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Kinetic characteristics of biological simultaneous anaerobic sulfide and nitrite removal

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ABSTRACT

The kinetic characteristics of biological simultaneous anaerobic sulfide and nitrite removal were studied in UASB reactor. The results showed that the process was capable of tolerating high influent sulfide and nitrite concentrations of 880 mg S/L and 252.78 mg N /L, respectively. It could hold a high sulfide and nitrite removal loading rates of 13.45 kg S/m^3/d and 2.26 kg N/m^3/d, respectively. Half-order model, first-order model, and second-order model were applied to predict the performance of the process, which indicated that second-order substrate removal model was best to predict the substrate removal in the bioreactor. Monod model, Michaelis–Menten model and Stover–Kincannon model were used to calculate the kinetic constants, which showed that modified Stover–Kincannon model and modified Michaelis–Menten model were ideal for determining maximum sulfide and nitrite removal rates, respectively.

Keywords: Anaerobic sulfide and nitrite removal; Kinetic constants; Substrate removal model; UASB reactor

1. Introduction

Sulfide-containing wastewaters are generated by a number of industries, such as petrochemical plants, tanneries, viscose rayon factories [1]. The produced sulfide would be toxic to human health; hence, it should be removed from wastewater before its discharge into the environment [2]. A variety of physicochemical and biological processes have been used to treat sulfide-containing waste streams [3]. Compared with physicochemical technologies, biological processes are cost-effective that operate at prevailing environmental conditions without any requirement for expensive chemicals and catalysts [4].

It has been shown that some bacteria-like *T. denitrificans* can oxidize sulfide to elemental sulfur simultaneously reducing nitrate or nitrite to dinitrogen [5]. For such reasons, nitrate or nitrite can be used as electron accepters to oxidize sulfide under anaerobic conditions. Short-cut nitrification process is a novel biological nitrogen removal process, which has been put into practical use. So it can serve as a nitrite source for the anaerobic sulfide oxidation process (ASO process). According to standard Gibbs free energy change, Nitrite-ASO process (Eq. (1)) takes place more easily than Nitrate-ASO process (Eq. (2)).

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So far, however, few researches have been carried out on Nitrite-ASO process [6–8].

$$\mathrm{HS} + 2/3\mathrm{NO}_2^- + 5/3\mathrm{H}^+ \rightarrow S + 1/3\mathrm{N}_2 + 4/3\mathrm{H}_2\mathrm{O} \quad \Delta Gm^\theta = 305.7\mathrm{kJ/mol} \tag{1}$$

$$\mathrm{HS} + 2/5\mathrm{NO}_3^- + 7/5\mathrm{H}^+ \rightarrow S + 1/5\mathrm{N}_2 + 6/5\mathrm{H}_2\mathrm{O} \quad \Delta Gm^\theta = 252.7\mathrm{kJ/mol} \tag{2}$$

Process modeling is an accepted route for describing and predicting the performance of biological treatment systems [9]. Many models exist in the wastewater treatment literature [10]. Wang et al. [11] established a kinetic model for simultaneous sulfide, nitrate, and acetate removal process based on Activated Sludge Model No. 1 (ASM1). Apparently, simplified models involving limited variables suit for practical application, in comparison with complicated ones which need numerous inputs and assumptions [12]. Monod model, Stover–Kincannon model, first-order substrate removal model and second-order substrate removal model are some of the popular models in biological treatment systems [13–15]. Half-order model and modified Michaelis–Menten model were used to predict substrate removal rates on desulfurization process [16, 17]. To date, however, very few researches have been focused on the substrate removal kinetics of Nitrite-ASO process based on simplified models. In this study, the Nitrite-ASO process was operated at different conditions, and different mathematical models were applied to study the process kinetic characteristics.

