



Dynamics and oxygen transfer of a novel vertical tubular biological reactor for wastewater treatment

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ABSTRACT

Dynamics and oxygen transfer of a novel vertical tubular biological reactor (VTBR) for wastewater treatment were investigated in this paper. It was showed that the dissolved oxygen concentration (DO) in VTBR is higher than that in the conventional bubble column. When the ratio of gas and liquid flow rates was greater than 6.44, there were no phenomena of deficiency oxygen in all reactors. The volume oxygen transfer coefficient (k_1a) was between 0.005 and 0.025 1/s. The multi-stage series CSTR and PFR model were developed to describe the dynamics of VTBR. It was revealed that the PFR model was proper to describe the dynamics of VTBR of which maximum error was only 25%. The industrial effluents from Dalian Bangchui Island Beer Company were utilized to verify the two models. It was suggested that when the ratio of gas–liquid was greater than 6.44, the removal efficiency of COD could be obtained more than 80%.

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1. Introduction

In recent years, with the rapid industrialization and the expansions of the chemical industry, a high number of polluting xenobiotic compounds has been released into environment [1,2]. Thus, the characteristics and composition of wastewater became more complex. Biological treatment has been accepted as one of the most feasible, eco-friendly and cost-effective options for the treatment of pollutants [3,4]. In general, bio-filter, suspended growth bioreactor, packed bed bioreactor and rotating rope bioreactor systems have been widely used for the biological treatment of such compounds [5,6]. However, these bioreactors come with some inheriting limitations such as poor oxygen transfer, low resistance to shock loadings, etc., which restricted the use of conventional reactors at higher concentration and loadings. Hence, research efforts have been made towards the development of novel bioreactors recently, which can overcome the disadvantages associated with conventional systems. Mohammed et al. evaluated the membrane bioreactor for treating municipal wastewater at different operating conditions [5]. Felix et al. review the bioreactor scale-up and oxygen transfer rate in microbial processes in detail [6]. Raul et al. developed two-phase partitioning bioreactors for treatment of volatile organic compounds, etc. [7]. Among these researches, various multi-phase bioreactors have been developed to improve the treatment efficiency, which have been widely applied in a

variety of biotechnological processes for wastewater treatment [8–10].

VTBR is a novel and recently developed liquid–gas co-current up-flow immobilized film wastewater treatment system, which was modified according to the principle of deep-well [11]. VTBR, a Chinese patent with No. CN1084831A, was firstly proposed by Zhou Jiti in 1992. As a novel bioreactor, VTBR possessed some advantages and specific characteristics concluding lower energy cost, higher efficiency and easy to operation, etc. while in detail: (1) the time for gas and liquid contact is enough to improve the oxygen transfer rate; (2) the pressure in VTBR can reach 2–3 atom, which is much higher than that in the conventional bioreactor, therefore, the maxim concentration of wastewater could increase to 5000 mg/L; (3) although VTBR comes from deep-well, its technical characteristics are far from deep-well because of the inputs; (4) As for the operation conditions, VTBR could be operated as aerobic art flow, anaerobic art flow and A/O art flow, and so on; (5) the surplus sludge is very low due to the immobilized biofilm operation; (6) considering the characteristics mentioned above, VTBR can be used for various treatment processes. From then on, some series processes of aerobic and anaerobic wastewater treatment devices have been developed from VTBR, which was used for high concentration wastewater treatment, nitrogen and phosphorus removal and sludge digestion, and so on [11,12]. Nowadays, VTBR has been successfully used in field treatments of many kinds of wastewater in China including effluents from chemical, mechanic, dyes and food industries. Although VTBR has been applied in the field treatments of various kinds of wastewater, there have been some fundamental studies to be performed.

