

Biohydrogen production: A new controllability criterion for analyzing the impacts of dark fermentation conditions

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Abstract. Biohydrogen production from renewable resources using dark fermentation has become an increasingly attractive solution in sustainable global energy supply. So far, there has been no report on the controllability analysis of biohydrogen production using dark fermentation. Process controllability is a crucial factor determining process feasibility. This paper presents a new criterion for assessing biohydrogen process controllability based on PI control. It proposes the critical loop gain derived via Routh stability analysis as a measure of process controllability. Results show that the dark fermentation using the bacteria from anaerobic dairy sludge and substrate source from sugarcane vinnasse can lead to a highly controllable process with a critical loop gain value of 4.3. For the two other cases, an increase of substrate concentration from 10 g/L to 40 g/L substantially reduces the controllability. The proposed controllability criterion is easily adopted to assess the process feasibility based on experimental data.

Keywords: Biohydrogen; Dark fermentation; Process controllability; Renewable energy; Multi-scale kinetic mode.

1. Introduction

The Global economy and energy supply currently have heavily relied on fossil fuels. The high demand for energy has led to a serious concern on fossil fuel consumption and uncontrolled carbon dioxide (CO₂) emissions from fossil fuels, and industries that have contributed to global climate change [1-3]. Furthermore, the extreme reliance on nonrenewable fossil fuels has led to an energy security issue. To minimise the depletion of fossil fuels and associated CO₂ emissions, sustainable and renewable energy sources are required. Thus, sustainable and renewable energy like hydrogen (H₂) [4], microalgae cultivation [5, 6], and biomethanation [7] play an important role in creating sustainable energy production.

Recently, H₂ becomes the most important alternative energy sources that offer a friendly climatic solution to the prevalent energy crisis [8-10]. High demand of H₂ in the global market has led to various strategic techniques to produce H₂ [11]. The sustainable strategy for the utilization of agricultural wastes in biohydrogen production has drawn intensive research attention recently because of several advantages. First, bioenergy is renewable which can lead to lower greenhouse gas emissions, such as CO₂ and methane emissions to the environment - the potential to reduce global warming.

Besides, for the agricultural-related industry, large volumes of solid, liquid, and gaseous wastes are being generated that have raised serious issues due to their environmental degradation factors [12]. Moreover, the partial management of waste approach is insufficient because of the slow process involved that cannot manage a large amount of the waste. Unfortunately, to date, most of these wastes remain underutilized and rather have become the source of environmental concerns in the industry. Presently, the lack of proper and technical resolution to these wastes has generated a lot of environmental problems in societies where these wastes are either composted or utilized for soil amendments. Hence, this has led to a steady increase in waste stockpiles causing environmental pollution.

Meanwhile, to counter and address this environmental pollution issue, anaerobic digestion can be a solution since it is a famous technology currently processing several feedstock types from the agricultural sector and other organic industrial waste streams [13]. Even though other biological H₂ production processes like direct bio-photolysis, indirect bio-photolysis, PF, DF, and hybrid fermentation technologies have been investigated and found of being economically feasible, and less energy-intensive. Besides, they can consume numerous different feedstocks and run at ambient temperature and pressure [14-16]. Hence, among the numerous H₂ production routes, the anaerobic DF process happens to be more efficient and significant since it has higher productivity and yield [17, 18]. Additionally, DF produces H₂ from numerous renewable biomass such as crop residues [19], lignocellulosic biomass [20], waste from organic material [21], and biomass algae [22, 23]. Therefore, the production of H₂ by DF is promising as it gives a higher H₂ yield, and lower production and capital costs as compared to the PF process [24]. Thus, these biological fermentations are less cost-efficient, and not a dirty procedure to generate biohydrogen [25].

