ISSN : 0974 - 7435 Volume 10 Issue 24

2014

BioTechnology

An Indian Journal

FULL PAPER

BTAIJ, 10(24), 2014 [16006-16017]

Mathematical modeling of a vertical tubular external airlift loop bioreactor for biomass production from natural gas

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ABSTRACT

In this study, a mathematical model of a vertical tubular external loop bioreactor (VTELB) was used for biomass production from natural gas. gas-liquid separator (top), bottom of the VTELB were mathematically modeled according to the ideal stirred reactors, and the vertical parts, riser and down-comer sections were modeled tank-in-series without back flow. Mathematical modeling of biological processes cell growth including, mass balance and kinetics Model. The kinetic model Monod is used. In this simulation, a tanks-in-series model describes the mixing characteristics. Acceptable results between the model used and the experimental data obtained.

KEYWORDS

Mathematical modeling; Vertical tubular loop bioreactor; Kinetic model; Microorganism.

INTRODUCTION

Airlift Bioreactor is main part of every biochemical process that is used to provide a controlled environment in order to reach to optimal growth and or forming product in a system. The transport phenomena in these reactors are achieved by pneumatic agitation, and circulation occurs in a defined cyclic pattern through a loop. Airlift bioreactors usage is increasing because of benefits of this type of bioreactor such as simple planning[1,2,3], providing adequate mixing[1,2], low pressure[4], high – velocity transfer mass and temperature[1,3,4], modeling bioreactors is necessary for planning, operation and accumulation speed control in reactors. The modeling of gad-liquid loop reactors was developed for airlift reactors with cultivation of microorganism[36,46]. The model is to include mass transfer, reaction kinetics. The methane is good candidate for biomass production because of its, non-toxicticity, selectivity, volatility[48]. bacteria which are able to utilize methane as the sole carbon and energy sourced have been known since 1906[47]. recently, Yazdian et al. (2009) studied biomass production from methane[41]. In the previous articles, modeling was carried out separately for every section and then equation were solved. The mixing characteristics of gas-liquid reactors are often intermediates between the characteristic of plug-flow and well-mixed flow. In this study each segment of the VTLB, such as bottom, gas-liquid separator (top) was considered based on mixed flow and the vertical parts of the loop, riser and down-comer were modeled according to a tanks-in series[3,4,46].

EXPERIMENTAL

Microorganism and growth medium

The microorganism (Methylomonas spp.) used in this research was isolated from samples obtained from an oil field in Iran.

The carbonless growth medium has been optimized by Yazdian et al. (2005)[39].

Bioreactor

A glass made VTELB with a non-line dissolved methane detector was used for the experiments. The schematic diagram and dimensions of VTELB are shown in Figure 1. And TABLE 1, respectively. The VTELB is made of a riser, liquid –gas separator, down-comer and gas supply distributor. The hydrostatic pressure between riser and down-comer (due to gas flow rates) results in circulation liquid. The length of VTELB was designed in such a way that it could be assembled in a variety of configurations, such that great flexibility in working volume, separator volume and riser diameter. The gas flow rates (methane and oxygen) were measured and controlled by two calibrated rotameters (No.12). Methane and oxygen mixed together, through a sparger (with six holes) located at the bottom of the riser (No.5). Streams of methane and oxygen could be opened and closed by valves (No.13). For safety, gases were passed for a single time through the bioreactor and vented out from the out let (No.14). The O2, CO2, and CH4 content of the exit stream from the separator were continuously monitored by Mettle Toledo In Pro 6800, Figaro- TGS4160, and FigaroTGS2611 sensors, respectively. In addition to water cooled condenser (No.1in Figure 1), one side of the loop was jacketed (No.11) to provide cool exchange capabilities. All experiments were carried out at 30(± 0.5) ◦C. This was done by a temperature loop controller (TLC) placed inside the methane detector (No.8) and connected to an electrical heater (No.3) positioned at the top of the VTELB. PH was measured with an online pH probe. NaOH and HCl were metered in to the loop to control the pH via a peristaltic pump, and the control circuit having adjustable work and pause time.