2. Materials and methods

2.1. Inoculum and enrichment of microbial communities

Inoculum was collected from the anaerobic methanogenic reactor operated at Dengta wastewater treatment plant located in Hangzhou City of China. Its total solids (TS) and volatile suspended solids (VSS) were 95.03 and 68.68 g/L, respectively, with VSS/TS ratio of 0.72. The simultaneous anaerobic sulfide and nitrite removal reactor was operated under lithoautotrophic conditions where sulfide was used as electron donor, and nitrite was employed as electron acceptor to accomplish denitrification. For initial one month, the reactor was fed with synthetic wastewater in order to acclimatize the bacteria to the new substrates and to enrich the functional bacteria.

2.2. Synthetic wastewater

The reactor was fed with synthetic influent containing $\mathrm{NaHCO_3}$, $\mathrm{MgCl_2}$, $\mathrm{KH_2PO_4}$, (1 g/L each), $\mathrm{(NH_4)_2SO_4}$ (0.24 g/L) and trace element solution (1 mL/L). The composition of trace element solution used was according to Mahmood et al. [6]. The sulfide/sulfur and nitrite/nitrogen concentrations were added in the form of sodium sulfide ($\mathrm{Na_2S{\cdot}9H_2O}$) and sodium nitrite ($\mathrm{NaNO_2}$), respectively, with their concentrations varying according to the experiments.

2.3. Anaerobic sulfide oxidizing reactor

The simultaneous anaerobic sulfide and nitrite removal reactor was an upflow reactor with biomass retention and was operated in a continuous mode. The reactor was made of perspex with a working volume of 1.3 L. The synthetic influent was pumped through a peristaltic pump from a 10 L influent vessel to the reactor. A recycling pump was used to mix the influent (substrate) and sludge (biocatalyst) well and hence to decrease possible substrate inhibition. The ratio of recycling flow to the influent flow was set at 2.5–3.0. The temperature was controlled between 29 and 31℃.

2.4. Experimental design

Initially, keeping hydraulic residence time (HRT) constant, the volumetric loading rate was enhanced through increasing the influent substrate concentration. The influent sulfide and nitrite concentrations were increased according to the stoichiometry of chemical reaction (Eq. (1)). Once the influent substrate concentrations were changed, the reactors were operated under the circumstances until the effluent quality became stable. If the substrate removal was less than 50%, the reactor was supposed to have reached its maximal loading rate, so further increase in influent concentrations was stopped.

After terminating the concentration experiment, the optimal influent substrate concentrations were kept constant, the volumetric loading rate was raised through decreasing the HRTs. When substrate removal became less than 50%, the HRT experiment was stopped. The influent pH was maintained at 7.0 ± 0.1 throughout the experiment.

2.5. Analytical procedures

Influent and effluent nitrite/nitrogen, pH, and sulfide/sulfur were analyzed during the operation of

ASO reactor. Nitrite/nitrogen (NO_2^--N) was analyzed through colorimetric method on daily basis [18]. The sulfide/sulfur was determined by iodometric method [18]. The pH was determined following standard method [18]. A three-point calibration of pH meter was performed daily. TS concentration was determined according to gravimetric method at 103℃, and volatile solids were analyzed through gravimetric method at 550℃ [18].

3. Kinetics approaches

3.1. Substrate removal model

3.1.1. Half-order substrate removal model

Koenig and Liu [16] described autotrophic denitrification rates in upflow sulfur packed-bed reactors by half-order kinetic model. The equation was expressed as follows:

$$S_e^{1/2} = S_i^{1/2} - \frac{1}{2}k_{(1/2)}\theta \tag{3}$$

3.1.2. First-order substrate removal model

Assuming that first-order kinetics was applied in the reactor and the reactor was under pseudo-steady-state conditions, the change rate of substrate concentration is negligible. The equation of first-order substrate removal model can be given as follows:

$$\frac{S_i - S_e}{\theta} = k_1 S_e \tag{4}$$

3.1.3. Second-order substrate removal model

After integration and linearization, the equation of a second-order substrate removal model is expressed as follows [19].