Furthermore, anaerobic digestion is also a widely applied bioprocess for biogas production using organic waste [26] such as biohydrogen production [27]. Producing biohydrogen via the bioprocessing route is preferable since it can perform at room pressure and temperature. Meanwhile, it requires lower energy-intensive and more environmentally friendly than

the conventional chemical methods [10]. This route can be divided into bio-photosynthesis using organic materials through dark fermentation (DF), and light fermentation [9, 28]. Recently, among renewable and sustainable sources, electrolysis is efficient since it provides 4% of total H₂ production worldwide. Moreover, water electrolysis generates H₂ with an efficiency of 52% with a corresponding cost of 10.3 \$/kg. However, this water electrolysis has the challenges of H₂ production in a cost-competitive way, with a specific cost (€/kW) compatible with market and fiscal requirements [29]. On the other hand, DF has higher stability and efficiency as compared to bio-photolysis which requires light fermentation. Besides having a relatively simple control system, the DF process can use a diverse group of organic wastes as substrate sources to produce H₂ [30].

Unfortunately, study on the dynamics controllability of the DF for biohydrogen production has not been reported so far. Crucial to the control strategy development is the process controllability of the given bioprocess design for the DF process, which depends on the type of bacteria, substrates, temperature, and substrate concentration. The present work attempts to narrow down the gap by proposing a new controllability measure suitable for the biohydrogen process.

2. Methodology

The controllability process indicates how easy it is to control the process. Meanwhile, in this study high controllability means that the process can be controlled easily. In another word, a high controllability process can attain high control performance, i.e., fast set point tracking, and fast disturbance rejection. Controllability can be measured using several indices, e.g., Relative Gain Array (RGA), Morari Index, nu-gap metric, and many more. This study proposes to use the loop gain of the Proportional Integral (PI) controller as a measure of single-loop process controllability application to biohydrogen process controllability analysis.

2.1 Stability of Process Integration (PI) Control

Assume the first-order plus dead time (FOPDT) process model, through the following equations for close loop characteristic and process transfer function respectively:

$$G_p = \frac{K_p e^{-\theta_p s}}{\tau_p s + 1} \tag{1}$$

Notations K_p , τ_p and θ_p denote the process gain, time constant, and dead time, respectively. Meanwhile, the Proportional-Integral (PI) controller transfer function in its ideal form is as follows:

$$G_c = K_c \left(1 + \frac{1}{\tau_I s} \right) \tag{2}$$

where K_c and τ_I represent the tunable parameters viz. controller gain and reset time, respectively.

Fig. 1 shows the block diagram of the standard single-loop control system where the corresponding process and controller transfer functions are as in (1) and (2). The block diagram assumes the dynamics of the actuator and sensor are fast compared to the dynamics of the process (i.e., pseudo-steady state of actuator and sensor). Thus, there is no need to include the actuator and sensor in the block diagram. Refer to

Fig. 1, Y_{sp} , E , C and Y denote the setpoint, error, controller output, and controlled variable signals; G_c and G_p are the controller and process transfer functions, respectively.

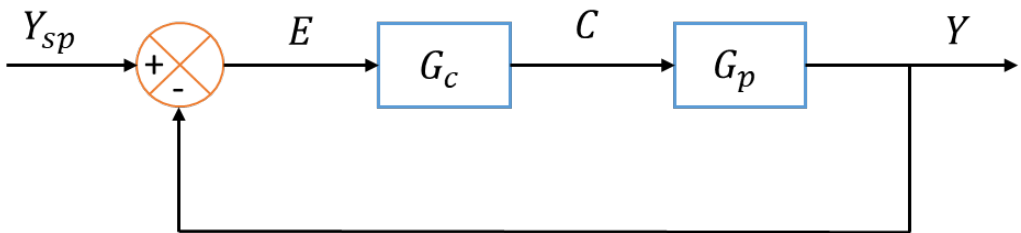


Fig. 1. Block diagram of the standard single-loop feedback control.

Based on the block diagram, one can derive the closed-loop setpoint transfer function as follows:

The closed loop set point transfer function is given as (Equation (3)):

$$H_r(s) = \frac{G_c(s)G_p(s)}{1 + G_c(s)G_p(s)} \tag{3}$$

Closed-loop input disturbance transfer function (Equation (4)):

$$H_{d_i}(s) = \frac{G_p(s)}{1 + G_c(s)G_p(s)} \tag{4}$$

Closed-loop output disturbance transfer function (Equation (5)):

$$H_{d_o}(s) = \frac{1}{1 + G_c(s)G_p(s)} \tag{5}$$

where R is a setpoint, E is an error, C is a controller output, D_i is an input disturbance, D_o is an output disturbance, Y is output or controlled variable, G_c is a controller transfer function or sub-system and G_p is a process transfer function or sub-system.