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Nomenclature

c_i	dissolved concentration of gas component in water (mg/L)
c_{si}	saturated dissolved concentration of gas component in water (mg/L)
v_{LS}	apparent velocity of the liquid (m/s)
k_{La}	volumetric liquid mass transfer coefficient (1/s)
c_0	initial dissolved concentration of gas component in water (mg/L)
β_L	volume proportion of liquid component
H	column height (m)
RO	utilization ratio of oxygen (mg/(Lh))
F	flow rate of wastewater (L/h)
S	unused dissolved oxygen (mg/L)
D	dilution rate (h^{-1})
b	microbial specific attenuation coefficient (h^{-1})
β	oxygen demand per unit mass of microorganisms
Y_g	microbial actual increment
S_b	concentration in the liquid phase (mg/L)
S_s	concentration on the surface of biofilm (mg/L)
μ_m	maximal specific growth rate (h^{-1})
V	volume of the liquid in a single VTBR reactor (L)
S_0	dissolved oxygen of inflow soluble substrate (mg/L)
q'_m	liquid flow rate (L/h)
A_s	filter effective sectional area (m^2)
K_s	saturation constant (mg/L)
D_a	equivalent diameter of reaction column (m)
X_0	initial concentration of microorganism (mg/L)
L	column length (m)

As for any bioreactors, some parameters are the most important for the reactors design. The mass transfer coefficients between the different phases together with reaction dynamics are important design parameters of gas–liquid–solid reactors for both chemical and biochemical engineering applications [13–16]. It has been reported that there have been considerable amount of work which was done for the purpose of mass transfer from gas phase to the liquid phase and different types of gas liquid contacting devices, such as wetted wall towers, bubble columns, etc. [17–19]. Hence, it is necessary to apply proper method to obtain the mass transfer coefficient for any novel multi-phase reactor, such as VTBR. Meanwhile, it is essential to acquire kinetic parameters for the operation of bioreactors optimization. Furthermore, the kinetic model should be verified by the field treatment process.

Therefore, the present paper was undertaken to study the oxygen transfer and dynamics of a co-current gas–liquid reactor for the field application experimentally. The volumetric oxygen transfer coefficient (k_{La}) was estimated by the least squares method, and the reaction dynamics were verified by the real wastewater effluents.

2. Materials and methods

2.1. Reactor design and operation

A schematic diagram of the experimental setup of VTBR is shown in Fig. 1(a), and the packing was fibrous filler (Fig. 1(b)). The VTBR consists of 10 reactors in sequential each with the volume of 12.449 L, and HRT about 2–4 h per column. And the inoculated activated sludge were grown adhere to the fibrous filler with concentration of 5–10 g/L in each reactor. And reactors joined by the baffles were 2 m in height and 90 mm inner diameter, through

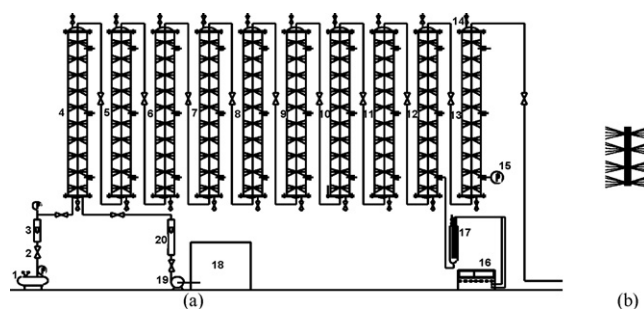


Fig. 1. Schematic description of the VTBR used in the experiment, (a) the structure of the VTBR; (b) the structure of the inputs: (1) DE-0.036/2-type air compressor; (2) control valve; (3) LZB-3WB-type gas meter; (4)–(13) 1–10-level reaction column; (14) release valve; (15) U-tube differential pressure meter; (16) BY1941A-type digital multi meter; (17) dissolved oxygen electrode device; (18) raw water flume; (19) 2J-W13/1.5-type sink-plunger metering pump; (20) LZB-type liquid flow meter; (21) emptying valve.

which flow patterns as well as the type of dispersion can be observed.

Two running models were adapted in this design, of which the cold-flow model means the wastewater does not contain the pollutants, and the hot-flow model means the system adapt the real wastewater. The temperature was set at 25 °C for both cold-model and hot-model. For cold-flow model, the distilled tap-water was used for experiments with initial DO = 0. While for the hot-flow model, the wastewater used was taken from Dalian Bangchui Islands Beer Company. And the pH values of both test water were adjusted to 7.0.

