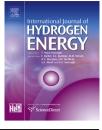


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Kinetic analysis on gaseous and aqueous product formation by mixed anaerobic hydrogen-producing cultures



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ABSTRACT

Anaerobic hydrogen production by mixed cultures is a complex process, and information about its kinetic analysis is sparse. In this study, an integrated approach with the weighted nonlinear least-squares analysis and accelerating genetic algorithm is proposed to evaluate the kinetic parameters of biohydrogen production from a sucrose by mixed anaerobic cultures. The weighted nonlinear least-squares analysis is used to calculate the differences in gaseous and aqueous product concentrations between the predicted and the measured results, while the accelerating genetic algorithm is utilized to optimize the objective function by minimizing the total sum of the squared weighted errors. The kinetic parameters for specific maximum substrate uptake rate, substrate uptake affinity constant, yield coefficient are calculated with this approach, and are validated by the results of the independent experimental results reported in literature. This integrated approach is effective and rapid to estimate the anaerobic hydrogen production kinetics by mixed cultures.

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1. Introduction

Due to the potential to meet the growing demand for energy and emission of pollutant gases from fossil fuels, hydrogen as a clean energy source is considered as a viable source of energy [1–3]. Photosynthetic and fermentative processes are the most commonly used technologies for biological hydrogen production [4–7]. Compared to photosynthetic bacteria, anaerobic acidogenic bacteria produce hydrogen at lower costs. Therefore, anaerobic fermentations of organic wastes to produce hydrogen have been widely demonstrated [8–14]. Dynamic modeling has been extensively used in environmental biotechnology, for process design, determination of optimal operating condition and the control of biological wastewater systems such anaerobic hydrogen-producing process [15–17]. By now, several approaches have been developed to model anaerobic biohydrogen production, i.e., kinetic study using the modified Gompertz equation [9,18], the modified Monod equation [18,19], and IWA anaerobic digestion model (ADM) and their modification versions [17,20–22]. However, the Gompertz equation is an empirical formula and the biochemical reactions in these models are expressed using

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nonlinear equations, resulting in complexity of the models and difficulty to estimate the kinetic parameters for anaerobic hydrogen production [23].

The integrated Monod or Michaelis—Menten equations are found to be effective to evaluate the kinetic parameters through progress curves from a few batch experiments or even one batch experiment [24]. The weighted non-linear least-squares analysis is an approach to minimize the differences between experimental data and model predictions when implicit expressions are used in models [24]. On the other hand, accelerating genetic algorithm (AGA), a parallel iterative and global search algorithm with certain learning ability could be used to perform the minimization in the weighted non-linear least-squares analysis. The AGA method has been successfully used for saving the computational time and accelerating the convergence [25].

In this study, an integrated approach was established to determine the gaseous and aqueous product formation kinetics of mixed anaerobic H_2 -producing cultures through the combination of weighted nonlinear least-squares analysis and AGA. The weighted nonlinear least-squares analysis was used to construct the objective function, and then AGA was used to minimize the objective function and obtain the best fit values of the kinetic parameters. Finally, the experimental data were used to validate this integrated approach. This is an attempt to evaluate the mixed anaerobic hydrogen production kinetics using the weighted nonlinear least-squares analysis and the AGA.

2. Materials and methods

2.1. Model development

Substrate uptake kinetics are usually used as the basis for all intracellular biochemical reactions [20]. When the biomass decay is neglected, the following equation based on ADM is used to describe the substrate uptake [20]:

$$-\frac{\mathrm{dS}_{\mathrm{S}}}{\mathrm{dt}} = k_m \frac{\mathrm{S}_{\mathrm{S}}}{\mathrm{K}_{\mathrm{S}} + \mathrm{S}_{\mathrm{S}}} \mathrm{X} \tag{1}$$

where S_S is the substrate concentration (kg COD/m³), X is the biomass concentration (kg COD/m³), K_S is the substrate uptake affinity constant (kg COD/m³), k_m is the maximum substrate uptake rate (1/d).

Correspondingly, for the anaerobic hydrogen-producing process, microbial growth rate can be described as:

$$\frac{\mathrm{d}X}{\mathrm{d}t} = \mathbf{Y} \cdot \mathbf{k}_m \frac{\mathbf{S}_{\mathrm{S}}}{\mathbf{K}_{\mathrm{S}} + \mathbf{S}_{\mathrm{S}}} \mathbf{X} \tag{2}$$

where Y is the microbial yield coefficient for growth (kg COD/ kg COD).

