Modelling of Nutrient Removal Processes in an Intermittently Aerated Bioreactor

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The general IAWPRC kinetics extended by volatilisation and the influence of wastewater composition on oxygen transport rate was used for description of lab-scale experiments simulating a Carrousel industrial reactor. Number of model equations and parameters were reduced by introduction of "switching functions", which express proportions of the reactor volume with different biochemical conditions. Majority of values of model parameters estimated for an industrial wastewater were close to those published in literature for municipal wastewater, except the hydrolysis rate constant of slowly biodegradable COD which was found significantly lower: 0.12 comparing to 3 g (g cell COD)⁻¹. The lower value was estimated due to the exploitation of a more precise model involving evaporation of volatile compounds. A strong inhibition effect of cadmium (II) salts was reflected by more than tenfold decrease in maximum growth rates of autotrophs and the rate constant of ammonia removal.

Key words:

Biokinetic parameter values, cadmium effect on biological processes, experimental and mathematical modelling, industrial wastewater treatment, intermittently aerated completely mixed activated sludge reactor, simultaneous nitrification and denitrification processes, volatilisation process

Introduction

A Carrousel type reactor is very frequently used to maintain zones with different concentration of dissolved oxygen, and in this way, various extents of reactions consuming either air oxygen, or oxygen from nitrates and nitrites, are established. Thus, simultaneous removal of COD, nitrates, ammonia, organic nitrogen compounds or eventually other types of substrates, can be achieved in the reactor. Because of high complexity of an exact model describing the Carrousel reactor, we have proposed an intermittently aerated, completely mixed, activated sludge reactor (CMASR), which uses different regimes in one reactor instead of considering a tankage of series of reactors, or a complicated continuous description by spatial differential equations¹.

The objective of the work was experimental and mathematical modelling of the processes mentioned above performed in an industrial Carrousel system. Another aim was to study the influence of a heavy metal on the processes carried out in the reactor and on biokinetic parameter values.

Experimental modelling

The volume of the reactor was 4.2 dm³. The cycle time 45 min. and the aeration time 15 min. were maintained when performing the pulse changes of the synthetic wastewater input composition. The reactor was operated at mean organic loading 0.19 g_{BOD5} g⁻¹_{MLVSS} d⁻¹ and solid retention time (SRT) was 20 d in order to maintain nitrifying activated sludge. The mean value of hydraulic retention time (HRT) was 63 h.

Measurements of the reactor response to pulse changes of input wastewater composition were carried out in order to verify the proposed mathematical models. Shock loading of the organic component and heavy metal (Cd²⁺) content in the synthetic wastewater, were performed. The concentration of COD in the reactor was about 900 mg \cdot l⁻¹, and the concentration of Cd²⁺ in the reactor reached approximately 10 mg \cdot l⁻¹ after performing this pulse input.

The measurements of the reactor responses to preliminary shock loading, which was carried out only with regard to organic component (cyclohexanone) content in the synthetic wastewater¹, were applied as reference measurements, i.e. without Cd^{2+} .

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The analyses of COD, MLSS, MLVSS, ammonium, nitrite, nitrate phosphorus and cadmium concentrations, were performed in accordance with standard methods². The dissolved oxygen concentration was measured by Syland Dissolved Oxygen Meter.

Mathematical modelling

Four mathematical models of simultaneous nitrification and denitrification processes in an intermittently aerated, completely mixed, activated sludge reactor, were developed and verified¹.

The IAWPRC activated sludge model^{3–5} was extended by including volatilisation process of volatile wastewater impurities. This extension was used for all verified models. Other extension of the above mentioned IAWPRC model is the implementation of influence of wastewater impurities on oxygen transport rate. Different approaches to oxygen implementation in biochemical reaction kinetics were applied in the development of the models. Consequently, the influence of wastewater impurities on oxygen transport implementation in individual models varies, too. Determination of some biokinetic parameter values based on evaluation of quasi steady-state measurements is also included in one of the models.