$$\frac{S_i\theta}{S_i - S_e} = \theta + \frac{S_i}{k_2 X} \tag{5}$$

If $a = S_i/k_2X$ and $E = (S_i{-}S_e)/S_i$, Eq. (5) will be modified as follows:

$$\frac{\theta}{E} = a + b\theta \tag{6}$$

3.2. Kinetic constants evaluation

3.2.1. Monod model

Monod model has been widely used to describe the process kinetics of anaerobic digesters [20]. The equation can be written as follows:

$$\frac{1}{R} = \frac{K_s}{R_{\max}}\frac{1}{S_e} + \frac{1}{R_{\max}} \tag{7}$$

3.2.2. Modified Michaelis–Menten model

Michaelis–Menten model is a popular model in wastewater treatment, too. Ma et al. applied the modified Michaelis–Menten model to determine H_2S removal rate in waste gases by an activated carbon bioreactor [17].

$$\frac{1}{R} = \frac{K_s}{R_{\max}}\frac{1}{S_{\ln}} + \frac{1}{R_{\max}} \tag{8}$$

$S_{\ln}$ is logarithmic mean concentration of substrate in the influent and effluent, which can be defined as followed:

$$S_{\ln} = \frac{S_0 - S_e}{\ln(S_0/S_e)} \tag{9}$$

3.2.3. Modified Stover–Kincannon model

Stover–Kincannon model is one of the most widely used mathematical models to determine kinetic constant in immobilized systems. However, this model also has been applied for granular sludge and flocculent sludge systems [15, 20].

The Stover–Kincannon model is described as below:

$$\left(\frac{\mathrm{dS}}{\mathrm{d}t}\right)^{-1} = \frac{1}{r} = \frac{V}{Q(S_i - S_e)} = \frac{K_B}{U_{\max}}\frac{V}{QS_i} + \frac{1}{U_{\max}} \tag{10}$$

4. Results and discussion

4.1. Substrate removal performance

The performance of the reactor at different influent concentrations and HRTs were shown in Tables 1 and 2. Keeping HRT constant, when the influent concentrations of sulfide and nitrite were increased to 880 mg S/L and 252.68 mg N/L, respectively, the maximum removal rate of sulfide and nitrite were 3.97 kg S/m^3/d and 0.65 kg N/m^3/d, respectively.

Table 1
The reactor performance at different influent concentrations

Influent concentration		Effluent concentration		Substrate removal efficiency		Substrate removal rate	
Sulfide (mg S/L)	Nitrite (mg N/L)	Sulfide (mg S/L)	Nitrite (mg N/L)	Sulfide (%)	Nitrite (%)	Sulfide (kg S/m^3/d)	Nitrite (kg N/m^3/d)
160	43.43	0.89	0.02	99.44	99.95	0.78	0.21
220	70.23	1.02	0.30	99.54	99.57	1.07	0.34
280	80.95	1.27	0.50	99.55	99.38	1.36	0.39
340	106.97	0.79	0.93	99.77	99.13	1.65	0.52
400	114.58	1.55	1.18	99.61	98.97	1.94	0.55
460	132.3	1.86	2.53	99.60	98.09	2.23	0.63
520	158.00	2.50	25.57	99.52	83.82	2.52	0.65
580	173.84	17.15	44.13	97.04	74.61	2.75	0.63
640	191.49	20.69	69.03	96.77	63.95	3.02	0.60
700	201.35	22.75	81.34	96.75	59.60	3.30	0.59
760	221.39	29.45	98.94	96.13	55.31	3.56	0.60
820	237.1	43.21	112.13	94.73	52.71	3.79	0.61
880	252.68	65.80	133.30	92.52	47.25	3.97	0.58

Keeping the concentrations of influent sulfide and nitrite constant around 460 mg S/L and 132.3 mg N/L, respectively, when HRT was shortened from 10 to 1.5 h, the maximum removal rates for sulfide and nitrite were as high as 13.45 kg S/m^3/d and 2.26 kg N/m^3/d, respectively. However, low HRT caused great loss of activated sludge, and the reactor was hard to continue working.