For the anaerobic hydrogen production process from organic wastes, the main gaseous and aqueous product may be changed significantly due to different operation strategies and environmental conditions. Generally, acetate, propionate, butyrate, lactate, formate and ethanol are the main aqueous products, and hydrogen and carbon dioxide are the main gaseous products. Thus, the product formation rates are expressed as follows:

$$\frac{\mathrm{d}P_{i}}{\mathrm{d}t} = f_{i}(1-Y) \cdot k_{m} \frac{S_{S}}{K_{S}+S_{S}} X \tag{3}$$

where P_i is the concentration of the aqueous and gaseous products (kg COD/m³), f_i represents the yield of products on substrate (kg COD/kg COD), and i represents the aqueous and gaseous products, i.e., acetate, propionate, butyrate, lactate, formate, ethanol and hydrogen, respectively.

According to Eqs. (2) and (3), the biomass and product associated with substrate uptake can be described as:

$$X = X_0 + Y \cdot (S_{s0} - S_s)$$
(4)

$$P_{i} = P_{i0} + f_{i} \cdot (1 - Y) \cdot (S_{S0} - S_{S})$$
(5)

where X_0 is the initial biomass concentration (kg COD/m³), S_{S0} is the initial substrate concentration (kg COD/m³), P_{i0} is the initial product concentration (kg COD/m³).

Eqs. (1), (4) and (5) are combined and integrated to obtain the following equation:

$$\begin{split} t_{ki} = & \frac{1}{k_m} \left\{ \frac{K_S}{X_0 + YS_{s0}} \ln \left(\frac{f_i (1 - Y)X_0 + Y(P_{ki} - P_{i0})}{f_i (1 - Y)K_SS_{s0} - K_S(P_{ki} - P_{i0})} \right) \\ & + \frac{1}{Y} \ln \left(X_0 + \frac{Y(P_{ki} - P_{i0})}{f_i (1 - Y)} \right) - \frac{K_S}{X_0 + YS_{s0}} \ln \left(\frac{X_0}{K_SS_{s0}} \right) \end{split}$$
(6)
$$& - \frac{1}{Y} \ln(X_0) \right\} \end{split}$$

where i represents the aqueous and gaseous products, $i = 1 \sim m$, where *m* is the number of the type of aqueous and gaseous products, i.e., acetate, propionate, butyrate, lactate, formate, ethanol and hydrogen, respectively; k represents the experimental data of the aqueous and gaseous products, $k = 1 \sim n$, where *n* is the number of the experimental data.

The weighted nonlinear least-square analysis is used to construct the objective function and to determine the kinetic parameters through the comparison of the model predictions with observed values of P_{ki} and t_{ki} using the known values. The use of the differences between the predicted and observed t values instead of those of the P values increases the calculated explicitly. Thus, the sum of the squared weighted errors (SSWE) can be obtained using the following equation:

$$SSWE_{i} = \sum_{k=1}^{n} \left[w_{ki} \left(t_{ki}^{obs} - t_{ki}^{pred} \right) \right]^{2}$$
⁽⁷⁾

where w_{ki} is an appropriate weighting factor; t_{ki}^{obs} is the time of the kth experimental data of the ith kind of aqueous and gaseous products; t_{ki}^{pred} is the time value predicted by the model for the measured kth concentration of the ith kind of aqueous and gaseous products.

The differences between the predicted and the observed aqueous and gaseous product values can be estimated by multiplying $t_{ki}^{obs} - t_{ki}^{pred}$ by the local slope of each product curve, $\Delta P_{ki}/\Delta t$. Therefore, the logical weighting factor is the local slope of the product curve:

$$SSWE_{i} = \sum_{k=1}^{n} \left[\frac{\Delta P_{ki}}{\Delta t} \left(t_{ki}^{obs} - t_{ki}^{pred} \right) \right]^{2} \approx \sum_{k=1}^{n} \left(P_{ki}^{obs} - P_{ki}^{pred} \right)^{2}$$
(8)

where P_{ki}^{obs} and P_{ki}^{pre} are the kth measured and predicted concentration of the ith kind of aqueous and gaseous products, respectively.

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