In Model 1, dissolved oxygen has been included as a substrate in the biochemical reaction rate terms. The process rate expressions are equal to those given in the Model No. 1. Matrix form of the process kinetics and stoichiometry⁵, including oxygen uptake rate, were applied to the formulation of the mass balance equations for the process components. Volatilisation of the substrate, as well as gas-liquid transport of oxygen, were also included into this model. Altogether, this model consists of 11 differential equations and 21 biokinetic parameters.

A mathematical description of volatilisation process in completely mixed, intermittently aerated bioreactor CMASR is based on mass balance of a volatile component of wastewater:

$$\frac{d\gamma_{V,2}}{dt} \cdot V = Q \cdot \gamma_{V,1} - Q \cdot \gamma_{V,2} - -(k_L a)_{agit} \cdot H \cdot \gamma_{V,2} \cdot V -$$
(1)
$$-(k_L a)_{aer} \cdot H \cdot \gamma_{V,2} \cdot p \cdot V - r_{V,S_v}$$

where

- Q wastewater volume flow rate, [L³ · T⁻¹]
- $\begin{array}{ll} \gamma_{V,1} & \mbox{ influent volatile organic component mass} \\ & \mbox{ concentration (COD), } [M \cdot L^{-3}] \end{array}$

- $\gamma_{V,2}$ effluent volatile organic component mass concentration (COD), $[M\cdot L^{-3}]$
- $(k_{\rm L}a)_{\rm agit}$ product of the partial mass transport coefficient and interfacial area per unit volume related to mechanical agitation, $[{\rm T}^{-1}]$
- $(k_{\rm L}a)_{\rm aer}$ product of the partial mass transport coefficient and interfacial area per unit volume; contribution due to aeration, $[{\rm T}^{-1}]$
- V reactor volume, [L³]
- $r_{V,SV}$ rate of volatile organic component consumption by biochemical reactions, $[M \cdot L^{-3} \cdot T^{-1}]$
- H Henry coefficient, [–]

The term $r_{V,Sy}$ in brackets is active only when biochemical reactions take place.

Parameter p relates the time of aeration (t_a) and the cycle time (t_c) , i.e.:

$$p = \frac{t_{\rm a}}{t_{\rm c}} \tag{2}$$

where

$$t_a$$
 – the time of aeration, [T]

 $t_{\rm c}$ – cycle time, [T]

The values of mass transfer coefficient for volatile component of wastewater due to mechanical agitation $[(k_La)_{agit} \cdot H]$ and due to aeration $[(k_La)_{aer} \cdot H]$ in Eq. (1), were obtained from the reactor response to pulse increase in concentration of volatile substance in the feed. The influence of wastewater composition on oxygen transport rate, more precisely on the value of the overall oxygen transport coefficient, was also considered in this model. Oxygen transport rate can be described as follows:

$$\frac{d\gamma_{Q}}{dt} = (k_{O_2}a)(\gamma_{O_2S} - \gamma_{O_2}) - (r_{V,O})$$
(3)

where

- $(k_{O_2}a)$ product of the overall mass transport coefficient and interfacial area for oxygen, $[T^{-1}]$
- $\gamma_{\rm O_2,S^-}$ saturation mass concentration of oxygen in solution, $[M\cdot L^{-3}]$
- γ_{O_2} mass concentration of oxygen in solution, $[M \cdot L^{-3}]$
- $r_{\rm V,O}$ rate of dissolved oxygen consumption by biochemical reactions, $[{\rm M}\cdot{\rm L}^{-3}\cdot{\rm T}^{-1}]$

The term $r_{V,O}$ in brackets is valid only when biochemical reactions take place.

For estimation of the parameter $(k_{O_2}a)$ value, Eq. (3) was integrated analytically. The term $r_{V,O}$ was not considered during this procedure.