Measuring methods

The differential hydrostatic pressures of gas–liquid separator, downstream and upstream of the bioreactor were determined by an inverted U-tube manometer and was Analyzed precisely by a high speed camera (Canon power shot S3 IS), too. The differential hydrostatic pressure between two heads of that zone is gas hold up (ε). The overall volumetric mass transfer coefficients ($K_La_{O_2}$ and K_La_{CH4}) were determined by the dynamic gassing out method (Christi, 1989) In addition to methane detector, two probes of dissolved oxygen (DO). (With response time of four seconds for 63% saturation), Values of optical density (DO) were recorded in a spectrophotometer (VARIAN CARRY 50 CONC, Australia at 595 nm).

TABLE 1: Reactor geometry and operational parameters

Descriptions	Unit	Value
Riser diameter, Dr	m	0.03, 0.06, 0.09
Down-comer diameter, D_d	m	0.03
Riser height, H_r	m	2.20
Down -comer height, H_d	m	2.20
Bioreactor height, h_b	m	2.40
Bioreactor volume, V_b	m	33-22
A_r/A_d	#	1, 4, 9
Separator diameter, D_s	m	0.11, 0.18, 0.25
Liquid level in separator, h_s	m	0.10
$S = V_s/V_d$	#	0.61, 1.65, 3.11
Holes size in sparger, D_0	mm	0.1
Number of holes in distributor, N	#	6

MATHEMATICAL MODELING

The mixing models used in most of the previous investigations dealing with loop bioreactors are an axial dispersion model and tanks- in – series[5]. The equation between these two models can be shown by Eq.(1)[3,5]:

$$\frac{1}{Pe} = \frac{b + 1/2}{N} \quad \textbf{(1)}$$

The b is the back flow. If b=0. The peclet (Pe) number should be calculated in order to select the modeling method and to estimate the number of tanks.

In investigation the airlift bioreactor system is modeled by dividing it the riser, gas separator (top section), down-comer, and the bottom section. In this equation, N and Peclet number are parameters used to identify the reactor behavior in the mixed model and axial model, respectively. In this research the bottom section and top section are treated as a well-mixed stage. For the riser and the down-comer is used tanks-in series model of the bioreactor. A tanks-in-series model descries the mixing characteristics. In this model is ignored b (back flow) in each stage.

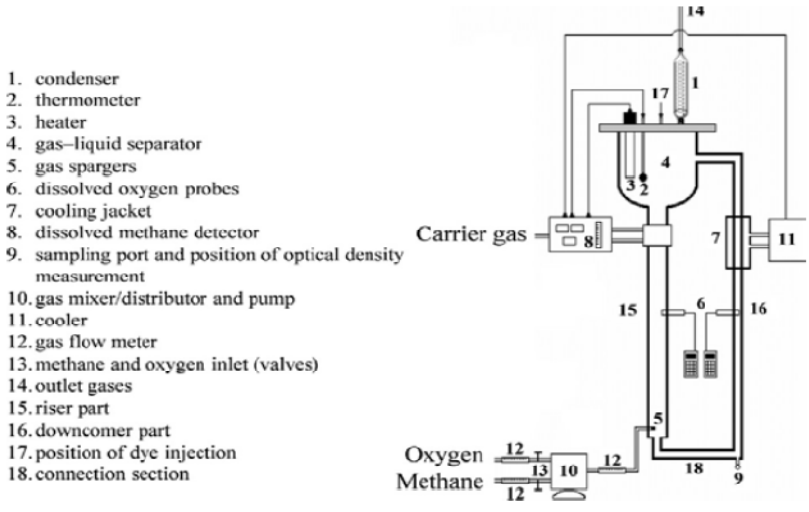


Figure 1: A schematic diagram of external airlift loop bioreactor

The following assumptions were considered for the present modeling:

1. Constant temperature, heat balances in the bioreactor.
2. Ideal behavior of the gas phase.

3. Reaction occurs just in the liquid phase.
4. The gas hold –up in the riser is constant.
5. The variation of hydrostatic pressure is ignored.
6. The Monod kinetics is used for biomass production[7].
7. Mass transfer coefficients constant along riser and down-comer.