The liquid flow rate of wastewater was between 3 and 6 L h⁻¹ and gas flow rates ranged from 0.1 to 0.5 L min⁻¹. The value of gas injection pressure was controlled as 0.125 MPa. The gas used in the experiments is atmospheric air pumped by a ventilator, which was higher than atmosphere. In the column, the gas and the liquid have a co-current up-flow circulation. The liquid flow rate was measured by a flow meter located at the tank outlet and controlled by a valve. And the air flow rate was measured by a flow meter.

2.2. Analytical methods

The samples were collected at the end of each column, of which COD_{Cr} and BOD₅ was determined by standard methods described elsewhere [20]. It was necessary to note that each sample point was equal to corresponding grade of VTBR.

Volumetric mass transfer coefficient k_{La} was measured by a dynamic technique. The dissolved oxygen concentration in the liquid was measured by a fast responding oxygen electrode manufactured by our lab. And the relationship between DO and voltage should be calculated by the experimental equation: $DO = -40.65 + 0.133U$. And the electrode response time was less than $1/k_{La}$, which was proved not affecting k_{La} value. And the volumetric oxygen transfer coefficient (k_{La}) was computed by the minimization of least squares method.

2.3. Mathematical model of oxygen transfer

In the cold-flow model system, oxygen was not consumed by any chemical reaction. Therefore, the oxygen balance could be described by the following equation:

$$\frac{\partial c_i}{\partial t} + \frac{d}{dx}(v_{LS}c_i) = k_{La}(c_{si} - c_i) \quad (1)$$

Initial condition : $t = 0, c_i = c_0$

Boundary conditions : $c_i(t, x = 0) = c_i \left(t - \frac{\beta_L H}{\nu_{LS}}, x = \beta_L H \right)$

And the following dimensionless variables were inputted:

$$\begin{aligned} c &= \frac{c_{si} - c_i}{c_{si} - c_0} \\ \tau &= \frac{t}{\beta_L H / \nu_{LS}} \\ X &= \frac{x}{\beta_L H} \end{aligned} \quad (2)$$

Then Eq. (1) was as follows:

$$\begin{aligned} \frac{\partial c}{\partial \tau} + \frac{\partial c}{\partial X} + mc &= 0 \\ \tau = 0, c &= 1 \\ c(\tau, 0) &= c[(\tau - 1), 1] \end{aligned} \quad (3)$$

where

$$m = \frac{k_L a \beta_L H}{\nu_{LS}} \quad (4)$$

Therefore, the DO can be determined from the following equation:

$$C_m = \frac{1}{m} \exp\left(-\frac{x+\tau}{2}m\right) \left[\exp\left(\frac{m}{2}\right) - 1 \right] \quad (5)$$

2.4. Dynamic model description for the reactor

In the hot-flow model system, two models were used in this paper as CSTR (model A) and PFR (model B). Under each model, the assumptions were made to develop the kinetic model for the VTBR.

2.4.1. Model A

Generally, CSTR culture systems consist of several separate CSTR in sequential. In order to develop the dynamic model, the following assumptions were given:

- (1) limited and dissolved substrates in each CSTR conform to the Monod equation;
- (2) kinetic parameters of each CSTR were kept consistent;
- (3) components of the limited and dissolved substrates were constant but only with concentration changes;
- (4) each CSTR was operated under steady state conditions.

Under these general assumptions the CSTR model was developed to describe reaction dynamics of the VTBR system. In this case, the utilization efficiency of oxygen is given by the following equation (the deduction process presents in Appendix A):

$$RO_{(n)} = \frac{F[(S_{(n-1)} - S_{(n)})D_{(n)} + b + D_{(n)}\beta_{(n)}Y_g] + X_{(n-1)}[D_{(n)}(\beta_{(n-1)} - \beta_{(n)}) + \beta_{(n-1)}b]}{D_{(n)} + b} \quad (6)$$

2.4.2. Model B

As we all know that flat flowing with the characteristics as:

- (1) there is no velocity distribution on the cross section which is vertical with flow direction;
- (2) there is no back mixing on the flow direction;
- (3) when the flow left the reactors, the HRT of each fluid particle is the same.