The main goal of our alternative model of simultaneous nitrification and denitrification processes in the intermittently aerated CMASR (Model 2) has been to reduce the number of biokinetic parameters to be estimated. The values of some biokinetic parameters are obtained from the evaluation of COD, NH_4^+ , NO_3^- and MLSS concentration measurements, carried out at quasi steady-state conditions, before performing dynamic measurements. The formulas for calculation of these biokinetic parameters of the model were derived from the mass balance equations for selected components that were formulated for steady-state (quasi steady-state) conditions. The mass balances for the following process components were used in order to obtain these formulas:

i) mass balance of readily biodegradable substrate in order to calculate the value of hydrolysis rate coefficient $k_{\rm h}$

ii) mass balance of nitrate nitrogen in order to calculate the value of autotrophic yield Y_A

iii) mass balance of autotrophic biomass in order to calculate the value of autotrophic decay coefficient $b_{\rm A}$

iv) mass balance of heterotrophic biomass in order to calculate the value of heterotrophic decay coefficient $b_{\rm H}$

v) mass balance of particulate biodegradable organic nitrogen in order to calculate the value of nitrogen fraction in particulate product coefficient $i_{\rm XD}$

For example, the following expression can be derived for heterotrophic decay coefficient b_H evaluation, using quasi steady-state measurements:

$$b_{\rm H} = \mu_{\rm max, H} \cdot \frac{\gamma_{\rm S, ST}}{K_{\rm S} + \gamma_{\rm S, ST}} \cdot \frac{\gamma_{\rm O_2, ST}}{K_{\rm OH} + \gamma_{\rm O_2, ST}} +$$

$$\mu_{\max,H} \cdot \frac{\gamma_{S,ST}}{K_S + \gamma_{S,ST}} \cdot \frac{\gamma_{NO_3,ST}}{K_{NO_3} + \gamma_{NO_3,ST}} \qquad (4)$$

$$\frac{K_{\rm OH}}{K_{\rm OH} + \gamma_{\rm O_2,ST}} \eta_{\rm g} - \frac{Q_{\rm w}}{V}$$

where

- $b_{\rm H}$ decay coefficient of heterotrophic biomass, $[T^{-1}]$
- $\mu_{\max,H}$ maximum specific growth rate of heterotrophic biomass, $[T^{-1}]$
- $K_{\rm S}$ saturation equilibrium constant for heterotrophs, [M \cdot L⁻³]
- $\begin{array}{lll} \gamma_{S,ST} & \mbox{ state readily degradable organic} \\ & \mbox{ substrate concentration (COD), } [M \cdot L^{-3}] \end{array}$

$$K_{\rm NO_2}$$
 – nitrate saturation constant, [M · L⁻³]

 $\gamma_{NO_3,ST}$ – steady state nitrate nitrogen concentration, $[M\cdot L^{-3}]$

- $\gamma_{\rm O_2,ST}$ steady state dissolved oxygen concentration, $[M\,\cdot\,L^{-3}]$
- K_{OH} oxygen saturation equilibrium constant for heterotrophs, [M · L⁻³]
- $\eta_{\rm g}$ correction factor for anoxic growth of heterotrophs, [–]
- $Q_{\rm w}$ volumetric flow rate of wasted sludge, [L³ · T⁻¹]

The values of the above mentioned biokinetic parameters, obtained by evaluation of quasi steady--state measurements, were next used in evaluation of dynamic experiments. The process of organic impurity volatilisation and influence of wastewater components on oxygen transport rate, were also included in this model in the same manner as in the above-described Model 1. 11 partial differential equations and 16 biokinetic parameters are used in this model.

In our next approach, i.e. Model 3 (simplified model) instead of mass balance of oxygen, weight factors⁵ also known as *switching functions*, are applied to control procedures of biochemical reactions under aerobic and anoxic conditions. Multiplying reaction rate terms with switching functions s1, s2 and s3 defined for individual sets of reactions as follows, performs this control:

- aerobic growth of heterotrophs and hydrolysis of entrapped organic

$$s_{1} = \frac{1}{N_{\rm m}} \sum_{i=1}^{N_{\rm m}} \frac{\gamma_{\rm O_{2}}(t_{i})}{K_{\rm OH} + \gamma_{\rm O_{2}}(t_{i})}$$
(5)

- anoxic growth of heterotrophs and hydrolysis of entrapped organic

$$s_2 = \frac{1}{N_{\rm m}} \sum_{i=1}^{N_{\rm m}} \frac{K_{\rm OH}}{K_{\rm OH} + \gamma_{\rm O_2}(t_i)}$$
(6)