4.2. Substrate removal model

4.2.1. Half-order substrate removal model

The value of $k_{(1/2)}$ was obtained from the slop by plotting $S_e{}^{1/2}$ vs. θ in Eq. (3). The values of $k_{(1/2)}$ for sulfide and nitrite were 1.02 and 1.69 mg$^{1/2}$/L$^{1/2}$/h, respectively. Fig. 1(A) shows the correlation coefficients of sulfide and nitrite were 0.5764 and 0.7142, respectively.

From Eq. (3), the formulas for predicting effluent substrate concentrations were given in Eqs. (11) and (12).

$$S_{e,\text{sulfide}} = (S_{i,\text{sulfide}}^{1/2} - 0.51\theta)^2 \tag{11}$$

$$S_{e,\text{nitrite}} = (S_{i,\text{nitrite}}^{1/2} - 0.84\theta)^2 \tag{12}$$

4.2.2. First-order substrate removal model

Fig. 1(B) shows the first-order substrate removal model applied to simultaneous anaerobic sulfide and nitrite removal process. The value of k_1 was calculated

Table 2
The reactor performance at different HRTs

	Influent concentration		Effluent concentration		Substrate removal efficiency		Substrate removal rate	
HRT (h)	Sulfide (mg S/L)	Nitrite (mg N/L)	Sulfide (mg S/L)	Nitrite (mg N/L)	Sulfide (%)	Nitrite (%)	Sulfide (kg S/m^3/d)	Nitrite (kg N/m^3/d)
9.84	465.00	132.46	1.86	2.53	99.60	98.09	2.23	0.63
7.44	453.33	135.69	1.36	4.03	99.70	97.03	2.96	0.85
5.04	455.56	131.13	1.64	4.97	99.64	96.21	4.37	1.20
4.08	465.00	135.84	1.86	5.95	99.60	95.62	5.39	1.53
3.60	461.03	133.35	6.27	27.75	98.64	79.19	6.05	1.41
3.12	459.72	136.45	13.24	41.25	97.12	69.77	6.87	1.46
2.00	460.36	134.55	20.21	64.29	95.61	52.22	10.55	1.69
1.50	460.02	139.73	39.70	69.11	91.37	50.54	13.45	2.26