From these characteristics, the mass balance equation can be developed from theoretical analysis as following:

$$\frac{dS_b}{dX} + \left[\frac{\mu_m V}{FL} \left(\frac{X_0}{Y} + S_0 - S_b \right) + \left(\frac{q''_m A_S}{FL} \right) \eta \right] \frac{S_b}{K_S + S_b} = 0 \quad (7)$$

The boundary conditions were:

$$X = 0, S_b = S_0 \quad (8)$$

Make $\bar{K} = K_S/S_b$ here, and then the effective coefficient η was defined as following (the deduction process presents in Appendix B):

$$\eta = \frac{(\bar{K} + 1) \left[1 - \bar{K} - D_a + \sqrt{(1 - \bar{K} - D_a)^2 + 4\bar{K}} \right]}{(1 + \bar{K} - D_a) + \sqrt{(1 - \bar{K} - D_a)^2 + 4\bar{K}}} \quad (9)$$

3. Results and discussion

3.1. Dissolved oxygen in the VTBR

It is well known that the volume oxygen transfer coefficient ($k_L a$) is one of the most important parameters, which affects the design and operation of the unit. Many factors could affect $k_L a$, including air flow rate, air pressure, temperature, vessel geometry and fluid characteristics, etc. In this paper, the variations of dissolved oxygen concentration (DO) in VTBR for cold-flow model and hot-flow model were investigated. The dynamics of dissolved oxygen between the first and second injection was determined. And the response of the electrode was rapid enough to determine. Time course of DO in the cold-flow model system with 50 Lh^{-1} of liquid flow rate was shown in Fig. 2(a). It was investigated that the

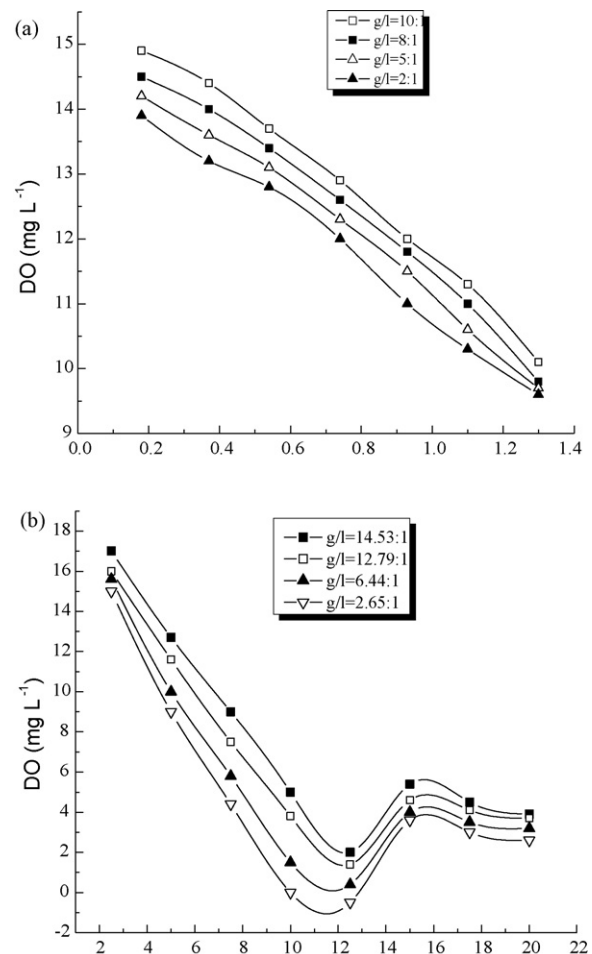


Fig. 2. Time course of DO changes in VTBR, (a) cold-flow model, (□) $g/l=10:1$; (■) $g/l=8:1$; (△) $g/l=5:1$; (▲) $g/l=2:1$; (b) hot-flow model, (□) $g/l=12.79:1$; (■) $g/l=14.53:1$; (▽) $g/l=2.65:1$; (▲) $g/l=6.44:1$.

DO decreased with time increased under the same g/l ratio, which meant DO in the former reactors was greater than that in the following reactors.