Model equation

The equation for pipe sections is[8,9,10]:

$$\frac{\partial C}{\partial t} = -U_L \frac{\partial C}{\partial x} + D_L \frac{\partial^2 C}{\partial x^2} \quad \textbf{(2)}$$

In this research the second term of equation (2) is neglected (without axial dispersion in pipe). For top (gas-liquid separator), and bottom of bioreactor equation is:

$$\frac{\partial C}{\partial t} = \frac{Q_L}{V_{sep}} (C_{sep,in} - C_{sep}) \quad \textbf{(3)}$$

Where $V_{sep9=}$ is the gas-liquid separator volume. In bench scale, the axial dispersion for liquid phase (D_G) is neglected[11].

Gas hold-up correlation

Yazdian et al. (2009) derived an experimental gas hold- up equation in EALB equation (4) which in used in the present model. The amount of gas hold-up changes by variation of gas and liquid velocity[12,13]:

$$\varepsilon_{gr} = 13.19 U_{sgr}^{1.43} (1 + \frac{A_r}{A_d})^{-0.62} (1+s)^{-0.58} (\frac{v_g}{v_{N_2}})^{-0.52} \quad \textbf{(4)}$$

There are linear correlation between ε_{gr} and ε_{gd} in external loop bioreactors[13].
This relationship between ε_{gr} and ε_{gd} of the EALB is estimated[14] by Eq. (5):

$$\varepsilon_{gr} = 0.47 \varepsilon_{gd} \quad \textbf{(5)}$$

Volumetric mass transfer coefficient

$$K_L a_r = 0.097 U_{sgr}^{0.46} (1 + \frac{A_r}{A_d})^{-0.63} (1+S)^{-0.61} (\frac{v_g}{v_{N_2}})^{-0.91} (\frac{D_g}{D_{N_2}})^{1.12} \quad \textbf{(6)}$$

According to Znad et al. (2004), the values of volumetric mass transfer coefficient in riser and down-comer are satisfied in the following equation[15,16]:

$$K_L a_d = \psi K_L a_r \quad \textbf{(7)}$$

An average amount of 0.34 for fraction of $K_L a_d / K_L a_r$ could be extracted from the experimental data Yazdian et al. (2009).

Kinetic model

The intrinsic cell growth kinetic of Methylomonas spp. was assumed oxygen limited and the rate of methane consumption was calculated based on the Monod model. The following relations were

considered for specific cell growth, cell growth dissolved methane, and oxygen consumption rates, respectively[17,18]:

$$\mu = \mu_m \left(\left(\frac{C_M}{K_{S,M} + C_M}\right) \left(\frac{C_O}{K_{S,O} + C_O}\right) \right) \quad \textbf{(8)}$$

$$r_M = \frac{dC_M}{dt} = \frac{-1}{Y_{X/M}} \frac{dC_x}{dt} \quad \textbf{(9)}$$

$$r_X = \frac{dC_X}{dt} = \mu C_X - K_d C_X \quad \textbf{(10)}$$

$$r_O = \frac{dC_O}{dt} = \frac{-1}{Y_{X/O}} \frac{dC_x}{dt} \quad \textbf{(11)}$$

The kinetic parameters were initially estimated based on the experimental results at constant gas and liquid flow rates (2and 20 L/min) and 2 μmol/L biomass concentration. Methane and oxygen yield were estimated by stiochiometric relation.

Material balance in different section of loop airlift bioreactor

The VTLB was modeled by dividing it into vertical flow section (riser), gas separator (top section), down-comer section, and bottom (Figure 1).

The model provides simulation first order partial for biomass concentration, dissolved methane, and oxygen without axial dispersion.

Vertical flow (riser), down-comer (flow) sections[19]:

Mass balance for biomass:

$$\frac{\partial C_X}{\partial t} = -U_L \frac{\partial C_X}{\partial x} + r_X \quad \textbf{(12)}$$

Mass balance for dissolved methane and oxygen:

$$\frac{\partial C_M}{\partial t} = -U_L \frac{\partial C_M}{\partial x} + K_L a_M (C_M^* - C_M) + r_M \quad \textbf{(13)}$$

$$\frac{\partial C_O}{\partial t} = -U_L \frac{\partial C_O}{\partial x} + K_L a_O (C_O^* - C_O) + r_O \quad \textbf{(14)}$$