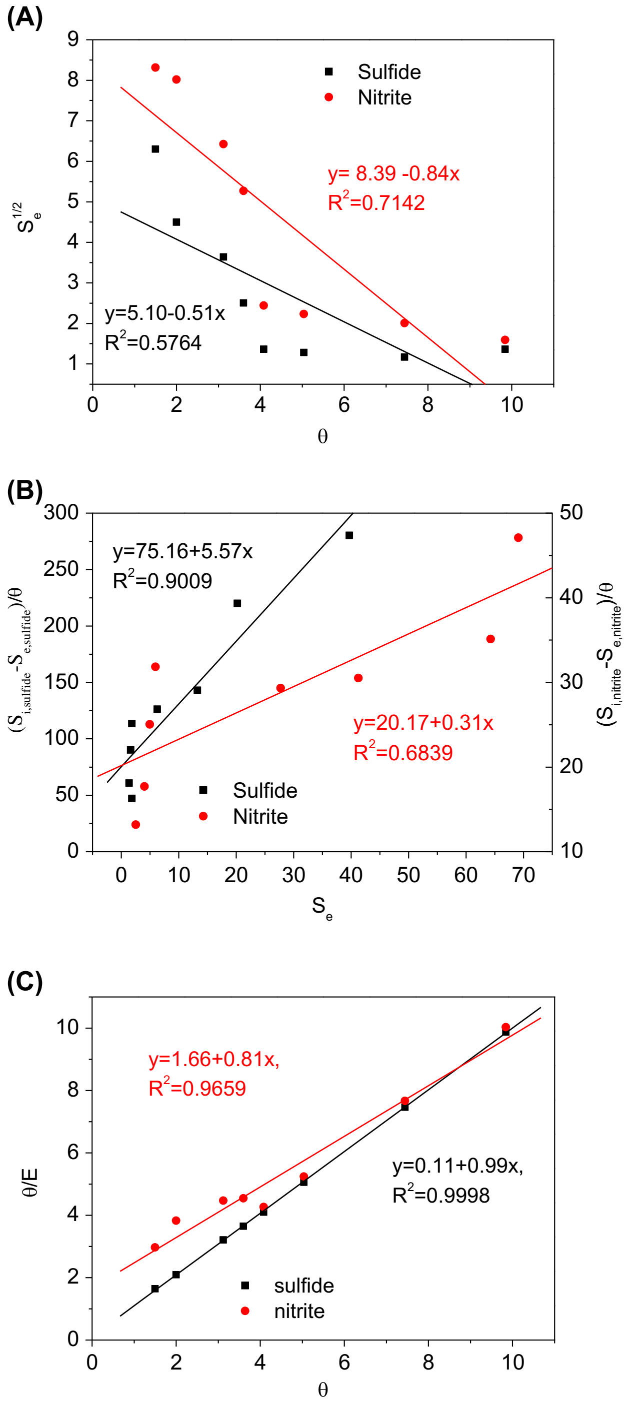

Fig. 1. Fitter curves for different substrate removal model. (A) Half-order substrate removal model. (B) First-order substrate removal model. (C) Second-order substrate removal model. The regression equations and related correlation coefficients were indicated in the figures.

from the slope by plotting $(S_i – S_e)/\theta$ vs. S_e. The values of k_1 for sulfide and nitrite were 5.57 and 0.31 per hour, respectively. The correlation coefficients of sulfide and nitrite were, 0.9009 and 0.6839, respectively.

From Eq. (4), the formulas for predicting effluent substrate concentrations were given in Eqs. (13) and (14).

$$S_{e,\mathrm{sulfide}} = \frac{S_{i,\mathrm{sulfide}}}{5.57\theta + 1} \tag{13}$$

$$S_{e,\mathrm{nitrite}} = \frac{S_{i,\mathrm{nitrite}}}{0.31\theta + 1} \tag{14}$$

4.2.3. Second-order substrate removal model

Fig. 1(C) shows the fitter curves for second-order substrate removal model. By plotting Eq. (6), the coefficients of second-order substrate removal model were determined. The values of a and b for sulfide were calculated to be 0.11 and 0.99 per hour, respectively; the values of a and b for nitrite were calculated to be 1.66 and 0.81 per hour, respectively. The correlation coefficients for sulfide and nitrite were 0.9998 and 0.9659, respectively.

From Eq. (6), the formulas for predicting effluent substrate concentrations were given in Eqs. (15) and (16).

$$S_{e,\mathrm{sulfide}} = S_{i,\mathrm{sulfide}}\left(1 - \frac{\theta}{0.11 + 0.99\theta}\right) \tag{15}$$

$$S_{e,\mathrm{nitrite}} = S_{i,\mathrm{nitrite}}\left(1 - \frac{\theta}{1.66 + 0.81\theta}\right) \tag{16}$$

4.3. Kinetic constants determination

4.3.1. Monod model

Fig. 2(A) showed that a double-reciprocal plot of $1/r$ against $1/S_e$, which would yield $R_{\max}$ and K_s values. The K_s and $R_{\max}$ constants for sulfide were calculated as 3.16 mg S/L and 4.39 kg S/m^3/d; the K_s and $R_{\max}$ constants for nitrite were calculated as 0.25 mg N/L and 0.62 kg N/m^3/d. It is suggested that maximum sulfide and nitrite removal rates in the UASB reactor were 4.39 kg S/m^3/d and 0.62 kg N/m^3/d, respectively. The correlation coefficients for sulfide and nitrite were 0.8602 and 0.9426, respectively.