- aerobic growth of autotrophs

$$s_{3} = \frac{1}{N_{\rm m}} \sum_{i=1}^{N_{\rm m}} \frac{\gamma_{\rm O_{2}}(t_{i})}{K_{\rm OA} + \gamma_{\rm O_{2}}(t_{i})}$$
(7)

where

- $\gamma_{O_2}(t_i)$ dissolved oxygen mass concentration in reactor ,[M · L⁻³]
- K_{OA} oxygen saturation equilibrium constant for autotrophs, [M · L⁻³]
- t_i duration of measurements, [T]
- $N_{\rm m}$ number of measured points in oxygen profile

For highly recycled systems this approach actually represents approximation of aerobic and anoxic portion of the reactor volume. The advantage of this simplified model is that the mass balance equation for dissolved oxygen has not to be solved. Another advantage of this model is implicit description of influence of organic components of wastewater on oxygen transport through the values of the above given switching functions. In this model, the process of organic wastewater impurity volatilisation was also considered. Mean values of switching functions (Eqs. 5 to 7) were calculated by using all dissolved oxygen profile data that were measured in the reactor during performing dynamic measurements. The switching function values were kept constant and equal to the obtained mean values for the overall time for which the experimental data were treated. In quantitative figures, this approach has enabled to reduce the number of model equations to 10 and number of parameters to 19.

The only difference of Model 4 comparing to Model 3 is gradual evaluation of *switching function* values, i.e., separately for each cycle. This approach was assumed to be more realistic because of inclusion of changeable conditions influencing the dissolved oxygen profile in the reactor. The number of differential equations and of biokinetic parameters are equal to the previous model.

Solution and calculation methods

The explicit fourth-order Runge-Kutta-Merson method⁶ was employed to solve a set of differential equations. The steady state values, before shock loading, were taken as starting values for the solution. The values of model parameters were determined by the least square method using the modified relaxation⁷ and Nelder-Mead algorithm for finding a minimum of the objective function⁸. The sum of residuum squares, which includes all process components considered in the models, was used as an objective function. During the parameter estimation, either all (kinetic and stoichiometric) parameters of the model were considered, or some of them were fixed, to help in the convergence of the minimisation procedure. Different starting values of parameters were used to find a global minimum.

Results and discussion

The concentration of COD in the bioreactor increased to about 900 mg \cdot l⁻¹ and the concentration of Cd²⁺ reached about 10 mg \cdot l⁻¹ after performing the shock loading. The COD, NO₃⁻, NH₄⁺ and MLSS responses of the reactor to input shock perturbations, were measured and evaluated by using the above briefly described models. The concentra-

tions of nitrites in the output of the reactor have shown low and negligible during the measurements.

The values of objective function, i.e. sums of process component residuum squares obtained by evaluation of the reactor responses to shock loading, are given in Table 1. For comparison also the values of objective function obtained by treatment of the reference measurements with regards to heavy metal content¹, are presented in Table 2.

Table 1 - The values of objective function.

	Model 1	Model 2	Model 3	Model 4
without Cd ²⁺ (Reference)	327 453	343 684	299 889	324 658
with Cd^{2+}	315 612	528 975	196 447	190 733

As can be seen from Table 1, there are not significant differences between the values of objective function obtained by fitting different models to the experimental responses, particularly in the case of the reference system measurements evaluation. The differences can be seen between the values obtained by treatment cadmium-laden system. The lowest and very close values were obtained by using Model 3 (mean values of *switching functions*) and Model 4 (transient values of *switching functions*).

The experimental and calculated COD responses of CMASR to organic (cyclohexanone) and cadmium shock loading, are presented in Fig. 1. The experimental and predicted NO_3^- and NH_4^+ responses to this pulse input, are plotted in Fig. 2 and 3, respectively.

Applying Model 4 was obtained the best agreement between the experimental and calculated values of COD. The best fit of NO_3^- experimental responses was obtained by using Model 3. Fig. 3 shows evident differences between experimental and calculated NH_4^+ values, particularly, during the first more or less 6 hours of experimenting. In fact, ammonium represents the synthetic wastewater component with the worst description of experimental response. On the other hand, it should be mentioned that ammonium concentrations in the reactor were significantly lower (at least by one order) comparing to other wastewater components, i.e. nitrates and COD.