It was proved that the changes of DO were different in both systems. In the cold-flow model system, the DO of each reactor was determined by cleaning water. The concentration of DO was mainly determined by the pressure inside of the reactor because there was no respiration of microorganisms. Therefore, DO increased with pressure increasing. Also, when the liquid flow rate was increased, the DO was improved to some extent with the increasing of gas and liquid ratio (g/l).

Time course of DO in the hot-flow model system with 5 L h^{-1} of liquid flow rate was shown in Fig. 2(b). The oxygen consumption rate (SOUR) of microorganisms during the first 3 reactors was more rapid than that of other reactors. The results indicated that the DO in the reactor changes in the different tendency when the microorganisms participate in the reaction. As mentioned above, in the cold-flow model system, the DO was determined only by pressure. However, in the hot-flow model system, DO depended not only on the pressure but also the SOUR. From Fig. 2(b), it could be easily observed that DO decreased quickly between the first and the third reactor. In the fourth and the fifth reactor, the DO also decreased and then turned to stable. As for the sixth to tenth reactor, DO was mainly determined by pressure according to the characteristics of VTBR, which lead to a little increase of dissolved oxygen concentration (Fig. 2(b)).

It was also showed that the DO was improved when g/l ratio was increased, which was the similarity to the results in the cold-flow model system. When g/l ratio was of 2.65, the fifth reactor exhibited anoxic conditions. It could be explained that the SOUR value was more than the oxygen transfer in this reactor. However, it was obvious that the dissolved oxygen was all above zero in the reactors when g/l ratio reached 6.44, which suggested that the oxygen transfer was more than the SOUR. Therefore, it was necessary to take into account that the g/l ratio should be higher than 6.44 when VTBR was in the field application.

3.2. Oxygen transfer

It was indicated that the value of $k_L a$ increased with the increase of liquid flow rate for a given gas and liquid flow rates (Fig. 3). When the ratio of gas and liquid flow rates was greater than 6.44, there were no phenomena of deficiency oxygen in all reactors. In order to determine the volume oxygen transfer coefficient ($k_L a$), a plug flow model describing gas–liquid two-phase up-flow in VTBR was proposed and the mathematics solution was given. By comparison of dynamic measurement and mathematics solution, the

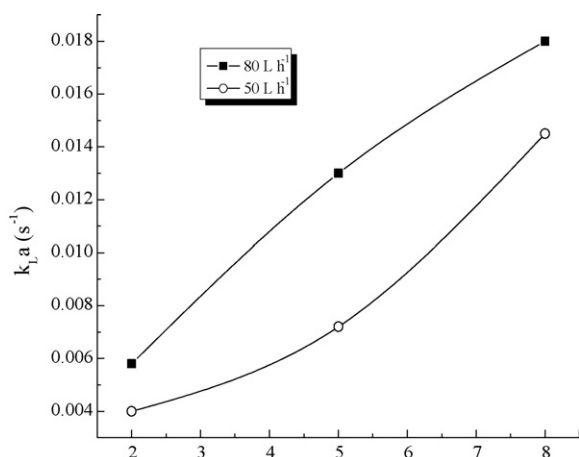


Fig. 3. Influence of different g/l on $k_L a$. (■) 80 L h^{-1} ; (□) 50 L h^{-1} .

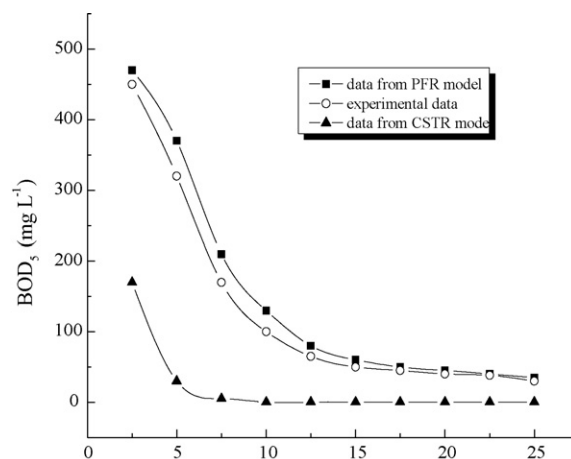


Fig. 4. Comparison of the data between two models and the experiments. (■) Data from PFR model; (□) experiment data; (▲) data from CSTR model.

volume oxygen transfer coefficient was obtained. The $k_L a$ of VTBR was between 0.005 and 0.025 $1/\text{s}$, which changed with liquid flow rate and the g/l ratio (Fig. 3). As reported previously, the $k_L a$ was about 0.045–0.019 $1/\text{s}$, which was a little lower (nearly 10–20%) than that of reporting here [21], which should be explained that the soft input in VTBR could improve the oxygen transfer.

