Experimental validation of a coupled nonlinear observer in a hydrogen production dark fermenter

A. Villa-Leyva[1], Axue Zhang[1], I. Torres-Zúñiga[1], A. Vargas[1], G. Buitrón[1],*

[1]*Laboratorio de Investigación en Procesos Avanzados de Tratamiento de Aguas, Unidad Académica Juriquilla, Instituto de Ingeniería, Universidad Nacional Autónoma de México, Blvd. Juriquilla 3001, Querétaro, Qro. C.P. 76230, México.*
**Tel: +524421926165; e-mail: gbuitronm@iingen.unam.mx*

ABSTRACT

In order to maximize the production of hydrogen from glucose in a continuous dark fermenter by modifying the OLR, specifically manipulating the inflow rate, the real knowledge of the substrate concentration at the reactor input is needed. Since it cannot be measured on-line, a Luenberger observer coupled to a Super-Twisting one can be used to estimate the substrate concentration at the reactor input. This study aimed to demonstrate the experimental validation of the proposed observer, which was tested by numerical simulations based on a mathematical model calibrated by experimental data in a previous research. Five conditions with different combinations of hydraulic retention time (HRT) and inflow substrate concentration were performed in the fermentative reactor for calibrating the mathematical model of the observer. These conditions were chosen based on Latin hypercube experimental design. The collected data allowed testing the proposed nonlinear observer and the results are encouraging, showing good estimations of the inflow substrate concentration. However the model is sensitive to perturbations, which leads to an erroneous estimation of the VSS concentration with the Luenberger observer. Nevertheless, the observer may be suitable to complement an optimization strategy previously proposed.

Keyword : bio-hydrogen production; nonlinear observer; bioprocess control

1. Introduction

The continued usage of fossil fuels is threatened the environment by increasing concentration of greenhouse gases and causing serious pollution problems in the atmosphere. Moreover gasoline, diesel and natural gas, with nearly 80% of global energy demand supplied are finite sources and are rapidly becoming scarcer and more expensive [1]. Hydrogen has been introduced as a potential replacement for energy resource due to its low generation of pollutants and high energy density [2].

Currently, most hydrogen is produced form non-renewable sources such as oil, natural gas and coal; about 50% is obtained using thermocatalytic techniques and gasification of natural gas, heavy oils, cola and naphtha [3]. On the other hand, the biological processes for the hydrogen production are presented as an alternative. These processes can be classified into three types [4]: bio-photolysis, photo-fermentation and dark-fermentation. Fermentative hydrogen production has been received increasing attention in recent years, it has the advantage of rapid hydrogen production rate and simple operation, moreover it can use organic solid wastes and the organic compound in the wastewater as substrate [5].

In fermentative hydrogen production the operation parameters have a crucial role on the metabolic pathway of the microorganisms, as a result, influence the process efficiency, product gas quality and energy inputs. Many researchers have been focusing on optimal operation parameters (pH, temperature, hydrogen partial pressure, and inoculum) to obtain the maximum hydrogen yield and hydrogen production rate. One of the operational conditions that affect the hydrogen production rate (HPR) in a dark fermenter reactor is the organic loading rate (OLR). According to [6] and [7] an optimum OLR exits in which the HPR is maximum, moreover this optimum OLR is close to the overloading one. In order to maximize the HPR, the OLR should be maintained optimal. The OLR depends on inflow rate and substrate concentration at the reactor input, a controlled variable and an uncontrolled one, respectively.

In order to optimize the productivity of hydrogen in a continuous dark fermenter by modifying the OLR, specifically manipulating the inflow rate, based on an optimization strategy proposed in [13], the real knowledge of the substrate concentration is needed. Since it is unpractical to measure on-line, a coupled observer is proposed [11]. It is able to estimate the substrate concentration at the reactor input by measuring the hydrogen flow rate at the reactor output. Although a validation of the proposed estimator has already been done using numerical simulations based on a mathematical model that was calibrated with previous experimental data, an experimental validation of the proposed estimator is needed as well. This work presents the results of this experimental validation.

