to track the maximum productivity by controlling the flow rate at the reactor input via a super-twisting controller.

The strategy is simple to implement and even if it consists of three element, the optimizer, the observer and the controller, the computing resources needed is not a critical issue. The observer represents the largest challenge because it consist of 6 ordinary differential equations. On the other hand, the nonlinear optimization problem has only one objective function, one optimization variable and one constraint, while the super-twisting controller has just the form of a proportional-integral controller.

Simulations demonstrate the feasibility of the strategy for hydraulic retention times over $5h$. The observer estimates correctly the glucose at the reactor input, the nonlinear optimization problem computes the maximum productivity and the optimal flow rate at the reactor input respecting the constraint, and the super-twisting controller correctly tracks the maximum productivity respecting the minimum hydraulic retention time allowed. However, it is well known that the hydrogen production process reach an optimal operation at hydraulic retention times around of $4h$. The Luenberger observer has to be therefore redesigned to correctly estimate the glucose and the biomass into the bioreactor around that hydraulic retention time.

ACKNOWLEDGEMENTS

This research was financed by CONACYT (project 100298) and PAPIIT-UNAM (project IN112114).

REFERENCES

Aceves-Lara, C., Latrille, E., Bernet, N., Buffière, P., and Steyer, J. (2008). A pseudo-stoichiometric dynamic model of anaerobic hydrogen production from molasses. *Water research*, 42, 2539–2550.

Aceves-Lara, C., Latrille, E., and Steyer, J. (2010). Optimal control of hydrogen production in a continuous anaerobic fermentation bioreactor. *International journal of hydrogen energy*, 35, 10710–10718.

Huang, S., Chen, H., Chung, C., Wu, C., Tsai, T., Chu, C., and Lin, C. (2012). Fermentative hydrogen production using a real-time fuzzy controller. *International journal of hydrogen energy*, 37, 15575–15581.

Levin, D., Pitt, L., and Love, M. (2004). Biohydrogen production: prospects and limitations to practical application. *International journal of hydrogen energy*, 29, 173–185.

Ramírez, J., Torres Zúñiga, I., and Buitrón, G. (2015). On-line heuristic optimization strategy to maximize the hydrogen production rate in a continuous stirred tank reactor. *Process Biochemistry*. URL http://dx.doi.org/10.1016/j.procbio.2015.03.003.

Shtessel, Y., Edwards, C., Fridman, L., and Levant, A. (2014). *Sliding mode control and observation*. Springer.

Torres Zúñiga, I., Vargas, A., Latrille, E., and Buitron, G. (2015). Robust observation strategy to estimate the substrate concentration in the influent of a fermentative bioreactor for hydrogen production. *Chemical Engineering Science*, 219, 126–134.