The closed-loop characteristic equation is expressible by a general characteristics polynomial equation [31, 32] (Equation (6)):

$$\psi = (h_n + K_L f_n)s^n + \dots + (h_1 + K_L f_1)s + (h_0 + K_L f_0) = 0 \tag{6}$$

Note that the parameters h_i and f_i for $i = 0, 1, \dots, n$ are often functions of the model parameters and the reset time. Meanwhile, K_L denotes the loop gain of the control system, i.e., $K_L = K_c K_p$ where K_c denotes the PI controller gain and K_p the process gain. Remember that the loop gain K_L is a dimensionless parameter directly related to the control performance. A high value of K_L means that the control system can exhibit higher performance than the smaller value of K_L and vice versa. Since K_L is directly related to the control performance, the loop gain value should be practically above zero. If its value is less than zero, then this implies that the control performance is lower than the open-loop system, i.e., the control system is practically useless.

2.2 Controllability Measure: Critical Loop Gain (CLG)

Notice that one of the upper limits of the loop gain, i.e., \bar{K}_{L_2} in is adjustable via the tuning value of τ_I . But another upper limit \bar{K}_{L_1} is fixed which is a non-function of the PI tuning parameter. The upper limit \bar{K}_{L_1} could be fixed by the process design characteristics as depicted by τ_p and θ_p . The minimum upper limit can be either \bar{K}_{L_1} or \bar{K}_{L_2} depending on the value of the reset time.

Both upper limits may have similar values, i.e. $\bar{K}_{L_1} = \bar{K}_{L_2}$. In this study, the critical loop gain (CLG) is defined for the case when $\bar{K}_{L_1} = \bar{K}_{L_2}$, which requires the reset time to have a critical value as follows (Equation (7)):

$$\tau_{I_{critic}} = \frac{\theta_p \tau_p}{\theta_p + \tau_p} \tag{7}$$

Therefore, CLG ($K_{L_{critic}}$) can be calculated in the following manner (Equation (8)):

$$K_{L_{critic}} = \frac{\tau_p}{\theta_p} = \frac{\tau_I}{\theta_p - \tau_I}, \quad \tau_I = \tau_{I_{critic}} \tag{8}$$

where τ_p , θ_p and τ_I denote the process model time-constant, deadtime, and PI control reset time, respectively.

Note that the CLG value represents process controllability. A large CLG value suggests that the process is easy to control. On the contrary, a small CLG value means that the process is hard to control. The highly controllable process can attain high closed-loop performance, i.e., as measured in terms of short settling time, fast disturbance rejection, small Integral Absolute Error (IAE), etc. High control performance is desirable because it can lead to energy saving, better product quality, profitability, and others.

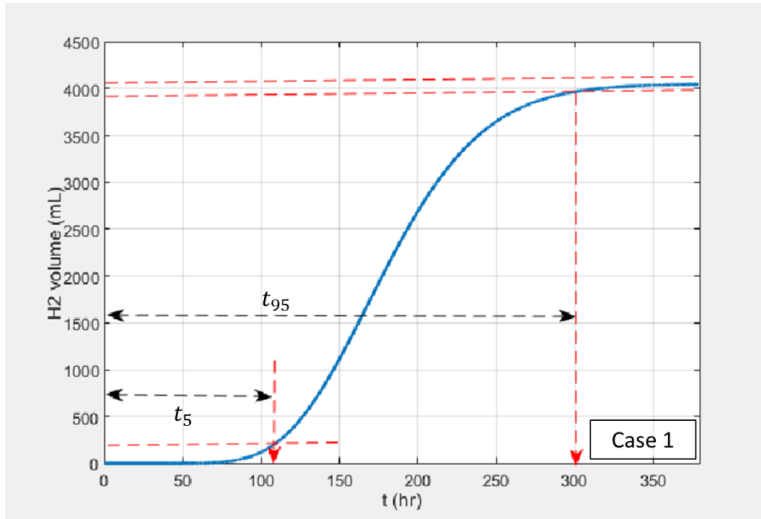


Fig. 2. Model-order reduction of biohydrogen production mode for case study 1 reported in [3].