2. Experimental

2.1 Inoculum, feed water composition and start-up of reactor

The inoculum was obtained from a UASB reactor used for treating the wastewater of a brewery plant. The sludge was subjected to a heat treatment described by [8]. The granular sludge was heated at 104°C during

24h in order to select the hydrogen-producing spore forming anaerobes and inhibit the activity of hydrogen consumers. The heated material was broken down in a mortar and sieved with a # 20 mesh (850µm). Glucose was used as organic substrate. For every gram of glucose 104 mg NH_4Cl and 50 mg K_2HPO_4 were added. For every liter of feed solution, the following amounts of mineral salts were added: 0.4 mg; $MnCl_2{\cdot}4H_2O$, 20 mg; $MgCl_2{\cdot}6H_2O$, 20 mg; $FeSO_4{\cdot}7H_2O$, 2 mg; $CoCl_2{\cdot}6H_2O$, 2 mg; $Na_2MoO_4{\cdot}2H_2O$, 2 mg; H_3BO_4, 2 mg; $NiCl_2{\cdot}6H_2O$, 2 mg; $ZnCl_2$, 2 mg. The experiment was carried in a continuous stirred tank reactor (CSTR) of 1.2 L with a working volume of 0.9 L equipped with a ez-biocontroler (Applikon Biotechnology, Schiedam, The Netherlands). The biocontroler allowed to keep the following operating conditions: temperature of 35 °C, stirring velocity at 100 rpm and the pH at 5.5. A lever sensor was set in the reactor to maintain the liquid volume.

The culture was started in batch mode with glucose and volatile suspended solids (VSS) concentration of 15 g/L and 4 g/L, respectively. After 5 cycles of 12 h the reactor was switched to continuous system. The feed solution was fed into the reactor by a peristaltic pump (Masterflex, Barnann, Illinois, USA).

2.2 Data acquisition system

Biogas flow was measured using a flow meter ADM 2000 (Agilent Technologies, Inc.) connected at a serial port of a personal computer. The percent of hydrogen in the biogas was measured with a HY-OPTIMA analyzer model 7000 (H2scan), which was connected to the personal computer using a device DAQ NI-USB-6008 (National Instrument Inc.).

2.3 Analytical method

The concentration of glucose at the influent and the effluent were determined according to the Dubois method [9]. The VSS concentration was determined according to the standard method [10]. To identify the biogas composition a gas chromatograph (SRI-8610C) was used, equipped with a thermal conductivity detector and two stainless steel columns packed. The carrier gas used was nitrogen at a flow rate of 20 ml/min. The temperatures of operation at the injector and the detector were 90 °C and 150 °C, respectively. The concentration of volatile fatty acids (VFA), acetic, propionic and butyric, and ethanol were measured using another gas chromatograph (Varian 3300) equipped with a flame ionization detector and a silica capillary column (Zebron ZB-FFAP) of 15 m long and 0.53 mm internal diameter. The samples were filtered firstly in glass fiber filter and then a nitrocellulose filter (0.45 µm), and later on acidified with hydrochloric acid. Nitrogen was used as carrier gas at flow of 9.5 ml/min. Injector and detector temperatures were maintained at 190 and 210 °C, respectively.

2.4 Coupled observer to estimate the glucose concentration at the reactor input.

The hydrogen production bioreactor has two inputs, the substrate concentration Glu_{in} (an uncontrolled input) and the flow rate Q_{in} (a controlled input). On the other hand, the total gas flow rate Q_{gas} and the hydrogen fraction $\%H_2$ at the reactor output are measured. Using both measurements, the hydrogen flow rate at the reactor output $q_{H2,gas}$ can be calculated.

In order to estimate the glucose concentration at the reactor input, the coupled observer proposed in [10] and shown in figure 1, is considered. The observer consists of a Luenberger observer followed by a super-twisting observer. By measuring the hydrogen flow rate at the reactor output the Luenberger observer estimates the glucose and the biomass concentrations inside the reactor. Then, the super-twisting observer uses these estimations to estimate the glucose concentration at the reactor input.