4.3.2. Modified Michaelis–Menten model

Fig. 2(B) shows Michaelis–Menten model applied to Nitrite-ASO process. According to Eq. (8), $K_s/R_{\max}$ is the slope and $1/R_{\max}$ is the intercept point. The K_s

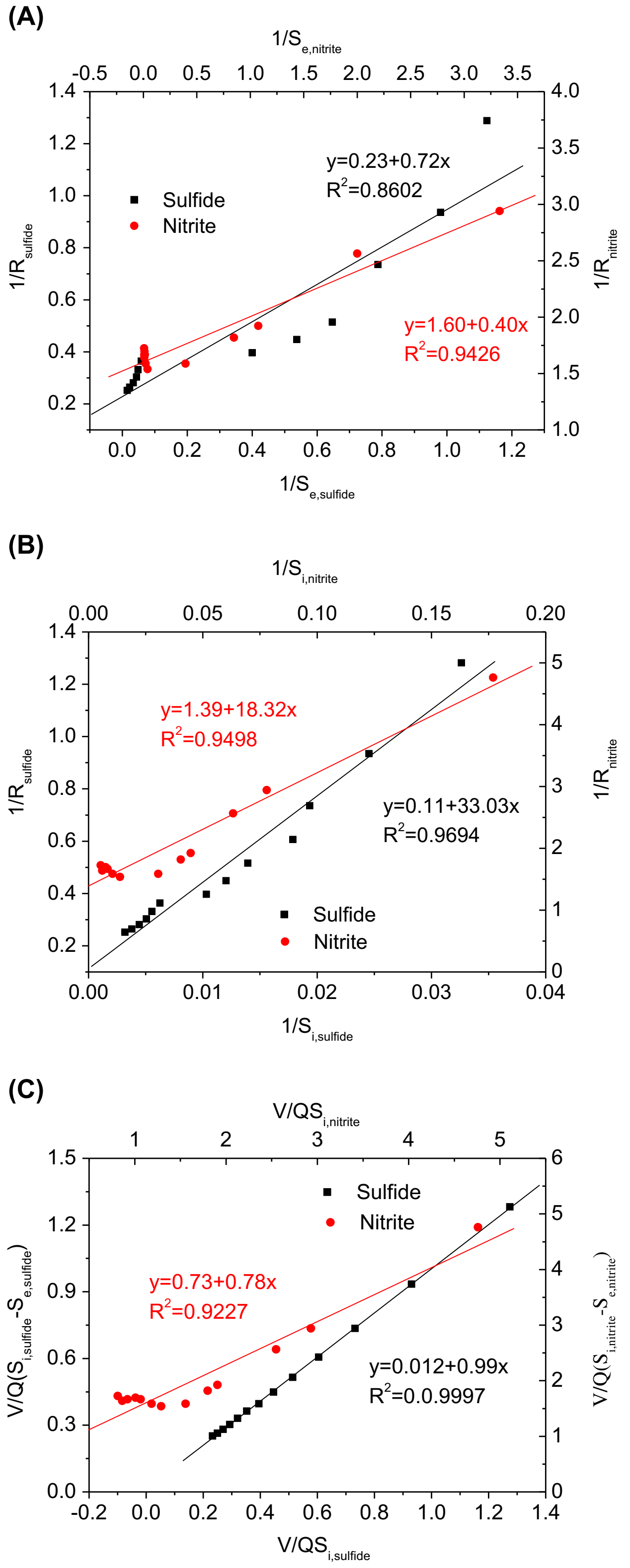

Fig. 2. Fitter curves for kinetic constants determination. (A) Monod model. (B) Michaelis-Menten model. (C) Modified Stover-Kincannon model. The regression equations and related correlation coefficients were indicated in the figures.

and $R_{\max}$ constants for sulfide were calculated as 293.66 mg S/L and 8.89 kg S/m^3/d; the K_s and $R_{\max}$ constants for nitrite were calculated as 13.15 mg N/L and 0.72 kg N/m^3/d. It indicated that maximum sulfide and nitrite removal rates in the UASB reactor were 8.89 kg S/m^3/d and 0.72 kg N/m^3/d, respectively. The correlation coefficients for sulfide and nitrite were 0.9694 and 0.9498, respectively.