2.3 Model-order Reduction

Recently, Lim and Nandong [4] conducted an extensive modeling study on biohydrogen production based on numerous data extracted from the literature. The researchers involved having applied the generalized multi-scale kinetic model formulated based on the multi-stage growth Hypothesis. Note that the generalized multi-scale kinetic model used in biohydrogen production modeling is an extended version of the original model proposed by [33]. The simplest form of the multi-scale kinetic model is the n-stage single-first order (SFO) model (Equation (9)):

$$G_H = \frac{K_H}{(\tau_1 s + 1)^n (\tau_2 s + 1)}, \quad \tau_1 > \tau_2 > 0 \tag{9}$$

Here, K_H is the cumulative hydrogen production, τ_1 the dominant time constant, τ_2 the non-dominant time constant and n the theoretical number of stages. For the analysis of the biohydrogen process controllability via the critical loop gain, it is essential to reduce the high-order model in Equation (9) to a simple FOPDT model. A simple way to minimise the high-order model to the FOPDT model is by using a simple graphical approach.

The three parameters of FOPDT are estimated as follows (Equation (10)):

$$K_p = K_H, \quad \theta_p = t_5, \quad \tau_p = \frac{t_{95}}{4} \tag{10}$$

Note that t_5 denotes the time the process takes to achieve 5% of the final steady-state value from the beginning of the process. Meanwhile, t_{95} denotes the time the process takes to reach 95% of its final steady-state value. Also, t_{95} represents the settling time, i.e., the time taken for the process to settle at its final steady-state value. For Case Study 1 reported in [4], the SFO model of the biohydrogen production is given by (Equation (11)),

$$G_H = \frac{4050}{(13.996s + 1)^{13} (0.584s + 1)} \tag{11}$$

Based on the visual inspection, the approximated FOPDT model of the biohydrogen production SFO model in Equation (11) is given as follows (Equation (12)):

$$G_H = \frac{4050e^{-108s}}{72.25s + 1} \tag{12}$$

The FOPDT model in Equation (12) shows that the given biohydrogen process is highly delay-dominated ($\theta_p/\tau_p = 1.5$). After the model-order reduction, the CLG value can now be calculated for the biohydrogen fermentation process using the reduced FOPDT model in Equation (12), which gives $CLG = 0.6968$. Notice that the CLG value is

inversely proportional to the dead time as shown in Equation (8). Many researchers have pointed out that the dead-time is one of the well-known factors that can reduce a control system performance as reported, e.g., [34-36]. Note that the models used in this paper for the controllability analyses were obtained from the previous study by [4]. The model reduction rule in Section 2.3 is applied to reduce the high-order models in [4].

3. Results and Discussion

Table 1 shows the calculated values of CLG for the 10 case studies selected from the biohydrogen modeling study in [4]. Besides calculating the values of CLG, the feedback PI control performance (Fig. 1) is also measured via closed-loop simulation in MATLAB Simulink (MATLAB R2015a). The PI control performance is measured using the normalized Integral Absolute Error (IAEn) (Equation (13)):

$$IAEn = \frac{1}{\tau_p} \int_0^{t_s} |e(t)| dt \tag{13}$$

Table 1. Controllability (CLG values) of different biohydrogen production case studies.

Case Study ^a	Microbial Source	Substrate Type	T (°C)	pH	Substrate Conc. (g/L)	$K_{Lcritic}$ (CLG)	IAEn
1	Anaerobic sewage sludge	Glucose	41	5.5	10	0.697	3.333
2	<i>Clostridium pasteurianum</i>	Starch	35	8	15	0.474	4.422
3	<i>Clostridium pasteurianum</i>	Xylose	35	7	20 ^b	0.548	3.881
4	Anaerobic cattle manure sludge	Glucose	55	5	25 ^b	0.799	2.923
5	Anaerobic sewage sludge	Cassava starch	37	6	32 ^b	0.221	7.618
6	Anaerobic paper mill sludge	Starch	35	5	20 ^b	0.214	7.876
13	<i>Clostridium thermocellum</i> and <i>Clostridium thermosaccharolyticum</i>	Cornstalk	55	7.2	10	1.344	1.461
14	<i>Enterobacter aerogenes</i>	Sugarcane molasses	30	-	10	2.006	1.097
15	<i>Enterobacter aerogenes</i>	Sugarcane molasses	30	-	40	0.808	2.939
16	Anaerobic dairy sludge	Sugarcane vinasse	36	5.5	11.6 ^b	4.304	0.641