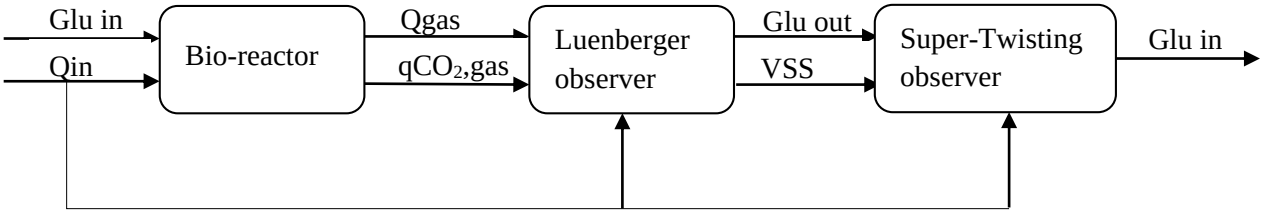

Figure 1. Coupled observer to estimate the glucose at the reactor input.

Since the coupled observer is based on the model of the process, firstly, the parameters of the model are computed. The model considered in [11] has two kinds of parameters: the pseudo-stoichiometric and the kinetic ones. On the other hand, the model has ten state variables, eight in liquid phase: glucose, acetate, propionate, butyrate, ethanol, biomass, carbon dioxide and hydrogen; and two in gas phase: carbon dioxide and hydrogen.

The biohydrogen production reactor was then operated in five nominal conditions resulting of the combination of hydraulic retention times (HRT) and inflow substrate concentrations according to a Latin hypercube experimental design [12] (see Table 1). Once the steady state was reached for each operation condition the analyses were performed, and then the reactor returned to the standard operation condition (HRT=8h and Glu_{in}=15g/L) before test the next condition.

Table 1. Nominal conditions of operation reactor according to Latin hypercube experimental design.

Number of condition	Glucose concentration (g/L)	HRT (h)
1	15	8
2	10	10
3	20	8
4	15	10
5	25	6

A variance analysis was performed to determine that two reactions explain 88% of the total variance of the experimental data measured between days 1 and 63. By considering three reactions 94% of the variance can be explained. This analysis shows that adding a third reaction would account for only 6% of the variance of the experimental data, which is lower than the 100/8 = 12.5 % of the variance of each state variable in liquid phase. In this way, the pseudo-stoichiometric parameters are grouped in an 8x2 matrix K. The reactions considered are based on the Monod model. Therefore, the kinetic parameters are grouped

in two 2-dimension column-vectors μmax and KGlu.

The Luenberger observer considers only the dynamics of the glucose, the biomass and the hydrogen in liquid and gas phases. Thus, the Luenberger observer correction term is a 4-dimension row-vector LL.

The super-twisting observer considers the dynamics of the glucose inside the reactor and the glucose at the reactor input. The super-twisting observer correction term is then a 2-dimension row-vector LST.

In order to compute the matrix K an optimization Non-Linear Problem (NLP) to minimize the product of the kernel of KT and the matrix of experimental data is solved by using the MATLAB function fmincon with the SQP algorithm [11]. In order to compute the vectors max and KGlu a NLP to minimize the difference between experimental data and simulation results is solved by using the MATLAB function RegNonO [10]. The parameters computed are then:

$$K^T = \begin{bmatrix} -1 & 0.3238 & 0.0174 & 0.2737 & 0.0219 & 0.1135 & 0.0087 & 0 \\ -1 & 0 & 0.0205 & 0.3028 & 0.0242 & 0.0934 & 0.0049 & 0.0351 \end{bmatrix}$$

$$\mu_{\max} = [37.32 \quad 27.24]$$

$$K_{Glu} = [0.29 \quad 0.26]$$

On the other hand, in order to compute the correction term LL, an optimization semi-definite problem (SDP) to decrease the influence of a disturbance (the unknown input Glu_{in}) on the estimation error is solved [11]. In order to compute the correction term LST, a SDP to decrease the influence of the uncertainties related to both the glucose and the input glucose dynamics on the estimation error is solved [11]. Both SDP are solved using the SEDUMI solver over the YALMIP toolbox in the MATLAB environment. The correction terms computed are then:

$$L_L = \begin{bmatrix} 108.93 \\ 0.67 \\ 0.06 \\ 0.08 \end{bmatrix}$$

$$L_{ST} = \begin{bmatrix} 0.43 \\ 1.79 \end{bmatrix} x10^6$$

The coupled observer is implemented in the MATLAB environment. The hydrogen flow rate at the reactor output is measured each 10 seconds. After 10 minutes, the mean is calculated and it is used together with the current input flow rate Qin to compute the current estimations of the glucose and the biomass inside the reactor, and the glucose at the reactor input.