Likewise, nitrate responses (Fig. 2), insignificant differences between the calculated MLSS responses were noticed by using verified models. From Fig. 5 follow very close experimental and predicted oxygen profiles, obtained by applying both models, i.e. Model 1 and Model 2. However, a bit better fit of the experimental dissolved oxygen (DO) concentrations has been achieved by applying Model 1.

Dynamic changes in the switching functions during this experiment, are shown in Fig. 6. As mentioned above, a more precise description of dynamic behaviour of the reactor was expected after applying the "dynamic" values of switching functions, as evaluated for each cycle in Model 4, comparing to the mean values used in Model 3. Although, there were significant changes of *switching function* values during the experiments (Figure 6), no significant improvement of the transient behaviour description of the reactor resulted from this approach (see Table 2). In comparison with literature⁵, a significant difference has been found for values of the hydrolysis rate constants (3.0 and 0.12) of slowly biodegradable COD. Besides other effects this finding can be explained by the exploitation of more precise model involving evaporation of volatile COD. If such an evaporation is not taken into account it seems that COD is consumed by microorganisms and consequently estimated rate constant is higher.

Values of biokinetic parameters obtained by evaluation of experimental data by the above described optimisation procedure, are given In Table 2. Initial values of the parameters were the same for each evaluation. In Table 2 there are only those parameter values, which have been obtained by selected models, i.e. with the lowest values of the objective function. For comparison, the typical parameter values recommended by IAWPRC task group⁵ for simulation of domestic wastewater treatment



Fig. 1 – Experimental and predicted COD responses of intermittently aerated CMASR to input organic and cadmium shock loading

processes are also listed in Table 2. From this Table it can be seen, that the calculated values of maximum specific growth rate for heterotrophs $\mu_{max,H}$ are very similar in all the cases, and are lower in comparison with the recommended ones. The calculated values of half-saturation coefficient for heterotrophic biomass K_s are higher than the recommended value.

A similar relation between these two parameter values was obtained also by the evaluation of or-



Fig. 2 – Experimental and predicted NO₃⁻ responses of intermittently aerated CMASR to input organic and cadmium shock loading



Fig. 3 – Experimental and predicted NH⁺₄ responses of intermittently aerated CMASR to input organic and cadmium shock loading



Fig. 4 – Experimental and predicted NH⁺₄ responses of intermittently aerated CMASR to input organic shock loading (reference system)



Fig. 5 – Experimental and predicted dissolved oxygen responses of intermittently aerated CMASR to organic and cadmium input shock loading

ganic, ammonium and cadmium pulse shock load, and reference system¹. From the point of view of the monosubstrate Monod equation, these two parameters are strongly correlated and such behaviour could indicate an inhibition of heterotrophic microorganisms growth under experimental conditions. This assumption is not valid directly in the multisubstrate Monod kinetics, which is applied in the presented model concept, due to correlation between more than two biokinetic parameters.



Fig. 6 – Values of switching functions evaluated from oxygen profile measurements in intermittently aerated CMASR – organic and cadmium shock loading

An inhibition effect of cadmium implies from the course of time dependencies of COD in experiments without cadmium¹ and experiment carried out with cadmium (Fig. 1). It has been expected that this inhibition could be also reflected by estimated model parameters. This assumption is valid for the Model 1 for which the maximum values of the $\mu_{\text{max},\text{A}}$ were found to be 0.94 and 0.075 for the system without and with cadmium, respectively. The Model 3 did not give such a nice reflection of the investigated system. This can be ascribed to the worse suitability of this model (high value of the objective function and strong correlation between $\mu_{\rm max A}$ and K_S). The calculated values of the other parameters are very close to the recommended ones, at least when considering the basic model, i.e. Model 1, and evaluation of transient measurements performed without cadmium¹.

The estimated values of biokinetic parameters by using different mathematical models also differ. The most significant seem to be lower values for fraction of autotrophic microorganisms in biomass f_A in the case of cadmium shock load application in comparison with the values which have been obtained by evaluating the pulse change of the only organic component concentration of wastewater input into the reactor (reference system). The values of oxygen half-saturation coefficient for autotrophic biomass K_{OA} are higher in the case of evaluation of shock loading with cadmium present, when compared with the values valid for the reference system and the values recommended for simulation of municipal wastewater treatment processes⁵.