4.3.3. Modified Stover–Kincannon model

Fig. 2(C) shows fitter curves for modified Stover–Kincannon model. According to Eq. (10), $K_B/U_{\max}$ is the slope and $1/U_{\max}$ is the intercept point. The K_B and $U_{\max}$ constants for sulfide were calculated as 85.43 and 86.13 kg S/m^3/d; the K_B and $U_{\max}$ constants for nitrite were calculated as 1.06 and 1.37 kg N/m^3/d. It showed that maximum sulfide and nitrite removal rates in the UASB reactor were 86.13 kg S/m^3/d and 1.37 kg N/m^3/d, respectively. The correlation coefficients for sulfide and nitrite were 0.9997 and 0.9227, respectively.

From Eq. (10), the formulas for predicting effluent substrate concentrations were given in Eqs. (17) and (18).

$$S_{e,\text{sulfide}} = S_{i,\text{sulfide}} - \frac{86.13\, S_{i,\text{sulfide}}}{85.43 + S_{i,\text{sulfide}}/\theta} \tag{17}$$

$$S_{e,\text{nitrite}} = S_{i,\text{nitrite}} - \frac{1.37\, S_{i,\text{nitrite}}}{1.06 + S_{i,\text{nitrite}}/\theta} \tag{18}$$

4.4. Model evaluation

When the correlation coefficients were compared, it showed that second-order substrate removal model was more appropriate than the other two substrate removal models to predict the performance of Nitrite-ASO process. Furthermore, model validity was evaluated by comparing the experimental data with predicted values calculated from different models by Eqs. (11)–(18) (Fig. 3). It clearly indicated that the effluent substrate concentrations predicted by half-order and Stover–Kincannon models were much higher than actual data. The predicted sulfide concentrations in the effluent by first-order model showed good correlations with actual data, while the predicted nitrite concentrations in the effluent did not. Only the predicted values calculated by second-order model nearly coincided with the actual line. Hence, second-order substrate removal model was most suitable for predicting the performance of simultaneous sulfide and nitrite removal process.

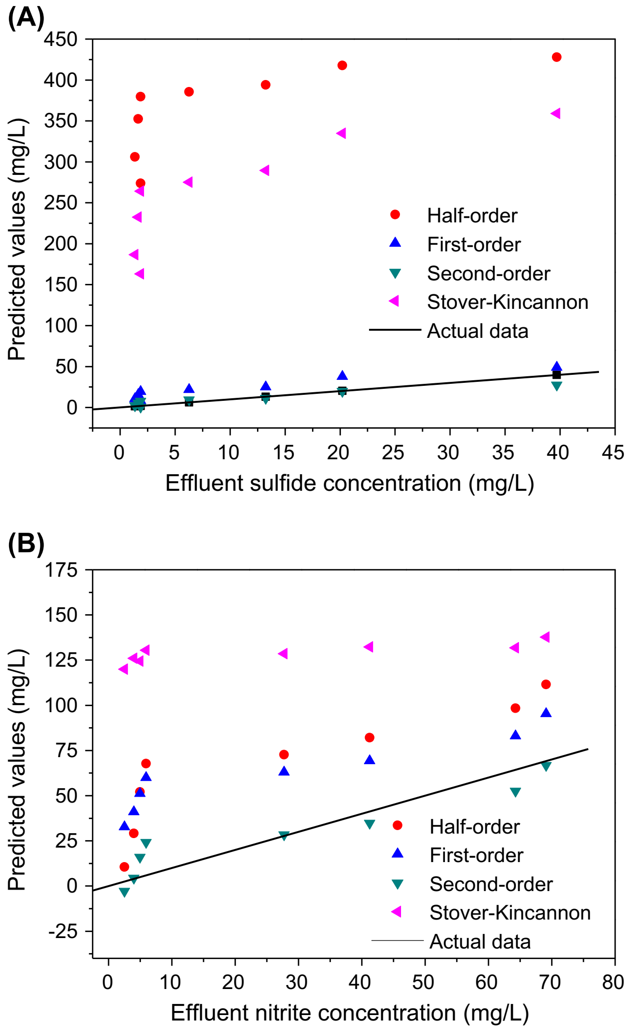

Fig. 3. Relationship between predicted values and effluent substrate concentrations. (A) Effluent sulfide concentration. (B) Effluent nitrite concentration.