3. Results and discussion

Once the model has been calibrated and both correction terms, from the Luenberger observer and the super-twisting observer, have been computed, the observer was validated on-line by using real-time hydrogen flow rates measured at the bioreactor output. In order to validate the observer, the reactor was operated for 15 days in three different nominal conditions of substrate concentration and HRT: on days 1-7 at 20 g/L and 6 h, on days 7-13 at 15 g/L and 4 h, and on days 13-15 at 10 g/L and 4 h, respectively.

Figures 1, 2 and 3 show the estimated VSS concentration in the reactor, glucose concentration at the effluent and at the influent, respectively. During the first condition (days 1-7) the estimation of the VSS was very close to the analytical method. After the day seven, in the second condition (15 g/L), the VSS concentrations estimated for the observer were below 1 g/L. Similar behavior was observed in the estimation of the substrate concentration at the input. During the operation at 20 g/L the value estimated was close to the value obtained with the analytical method, and at 15 g/L (days 7-13) the estimation of the substrate at the input was lower than the real concentration. Apparently, the disturbance (the unknown input) was not correctly rejected by the Luenberger observer because of the uncertainty on the glucose dynamics grew, the glucose at the reactor input was not correctly estimated as well. In order to have a better estimation the operating point of the Luenberger observer was modified in the ninth day of the bioreactor operation. Nevertheless, even if the error estimation decreased, the coupled observer performance was not satisfactory. Then, the condition of operation was changed to 10 g/L of substrate concentration. In this condition the estimation of substrate concentration at the input was very close to the real concentration despite the error estimation of VSS concentration, demonstrating the robustness of the super-twisting observer.

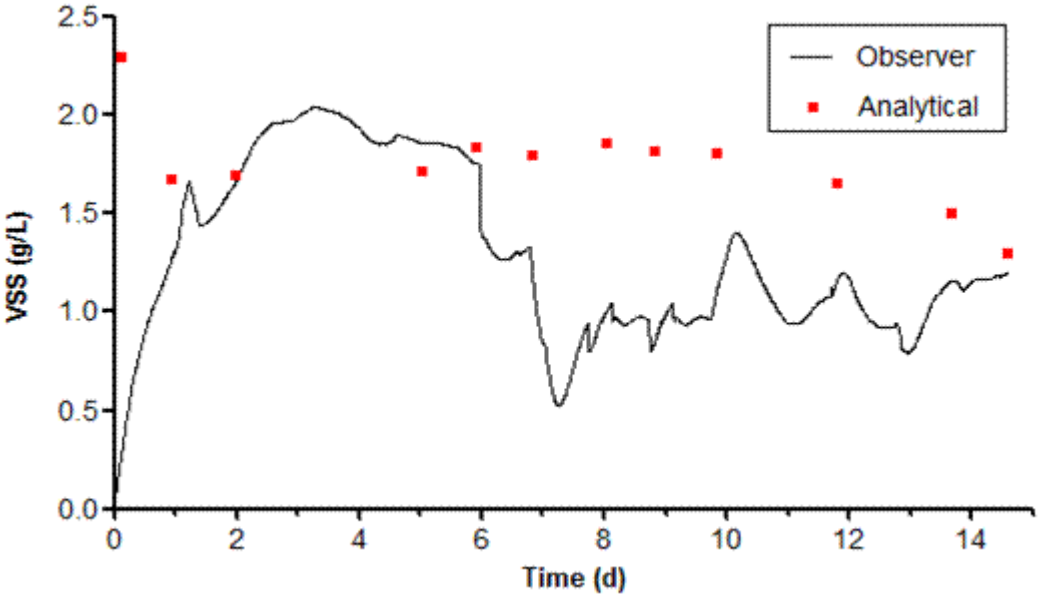

Figure 1. Estimation of VSS and the result of analytical method.