	1					
D	Unit	Model No. 1 Henze, 1987	without Cd ²⁺		with Cd ²⁺	
Par.			Model 1	Model 3	Model 1	Model 3
$\mu_{\rm max, A}$	d ⁻¹	0.80	0.94	0.90	0.075	0.71
$\mu_{\rm max,H}$	d^{-1}	6.00	1.10	1.13	1.51	1.46
$K_{\rm S}$	$g m^{-3} COD$	20.00	37.26	43.28	16.26	99.27
$K_{\rm NH}$	$g m^{-3} NH_4-N$	1.00	0.32	1.21	0.25	2.00
K _{NO}	g m ⁻³ NO ₃ -N	0.50	0.43	0.45	0.34	0.46
$Y_{\rm A}$	g cell COD formed (g N oxidised) ⁻¹	0.24	0.15	0.15	0.22	0.28
Y_{H}	g cell COD formed (g COD oxidised) $^{-1}$	0.67	0.45	0.45	0.62	0.50
$b_{\rm A}$	d^{-1}	0.12	0.16	0.17	0.19	0.17
$b_{\rm H}$	d^{-1}	0.62	0.20	0.20	0.47	0.27
$\eta_{ m g}$	dimensionless	0.80	0.74	0.75	0.69	0.62
$K_{\rm OH}$	g m ⁻³ O_2	0.20	0.26	0.26*	0.23	0.26*
K _{OA}	g m ⁻³ O_2	0.40	0.62	1.19*	1.73	1.50*
$k_{\rm h}$	g slowly biodegradable COD g^{-1} cell COD $\cdot \ d^{-1}$	3.00	0.12	0.12	0.12	0.11
$K_{\rm X}$	g slowly biodegradable COD g^{-1} cell COD $^{-1}$	0.03	0.025	0.023	0.040	0.048
$\eta_{ m h}$	dimensionless	0.40	0.60	0.60	0.55	0.59
k _a	$m^3 \cdot g^{-1} \cdot d^{-1} \text{ COD}$	0.08	0.15	0.16	0.038	0.065
$f_{\rm P}$	dimensionless	0.08	0.079	0.079	0.060	0.062
$i_{\rm XP}$	$g_{\rm N} \; g_{\rm COD}^{-1}$ in endogenous mass	0.06	0.064	0.061	0.060	0.061
$i_{\rm XB}$	$g_N g_{COD}^{-1}$ in biomass	0.086	0.050	0.051	0.080	0.040

Т	a b	l e	2	_	Biokinetic	parameter	values
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*calculated from mean values of switching functions by using Eqs. (5) to (7)

 $f_{\rm A}$ – fraction of autotrophs

dimensionless

dimensionless

 $f_{\rm B}$ – fraction of heterotrophs

Conclusions

 $f_{\rm A}$

 $f_{\rm H}$

Good agreement between the experimental and predicted dynamic values of COD, NO_3^- , MLSS and DO was obtained by applying the verified mathematical models of intermittently aerated CMASR. A time delay between experimental and predicted ammonium responses, was observed. A significant decrease of ammonium content in the reactor after this delay indicates temporal inhibition of nitrification process by autotrophs activity suppressing due to high organic loading.

0.012

0.583

0.005

0.55

0.009

0.60

0.014

0.585

The lowest and very close values of objective function (sum of process component residuum squares) were obtained by using Model 3 (mean values of *switching functions*) and Model 4 (transient values of *switching functions*). No significant improvement of transient approach (Model 4) with regard to the quality of dynamic behaviour description followed from the results. The combination of quasi steady-state measurements and dynamic responses allows decreasing number of biokinetic parameters, which have to be estimated by five.

In general, there are not significant differences between the estimated biokinetic parameter values and those recommended for simulation of municipal wastewater treatment processes. However, the indication of inhibition of some processes, e.g. autotrophic growth and heterotrophic growth in the presence of cadmium, follows from the comparison of the biokinetic parameter values of heavy metal-laden system in comparison to the reference system.

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