By Henry's law the saturated methane and oxygen concentration (C_M^*, C_O^*) are calculated:

$$C_M^* = \frac{P}{H_M} y_M \quad \textbf{(15)}$$

$$C_O^* = \frac{P}{H_O} y_O \quad \textbf{(16)}$$

Bottom section and separator (top) section with hold up:

For biomass:

$$\frac{\partial C_{X,i}}{\partial t} = Q_L \frac{C_{X,i-1} - C_{X,i}}{A_i L_i (1-\varepsilon_d)} + r_{X,i} \quad \textbf{(17)}$$

For dissolved methane and oxygen:

$$\frac{\partial C_{M,i}}{\partial t} = Q_L \frac{C_{M,i-1} - C_{M,i}}{A_i L_i (1-\varepsilon_d)} + K_L a_{M,d} (C^*_{M,i} - C_{M,i}) + r_{M,i} \quad \textbf{(18)}$$

$$\frac{\partial C_{O,i}}{\partial t} = Q_L \frac{C_{O,i-1} - C_{O,i}}{A_i L_i (1-\varepsilon_d)} + K_L a_{O,d} (C^*_{O,i} - C_{O,i}) + r_{O,i} \quad \textbf{(19)}$$

(a) Initial and boundary condition

It is assumed that methane, oxygen, and biomass concentrations are the same all through the VTELB for initial condition. Since the process is cyclic, the end point, differentially, is adjacent to the starting point; therefore, the boundary condition in the ending point could be considered equal to the condition in the starting point. The mixed model was used for simulation. PDE equations were applied for dissolved methane and oxygen as well as the biomass concentrations. One initial condition and two boundary conditions were required for PDE equation. one of the boundary conditions was related to the circumstances of initial inlet where the concentrations of compositions were equal to the former point The other boundary condition is the final point of volume control where the concentrations of compositions equal to the former points. Using the mentioned boundary conditions and the mathematical method, e.g., Finite difference, the PDE equations will be converted to ODE equations. Only one condition is required for ODE equations.

In CSTR simulation only one initial condition is required: the system should be at t=0 and every part of the CSTR should be uniform. It means that at t=0, we know all of the concentrations in the tubes and any other vessels; therefore, we start from those parts which are modeled by CSTR and, after Δt, we know the concentrations in the beginning and the end of the tube. Thereafter, we have the boundary conditions for the tube especially for the entrance of each tube. At first, the growth rate of microorganism was low. Therefore, the consumption of methane and oxygen was very low as well. Also, we assumed that the dissolved methane and oxygen concentrations were the same at initial time. Therefore, it was assumed that methane, oxygen, and biomass concentrations were identical through the hole of VTELB for initial conditions.

TABLE 2: The hydrodynamic parameters in VTELB

Q_L(L/S)	Q_G(L/S)	$\varepsilon_{G,u}$	$\varepsilon_{G,d}$	$K_L a_O$ (s^{-1})	$K_L a_M$ (s^{-1})	D_L (cm^2/s)	D_G (dm^2/s)
12	1.2	0.324	0.189	0.0121	0.0104	0	0

(b) Numerical solution

The model equations were solved numerically by the method of lines in MATLAB software. In each section for PDEs solution in each section which considered mixed flow a set of N first order equations and three components (biomass, dissolved methane, and oxygen) were supplied. In order to tune the kinetic parameters, a set of ordinary (ODE) and partial first-ordinary equations (PDEs) were solved making use of MATLAB.

RESULT AND DISCUSSION

The model parameters

TABLE 2 shows the hydrodynamics and mass transfer parameters. For modeling, the hydrodynamic parameters, in the VTELB, were obtained experimentally. The values of volumetric methane and oxygen mass transfer coefficients and gas hold-up were calculated by Esq. (4-7). TABLE 2 shows the hydrodynamics and mass transfer parameters. Moreover, the kinetic parameters are explained in TABLE 3. The parameters were optimized by the Nelder-mead optimization algorithm. Derived K_d is very low; therefore the death of microorganism is not considered in this model practically. This model predicted Biomass growth until 35h, and it is not for after growth phase. So the yield coefficient of

biomass on methane $Y_{X/M}$ obtains extraordinary high. This parameter was not obtained from theoretical or operational predictions. It is estimated of proposed model. It is difficult to decide on single model for the quantitative description of the process in with range of initial substrate concentration[49].