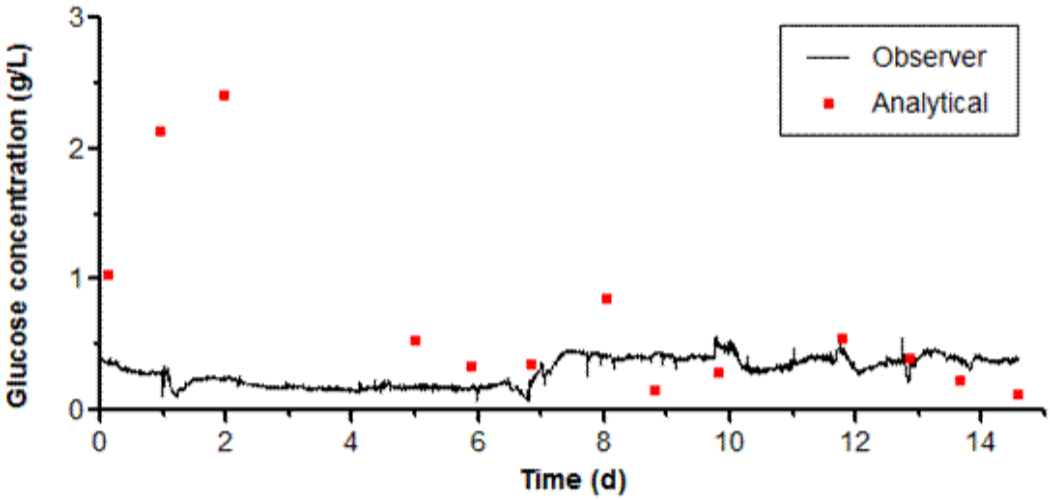

Figure 2. Estimation of substrate concentration inside the reactor.

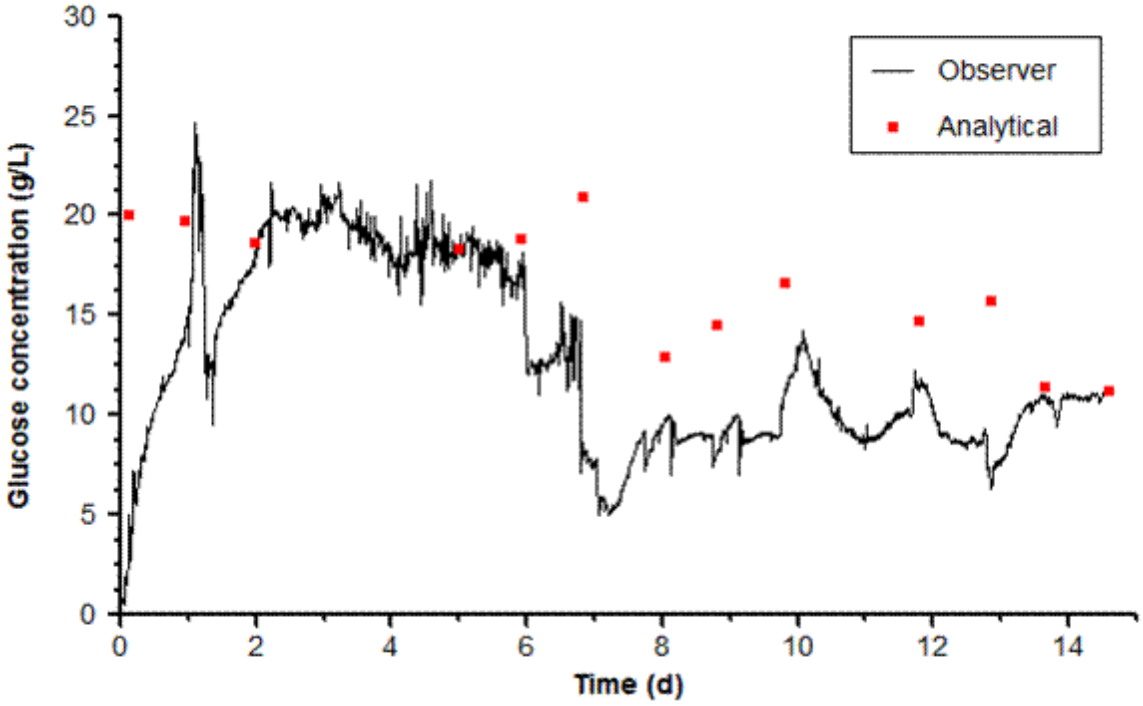

Figure 3. Estimation of substrate concentration at the input of the reactor.