3.3. Dynamic model development of VTBR

From the view of biology, bio-treatment of wastewater could be considered as continuous processes in which the microorganisms were largely generated. This paper analyzed and discussed the kinetics for VTBR. Two flow models of the ideal reactors were proposed, i.e. multi-stage series CSTR model and PFR model without the resistance of mass transfer outside. When the ratio of gas and liquid flow rates was greater than 6.44, the removal efficiency of COD was greater than 80%. This study showed that all experimental data were within 25% of predictions from PFR model without the resistance of mass transfer outside.

Comparison of the results from model calculation and experiments under g/l 6.44 was shown in Fig. 4. It was observed that the kinetic parameters of VTBR located between CSTR and PFR model. And the experiment value was closer to the data from the PFR model. Although the value from PFR model was larger than the experiment data, it was still consistent with the real data. As shown in Table 1, the PFR model was proper to describe the dynamics of VTBR with its maximum error only being 25%.

3.4. Application of VTBR for industrial effluents

As described above, the key parameter $k_L a$ was determined and the PFR model was proved to be fit for describing the VTBR sys-

Table 1
Data comparison of PFR model and experiments.

No.	BOD ₅ (mg/L) from PFR model	BOD ₅ (mg/L) from experiments	Relative error (%)
1	468	449.7	4.07
2	370	314.09	17.80
3	210	176.39	19.05
4	135	126.06	7.09
5	85	71.98	18.09
6	65	52.84	23.01
7	52	43.69	19.02
8	45	38.70	16.28
9	40	37.03	8.02
10	32	26.22	22.04

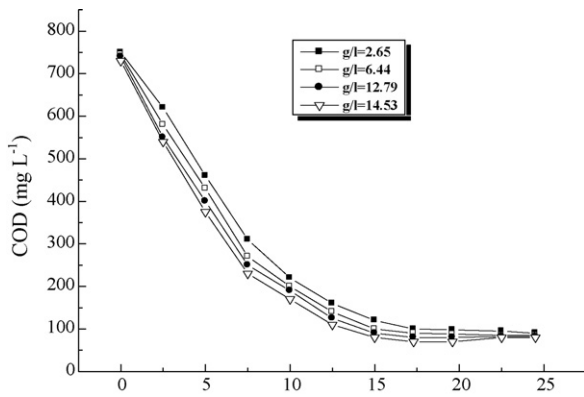


Fig. 5. Time course of COD removal in VTBR for real wastewater treatment. (■) $g/l=2.65:1$; (□) $g/l=6.44:1$; (▲) $g/l=12.79:1$; (△) $g/l=14.53:1$.

tem. However, it is still necessary to investigate whether the VTBR could be used for real wastewater treatment. Therefore, the effluent from Dalian Bancuidao Island was collected to verify the treatment ability. Firstly, the VTBR was under the acclimatization stage with 738.05 mg/L of the influent COD. After 15 days acclimatization, the biofilm was formed with the predominant Protozoa. In this period, the COD removal reached nearly 90.83% (data not shown), which indicated that the VTBR possessed high efficiency for such wastewater treatment.

During stable operation stage, COD value of each level was determined (Fig. 5). With the g/l ratio increased, COD of effluent decreased, which indicated that large gas amount would enhance the COD removal. However, when the gas quantity reached some extent, COD removal kept stable as shown in Fig. 5. It was showed that tend of line 3 ($g/l=12.79$) and line 4 ($g/l=14.53$) was almost consistent, which exhibited that it would largely increased the power consumption if the gas quantity was continuously increased. From Fig. 5, it was also obvious that the g/l ratio played the key role in the COD removal within the short HRT, which suggested that oxygen transfer was important for the target compounds degradation.