Simulation of biomass production

Figure 2 shows the model fitting to the experimental data at constant gas and liquid flow rates, biomass concentration, and oxygen fraction, 1.2 L/min, 12 L/min, 1mg/L, and 0.25, respectively. The biomass concentration exponentially increased with cultivation time. The graphs in this figure were obtained based on the suggested kinetic model. Figure 2 shows that oxygen concentration reached almost zero after 28h. The concentration of methane also became constant at the same time. However, it shows increase in cell concentrations after 20h from the beginning of reaction. In Figure 3, from 27 to 32 h, the biomass concentration increases fast, because it is resulted from methane and oxygen gases injection.

TABLE 3: The tuned kinetic parameters obtained by minimizing sum of squares relative error

Kinetic parameters	Value
$\mu_{max}(h^{-1})$	0.212
$K_{S,M}(\mu M)$	0.00058
$K_{S,O}(\mu M)$	0.00003
$Y_{X/M}(g/g)$	101.54
$Y_{X/O}(g/g)$	1.0002
$K_d(h^{-1})$	0.322
$K_l(g/L)$	0.00025

Validation of biomass production at various oxygen fractions

Figure 3(a–c) shows the reduction of dissolved methane and oxygen along with time. It is observed that the values of oxygen fraction in all cases are reduced to the same amount. While the methane concentration in gas phase was increased from 0.002 to 0.006, the amount of oxygen was decreased. Therefore, there was an equilibrium condition between gas and liquid concentrations for a certain gas. This is when the initial concentration of methane changed to the thermodynamic concentration (based on Henry's law). Also, if we wish to use the same initial methane concentration in Figure 3(a–c), we are to utilize the inert gas, which is not economic for applications in industry. In all the experiments, the growth rate was restricted by oxygen limiting. Utilizing oxygen by microorganism resulted in the consumption of methane. The amount of methane consumption was dependent on $Y_{X/M}$ and K_La. Figure 3 (a–c) shows the comparison of the results of the model with the experimental data at respective constant gas and liquid flow rates and biomass concentration1.2 L/min, 12 L/min, 1mg/L, and oxygen fraction from 0.002 to 0.006 (if the air fraction is 0.2, 0.5, 0.75) the remaining is 0.004–0.008 for methane) using the fitted kinetic parameters. This figure shows the model is able to optimize the gas fractions for biomass production and these predicted optimum fraction of gases are consistent with the experimental observation of Yazdian et al. (2009)[41]. 32 h, the amount of biomass production reached to2.25 mg/L only when oxygen fractions are 0.002–0.004 and 0.006. In order to investigate the biomass production in terms of optimum gas fraction, Yazdian et al. (2009) designed five experiments[5] where gas stream was a mixture of oxygen and methane. In the cited study, optical density for every stream was plotted versus gas fraction. It should be mentioned that the time scale of the experiments was arranged based on the microorganism growth rate. Therefore, we observed three stages of growth during 35 h. After 35 h, the VTLB was restricted by oxygen limiting and not methane concentration. The values of R-squared are between 99.4 and 99.8 for dissolved methane and oxygen, and biomass production. While the methane concentration in gas phase was increased from 0.2 to 0.6, the amount of oxygen was decreased. Therefore, there was an equilibrium condition between gas and liquid concentrations for a

certain gas. This is when the initial concentration of methane changed to the thermodynamic concentration (based on Henry's law).