4. Conclusions

This study presented the results of experimental validation of a Luenberger observer coupled to a Super-Twisting for the estimation of substrate concentration at the input of the reactor. When the glucose concentration was 20 and 10 g/L, the observer was able to estimate the concentration to close to the real. The result was not completely successful at 15 g/L, although the operating point of the Luenberger observer was modified, the error estimation was not sufficiently decreased.

Therefore, further experimental implementation of the coupled observer must be carried out in order to compute the observer gains to correctly estimate the substrate at the reactor input for any operation condition.

Once the observer correctly estimates the glucose at the reactor input, the optimization strategy will be implemented to compute the optimal flow rate at the reactor input for maximizing the hydrogen productivity rate.

Acknowledgements

This research was supported through FOMIX CONACYT project (192341). The authors are grateful to Jaime Perez for the technical support and fruitful discussions.

References

[1] R. L. Evans. Fueling our future: An introduction to Sustainable Energy. 1st ed. Cambridge University Press. N. Y. 2007.

[2] B. Rohland, J. Nitsch, H. Wendt. Hydrogen and fuel cells: the clean energy system. J. of power source. 1992; 37: 271-277.

[3] Mohan, S. V., Bhaskar, Y. V. Sarma, P. Bio-hydrogen production from chemical wastewater treatment in biofilm configured reactor operated in periodic discontinuous batch mode by selectively enriched anaerobic mixed consortia. Water Research. 2007; 41: 2652–2664.

[4] K. K. Ilgi, K. Fikret. Bio-hydrogen production from waste materials. Enzyme and Microbial Technology. 2006; 38: 569-582.

[5] J. Wang, W. Wan. Factors influencing fermentative hydrogen production: Areview. Int. J. of Hydrogen Energy. 2009: 799-811.

[6] H. Hafez, G. Nakhla, M. Hesham, E. Elbeshbishy, B. Baghchehsaraee. Effect of organic loading on a novel hydrogen bioreactor. Int. J. of Hydrogen Energy. 2010; 35: 81-92.

[7] L. Shen, D. M. Bagley, S. N. Liss. Effect of organic loading rate on fermentative hydrogen production from continous stirred tank and membrane bioreactor. Int. J. of Hydrogen Hydrogen Energy. 2009; 34: 3689-3696.

[8] G. Buitrón, C. Carbajal. Biohydrogen production from Tequila vinasses in an anaerobic sequencing batch reactor: Effect of initial substrate concentration, temperature and hydraulic retention time. Bioresource Technology. 2010; 101: 9071–9077.

[9] M. Dubois, K. A. Gilles, P. A. Rebers, F. Smith. Colorimetric method for determination of sugar related substrate. Analytical Chemistry. 1956; 28: 350-356.

[10] APHA. Standard Methods for the Examination of Waters and Wastwater. 11th ed. Port City Press. Baltimore. 2005.

[11] I. Torres-Zúñiga, J. E. Ramírez, A. Vargas, E. Latrille and G. Buitrón. Coupled robust observer to estimate the substrate at the input of a biohydrogen production reactor. Submitted to Chemical Engineering Journal. 2014.

[12] A. Saltelli, M. Ratto, T. Andres, F. Campolongo, J. Cariboni, D. Gatelli, M. Saisana, S. Tarantola, Global sensitivity analysis, John Wiley and Sons, Ltd, 2008.

[13] J. E. Ramírez, I. Torres Zúñiga, G. Buitrón. Real time optimization for fermentative hydrogen production in a continuous reactor. Submitted to Process Biochemistry.