There is a great disparity among the maximum substrate removal rates calculated by different models. The maximum substrate removal rates by Stover–Kincannon model were much higher than those by Monod and Michaelis–Menten models, especially maximum sulfide removal rate. When the correlation coefficients were compared, it showed that Stover–Kincannon model was more appropriate than Monod and Michaelis–Menten models to determine maximum sulfide removal rate. Mahmood et al. [6] operated a laboratory-scale reactor to remove sulfide and nitrite simultaneously, whose sulfide removal rate could achieve 13.82 kg S/m^3/d. In this study, the actual maximum removal rates of sulfide was 13.45 kg S/m^3/d, which were only 15.62% of the calculated $U_{\max}$ by Stover–Kincannon model. The reactor stopped operating due to great loss of activated sludge caused by low HRT. If the reactor configuration was modified and the settleability of activated sludge was improved, the calculated $U_{\max}$ could be achieved.

Comparing the correlation coefficients, it showed that Michaelis–Menten model was more appropriate than Monod and Stover–Kincannon models to determine maximum nitrite removal rate. In this study, the actual maximum removal rate of nitrite was 0.65 kg N/m^3/d, which was 90.28% of the calculated $R_{\max}$. It was suggested that the reactor had performed most of denitrification capability.

According to Monod model, the half-saturation concentrations of sulfide and nitrite were 3.16 mg S/L and 0.25 mg N/L, respectively. According to reports in the literature, the half-saturation concentrations of sulfide and nitrite were 0.63–1.99 mg S/L and 0.21 mg N/L [21,22]. The results were in general agreement with the values reported in literature.

5. Conclusion

In simultaneous anaerobic sulfide and nitrite removal process, the maximum influent sulfide and nitrite concentrations were 25.78 mg S/L and 880 mg N/L, respectively. The maximum sulfide and nitrite removal rates were 13.45 kg S/m^3/d and 2.26 kg N/m^3/d, respectively.

Six kinetic models were applied to the process. Second-order substrate removal model was more appropriate to predict the performance of simultaneous sulfide and nitrite removal process. Stover–Kincannon model and Michaelis–Menten model gave higher correlation coefficients of 0.9997 and 0.9498, respectively, which were ideal for determining maximum sulfide and nitrite removal rates, respectively. These models may be used in the design of biological simultaneous anaerobic sulfide and nitrite removal.

Nomenclature

a — S_i/k_2X (per hour)
b — constant for Grau second-order model (dimensionless)
dS/dt — substrate removal rate (kg/m^3/d)
E — substrate removal efficiency (%)
$k_{(1/2)}$ — half-order reaction rate constant
k_1 — first-order kinetic constant (per hour)
k_2 — second-order kinetic constant (per hour)
K_B — saturation value constant (kg/m^3/d)

K_s — half saturation concentration (mg/L)
Q — inflow rate (L/h)
R — substrate removal rate (kg/m^3/d)
$R_{\max}$ — maximum substrate removal rate (kg/m^3/d)
S_e — effluent substrate concentration (mg/L)
S_i — influent substrate concentration (mg/L)
$S_{\ln}$ — logarithmic mean concentration of substrate in the influent and effluent (mg/L)
$U_{\max}$ — maximum utilization rate constant (kg/m^3/d)
V — reactor volume (L)
X — average biomass concentration in the reactor (g/L)
θ — hydraulic retention time, HRT (h)

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