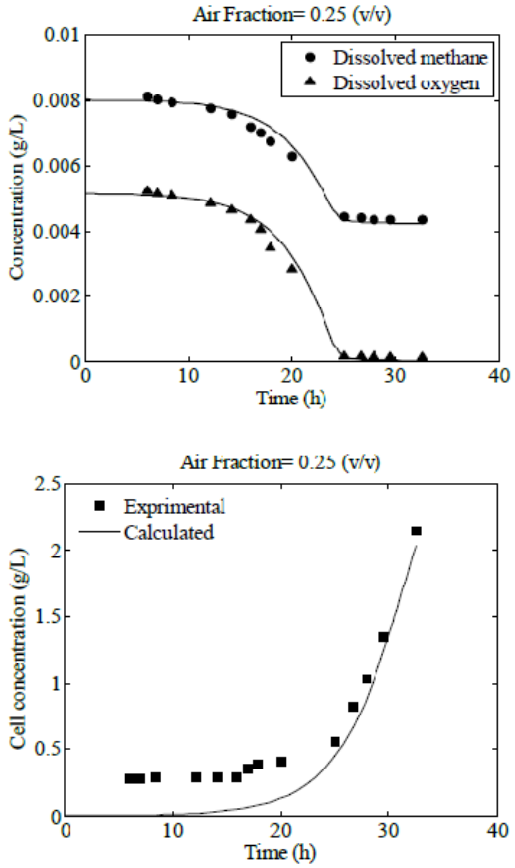
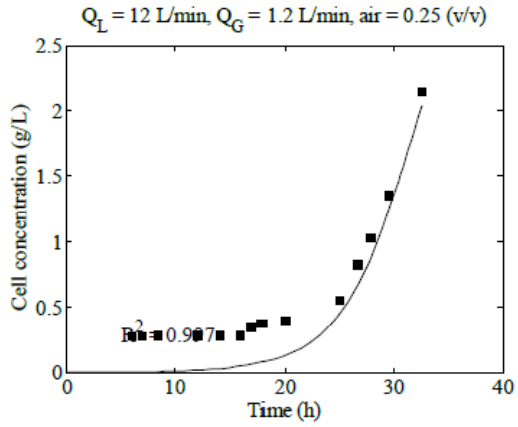
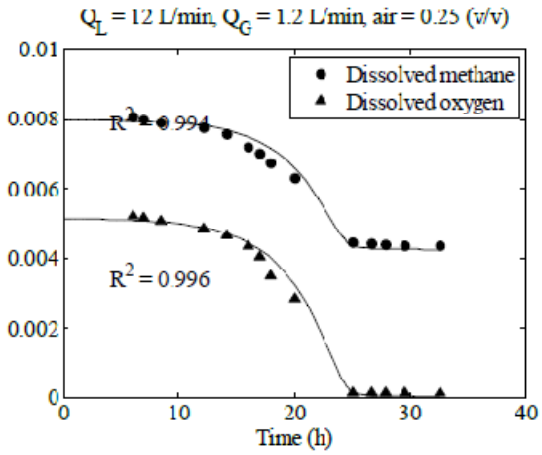
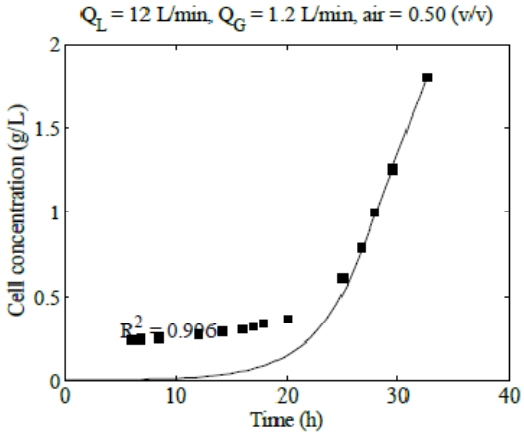
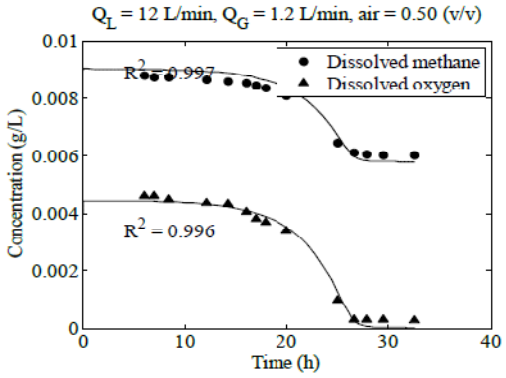