^a Reference no of case study as in Lim and Nandong [4] ^b Substrate concentration unit is g-COD/L

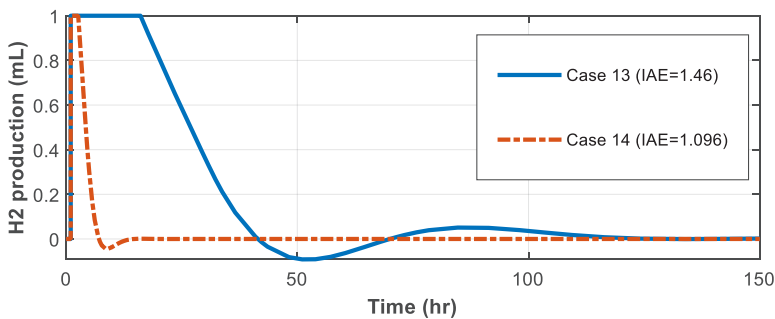


Fig.3. Model-order reduction of biohydrogen production mode for case study 13 and 14

Fig. 3 shows the comparative plots for the case studies 13 and 14. The IAE values of the case studies 13 and 14 are 1.46 and 1.096 respectively. Therefore, a higher IAE value (lower performance) is due to a lower CLG (controllability). For the case study 14 with a lower IAE value (1.096) corresponds to a high CLG (better controllability), which means

that this system becomes easier to control. This is clearly illustrated by Fig. 3 where the DF of case study 14 shows faster and smooth response than that of the case study 13. Keep in mind that the control system performance of the given process is determined by the controllability that is related to various factors such as substrates, temperatures, microbial types, bioreactor design, etc.

For case studies 14 and 15 described using the same microbial type, substrate, and temperature but different fermentation substrate concentrations. For the two cases, an increase of substrate concentration from 10 g/L to 40 g/L substantially reduces the controllability, i.e., $K_{L_{critic}}$ drops from 2 to 0.8. One of the possible reasons for the decrease in controllability is substrate inhibition at a high substrate concentration. Such an inhibitory effect of high substrate concentration is reported in [37, 38] where the hydrogen yield decreased significantly as the hexose concentration increased from 10 g/L to 50 g/L. Bear in mind that some soluble by-products released during fermentation, e.g., ethanol can also inhibit the biohydrogen productivity and yield as reported in [39].

The substrate type shows a strong influence on biohydrogen production controllability. For example, case studies 4, 5, and 6 suggest that the use of glucose (simple sugar) leads to higher process controllability than the use of starch (complex substrate) as the feedstock. The reason for the lower controllability associated with the use of starch is attributable to the need to break down the complex substrate molecules into simple sugars required for cellular uptake and conversion into hydrogen. Thus, there are more steps involved in the digestion of complex substrate than in case of the simple sugar, which leads to a larger dead time in the former fermentation. An increase in the dead time will reduce the upper limit on control performance, i.e., lower controllability.

4. Conclusion

The new controllability criterion for analyzing the impacts of dark fermentation conditions was conducted and the analyses showed that the optimum temperature of dark fermentation depends on the microbial species, either mesophilic or thermophilic types. Hence, temperature plays a vital role in influencing biohydrogen productivity and controllability. A high temperature, e.g., the case study 13 (the operating temperature at 55 °C) can favour better process controllability. However, case study 4 shows low process controllability (less than 1) even though the operating temperature is at 55 °C. For mesophilic fermentation, case study 16 shows the highest process controllability ($K_{L_{critic}} = 4.3$), which operated at 36 °C using the anaerobic dairy sludge. Unfortunately, the maximum volumetric productivity of case study 16 is very low (31.1 mL-H₂/L.hr) compared to other case studies, e.g., case study 6 has a maximum productivity of 441.8 mL-H₂/L.hr but a low CLG value of 0.2.

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