As shown in Fig. 5, it suggested that the COD removal was nearly 89% under enough HRT during the whole operation of the VTBR. When g/l was 14.53, it needed 11 h for 80% COD removal. While g/l was 2.65, it needed more time (nearly 13 h) to obtain the same COD removal. It was concluded that relatively few series of VTBR with high g/l ratio would obtain the same removal efficiency. However, the g/l ratio was not the sole determinants for COD removal in VTBR.

4. Conclusions

In conclusion, the most important parameter affecting the design and operation of VTBR is the volume oxygen transfer coefficient $k_L a$. Therefore, this study was firstly performed to determine the $k_L a$, which between 0.005–0.025 1/s. And it was proved that multi-stage series CSTR and PFR model could be used to describe the dynamics of VTBR. According to the oxygen transfer and dynamic analysis, VTBR was proved to be efficient for industrial effluents such as some effluents from Dalian Bangchui Island Beer Company, of which the COD removal was more than 80%. Although VTBR has been widely used in the wastewater treatment in the industry of China, the basic characteristics such as hydrodynamics mass transfer should also be deeply studied. As a novel bioreactor, VTBR is considered to be the predominant sewage treatment equipment in the future.

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Appendix A.

According to the assumption mentioned above, the following equation can be obtained:

$$\mu_n = D_n \left(1 - \frac{X_{V(n-1)}}{X_{V(n)}} \right) + \gamma + b$$

$$X_{V(n)} = \frac{D_n [X_{V(n-1)} + Y_g (S_{(n-1)} - S_{(n)})]}{D_n + b + \gamma}$$

$$X_{d(n)} = \frac{D_n X_{d(n-1)} + \gamma X_{V(n)}}{D_n + b}$$

$$X_{(n)} = \frac{D_n [X_{(n-1)} + Y_g (S_{(n-1)} - S_{(n)})]}{D_n + b}$$

$$[\mu_m - (D_n + \gamma + b)] S_{(n)}^2 - \left[\mu_m \left(\frac{X_{V(n-1)}}{Y_g} + S_{(n-1)} \right) + (K_S - S_{(n-1)}) (D_n + \gamma + b) \right] S_{(n)} + S_{(n-1)} K_S (D_n + \gamma + b) = 0$$

$$v_{(n)} = \left(\frac{D_n + b}{D_n + \gamma + b} \right) \left(\frac{X_{V(n-1)} + Y_g (S_{(n-1)} - S_{(n)})}{X_{V(n)} + Y_g (S_{(n-1)} - S_{(n)})} \right)$$

$$RO_{(n)} = \frac{F[(S_{(n-1)} - S_{(n)}) (D_n + b + D_n \beta_{(n)} Y_g) + X_{(n-1)} \{D_n (\beta_{(n-1)} - \beta_{(n)}) + \beta_{(n-1)} b\}]}{D_n + b}$$

Appendix B.

The equation of mass transfer rate was as following:

$$N_S = K_L (S_b - S_S)$$

According to the Monod equation, it could obtain the following equation:

$$-r_S'' = \frac{q_m'' S_S}{K_S + S_S}$$

Under steady state, $N_S = -r_S''$, therefore, the following equation could be got:

$$S_S = \frac{S_b - K_S - (q_m''/K_L) + \sqrt{(S_b - K_S - (q_m''/K_L))^2 + 4K_S S_b}}{2}$$

Considering the η using in the chemical reaction engineering, it could get the following equation:

$$\eta = \frac{r_S''}{r_b''} = \frac{q_m'' S_S / (K_S + S_S)}{q_m'' S_b / (K_S + S_b)}$$

And these dimensionless variables were introduced:

$$\bar{S} = \frac{S_S}{S_b}$$

$$\bar{K} = \frac{K_S}{S_b}$$

$$D_a = \frac{q_m''}{K_L S_b}$$

According to all the equation mentioned above, the η could determine as:

$$\eta = \frac{(\bar{K} + 1) \left[1 - \bar{K} - D_a + \sqrt{(1 - \bar{K} - D_a)^2 + 4\bar{K}} \right]}{(1 + \bar{K} - D_a) + \sqrt{(1 - \bar{K} - D_a)^2 + 4\bar{K}}}$$

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