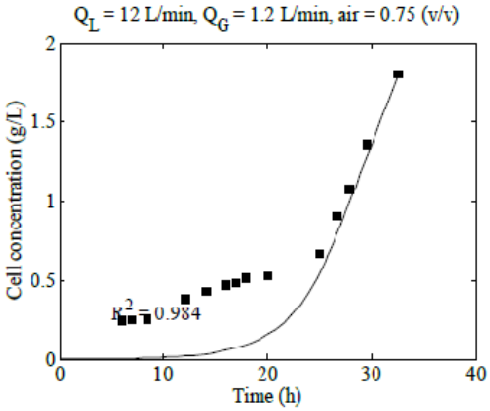
Figure 2: Comparison between the model and experimental concentrations versus time. Gas volumetric flow rate = 1.2 L/min, liquid volumetric flow rate = 12 L/min, and oxygen fraction = 0.25. The points show experimental data and the lines are obtained from the model after first static mixer




Q_L = 12 L/min, Q_G = 1.2 L/min, air = 0.25 (v/v)
Dissolved methane
Dissolved oxygen
R^2 = 0.994
R^2 = 0.996
Time (h)


Q_L = 12 L/min, Q_G = 1.2 L/min, air = 0.50 (v/v)
Cell concentration (g/L)
R^2 = 0.986
Time (h)


Q_L = 12 L/min, Q_G = 1.2 L/min, air = 0.50 (v/v)
Dissolved methane
Dissolved oxygen
R^2 = 0.997
R^2 = 0.996
Concentration (g/L)
Time (h)


Q_L = 12 L/min, Q_G = 1.2 L/min, air = 0.75 (v/v)
Cell concentration (g/L)
R^2 = 0.984
Time (h)

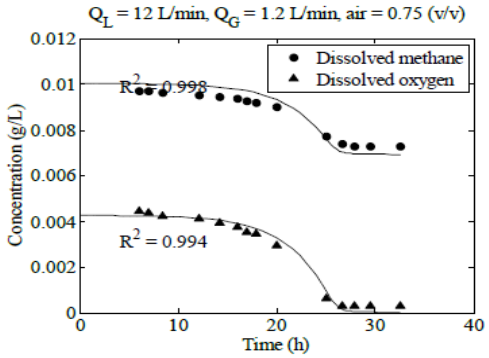


Figure 3: Validation of the model for biomass concentration, dissolved methane and dissolved oxygen versus time. Gas volumetric flow rate = 1.2 L/min and liquid volumetric flow rate = 12 L/min. The points show experimental data and the lines are obtained from the model after first static mixer; (A) oxygen fractions 0.25 (v/v); (B) oxygen fractions 0.5 (v/v); (C) oxygen fractions 0.75 (v/v)

CONCLUSION

In this research, the mathematical modeling of biomass production from natural gas was produced in a vertical tubular loop bioreactor (VTLB) in the vicinity of Methylomonos spp. The present model has shown the authority of predicting the dynamic behavior of biological process in the loop tubular bioreactors with gas circulation. The profile of biomass concentration, methane, and dissolved oxygen can be determined at various liquid flow rates and methane to air ratios. This is valuable information for scaling up for biomass production process from methane.

Notation

A_r/A_d	riser to down-comer cross sectional a ratio, dimensionless
C*	saturated oxygen concentration, ppm
C_L	concentration, ppm
D_0	holes size in sparger, mm
D_d	down-comer diameter, m
D_g	diffusion coefficient, m^2/s
D_r	riser diameter, m
D_s	separator diameter, m
h_b	bioreactor height, m
h_s	liquid level in separator, m
H_d	down-comer height, m
H_r	riser height, m
$K_L a_{CH4}$	volumetric methane mass transfer coefficient, s^{-1}
$K_L a_{O2}$	volumetric oxygen mass transfer coefficient, s^{-1}
N	number of holes in distributor, dimensionless
OD	optical density, dimensionless
S	separator to down-comer volume ratio, dimension- less
T_c	circulation time, s
t_m	mixing time, s
U_{sgr}	superficial gas velocity in riser, m/s
V	bioreactor volume, L
V_L	liquid velocity, m/s

Greek letters

μ	Specific growth rate, 1/s
μ_m	maximum specific growth rate, 1/s
ε	Gas hold- up, dimensionless

Subscripts

d	down-comer
i	number of point

G gas phase
L liquid phase
M Methane
O oxygen
R Riser
X biomass

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